21:30-21:45	**Discussion and Summary**

Keynote Presentation Abstracts

K-1 Aerosols and Precipitation

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The effect of aerosols on precipitation is extremely important to the environment and a major scientific research subject. In this talk, we review key observational evidences for the effect of aerosols on precipitation. In addition, we analyze changes in precipitation and precipitation intensity in China (1961-2009), Taiwan (1961-2011), as well as in global data from the Global Precipitation Climatology Project (GPCP) covering the period 1979–2007. A large number of works have been carried out on the effect of aerosols on precipitation, but there still is no convincing scientific proof of the suppression of precipitation proposed as one of the indirect effects of aerosols. This is supported by results of our analysis which show little correlation between changes of annual total precipitation and emissions of air pollutants. On the other hand, the annual total precipitation and precipitation intensity are found to correlate significantly with global temperature rather than anthropogenic aerosols. In particular, the annual top 10% heavy precipitation increases by about 110% for each degree Kelvin (K) increase in global mean temperature, while the 30%–60% moderate precipitation decrease by about 30% /K. The global average precipitation intensity increases by about 23% /K, substantially greater than the increase of about 7% /K in atmospheric water-holding capacity estimated by the Clausius-Clapeyron equation. The large increase of precipitation intensity is qualitatively consistent with the hypothesis that the precipitation intensity should increase by more than 7% /K because of the additional latent heat released from the increased moisture. Nevertheless the bottom 10% light precipitation in industrialized regions has decreased significantly over the last 50 years, apparently as a result of suppression of precipitation proposed as one of the indirect effects of aerosols. In comparison, the bottom 10% light precipitation over the ocean where is relatively free from anthropogenic aerosols has increased slightly.

K-2 Ozone in the Anthropocene: Lessons from urban to remote measurements at northern mid-latitudes

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For more than six decades ozone concentrations have been quantitatively measured in the lower troposphere. Most of these measurements were made at northern mid-latitudes, the same region most impacted by anthropogenic emissions of ozone precursors. During these decades ozone precursor emissions increased dramatically, but also became the focus of increasingly stringent control efforts in many areas. The goal of this work is to determine as quantitatively as possible how ambient concentrations of ozone have responded to these changes in emissions (and perhaps changes in other atmospheric processes); both urban, regional and more remote (i.e. baseline) ozone concentrations will be considered. These determinations are designed to provide benchmarks to which chemistry-climate models can be compared, and address the following points.

Some major urban areas (e.g., Los Angeles and Mexico City) developed quite severe ozone pollution problems due to photochemical production on urban to regional scales. These problems have been well publicized and widely discussed, but perhaps less attention has been paid to the remarkable progress that has been made in alleviating these problems. Comparison of progress in different cities can provide useful guides for other urban areas, particularly rapidly developing mega-cities, both in controlling current problems and in avoiding some of the more severe problems experienced during earlier urban development.

On regional scales, transport between adjacent urban areas or air basins can be quite important. In urban areas with modest ozone pollution levels, transported baseline ozone can substantially contribute to local concentrations. Consequently, as local air quality standards are tightened, long-range and regional transport of ozone contributes an increasing fraction of allowable concentrations. Thus, to be optimally effective, air quality strategies must encompass local, regional and hemisphere-wide scales and consider varying baseline concentrations.

On the broadest spatial scale, available data (although limited) indicate that baseline ozone at northern mid-latitudes increased substantially since about the middle of the 20th century, and this increase continues today, at least in some regions. The longest and most reliable data sets (ten ground-based and one airborne) including six European, (beginning in the 1950s and before) three North American (beginning in 1984) and two Asian (beginning in 1991) are considered. When the entire time periods of the data records are considered a consistent picture emerges; ozone has increased at all sites in all seasons. At European and North American sites the average increase of ozone before 2000 was approximately 1%/year relative to the site's 2000 year mixing ratio in each season. For perspective, this rate of increase sustained from 1950 to 2000 corresponds to a doubling. At most European sites and some North American sites the rate of increase has slowed over the last decade (possibly longer) of the records.

The increase in baseline ozone has significant differences among the seasons, which implies that the ozone seasonal cycle has changed. At northern mid-latitudes the seasonal cycle has shifted so that the observed peak concentrations now appear earlier in the year than they did twenty years ago. Several possible reasons for this change will be discussed.

K-3 Global Remote Sensing of Tropospheric Trace Gases: GOME SCIAMACHY and GOME-2

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Since 1995 passive remote sensing measurements of the up welling electromagnetic radiation have been made using the following instruments: GOME on ERS-2 (1995-2011), SCIAMACHY on ENVISAT (2002 to 2012), and GOME-2 on Metop (2006/7 to present). The three platforms fly in sun synchronous low earth orbit having equator crossing times in da descending node of 10:30, 10:00 and 9:30 respectively for ERS-2, ENVISAT and Metop. In a nadir viewing geometry GOME and GOME-2 measure from 230nm to 793 nm whereas SCIAMACHY measures from 214 to 2380 nm: improving data rate, leads to the spatial resolution respectively of 320kmx40km, to 30kmx60km and 40kmx80km. Inversion of the measurements leads to the retrieval of the tropospheric columns of O_3, the O_3, precursors such as NO_2, HCHO and HCHO, the precursors NO_2 and SO_2, the halogen oxides BrO and IO and water vapour. The long term measurements of the gases will be presented and their changes or trends involving natural phenomena and anthropogenic activity will be discussed.

Invited Oral Presentation Abstracts

S1-I-1 Natural and anthropogenic changes in CO_2, CH_4 and N_2O over the past 2 millennia

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The changes in atmospheric trace gas composition during and before the anthropocene are best determined from the record of air enclosed in ice sheets. Air extracted from the open pores in firn and the bubbles in ice is measured to derive the past concentrations and isotopes of the long lived trace gases. The significant increases in CO_2, CH_4 and N_2O since about 1750 are caused mainly by energy, industrial and agricultural sources and are a key feature of the anthropocene.

The millennia preceding the anthropocene, the Late Pre-Industrial Holocene (LPIH), show evidence of natural changes in trace gases that can be used to constrain models and improve their ability to predict future changes under scenarios of anthropogenic emissions and climate change. The advantages of this period, as compared with periods further back in the past, are that many elements of the climate system, such as sea level, ice sheet extent and orbital arrangement were not greatly different from present, and information on other drivers of atmospheric composition such as climate variations and biomass burning is relatively abundant. However, precise measurements and accurately dated ice core air samples that are highly resolved in time (narrow air age spread) are required to record the small and rapid trace gas signals of this period.

The atmospheric composition records from Law Dome, Antarctica are widely used in chemistry, carbon and climate model simulations of the anthropocene and the LPIH. The air is enclosed with narrow age spread and recent samples overlap with the beginning of direct atmospheric observations. Comparisons between ice core measurements, firn air measurements and direct observations, as well as repeat measurements over many years, confirm that the air is enclosed and preserved reliably.

We have revised the records of atmospheric trace gases from Law Dome by applying updated calibrations and analytical corrections, consistent sample selection criteria, new ice and air dating and including new measurements. The revised records of CO_2, CH_4 and N_2O concentrations are almost identical to our previously published records, including significant variations in concentrations during the anthropocene and the LPIH. The likely causes of the variations will be discussed in light of evidence from isotopes and other trace gases that is emerging from the Law Dome cores and recently published results from other ice core studies.

S1-I-2 Multi-resolution Emission Inventory for China (MEIC): model framework and 1990-2010 anthropogenic emissions

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Developing an understanding of China's anthropogenic emissions has largely relied on bottom-up approaches that aggregate activity data and emission factors. Bottom-up inventories have been developed for China during past two decades and several regional and national emission inventory datasets were available. However, bottom-up inventory in China is still thought to be highly uncertain due to lack of detailed statistical data and local emission factor measurements, etc. This talk will cover several aspects of recent efforts in our group for continuously improving bottom-up emission inventory in China. We implemented an improved technology-based methodology to estimate anthropogenic emissions in China, in order to be able to reflect the types of technology presently operating in China. We also developed a regional emission processing system to convert annual emission inventory to model-ready emission data. With improved methodology, we have developed an open accessed emission database, which is named Multi-resolution Emission Inventory for China (MEIC). Anthropogenic emissions for the period of 1990-2010 will be presented in this talk.

S2-I-1 Impacts of Emissions from Megacities on Air Quality and Climate

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About half of the world's population now lives in urban areas because of the opportunities for better jobs, access to city services, cultural and educational activities, and a desire for more stimulating human interaction. At the same time, many of these urban centers are expanding rapidly, transforming themselves from villages to towns, towns to cities, and cities to megacities. The concentrations of people and activity are exerting increasing stress on the natural environment that it is beginning to have extensive urban, regional and global impacts. However, as the centers of economic growth, technological advances, social dynamics and cultural production, these urban centers also offer opportunities to manage a growing population in a sustainable way. Managing megacities and large urban complexes sustainably will be one of the major challenges in the coming years.

Population growth, increasing motorization and industrialization have resulted in a higher demand for energy, greater use of fossil fuels, and more emission of pollutants into the atmosphere. As a result, in recent decades air pollution has become not only one of the most important environmental problems of megacities, but also presents serious consequences to human health, ecosystems and climate change, as well as imposes large economic costs to society.

The talk will select a few megacities to identify similarities and differences among the air pollution problems and control strategies that are important to megacities throughout the world. While the progress to date in combating air pollution problems in developed and some developing world megacities has been impressive, many challenges remain including the need to improve air quality while simultaneously mitigating climate change. Recent scientific evidence demonstrates that control of pollutants that are short-lived in the atmosphere such as black carbon particles and methane (precursors of tropospheric ozone) through rapid implementation of proven emission reduction measures would have immediate near-term climate protection, in addition to significant air quality benefits. This talk will present some case studies of measures implemented in different world regions and discusses the opportunities and barriers to much wider implementation.

While each city – its problems, resources and outlook – is unique, the need for a holistic approach of the complex environmental problems is the same. There is no single strategy in solving air pollution problems in megacities; a mix of policy measures will be needed to improve air quality and mitigate climate change.

S2-I-2 Increase of total oxidant in Beijing atmosphere in summer, 1995-2010

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Changing of ground-level ozone, reflecting the oxidative capacity of the atmospgere, has been of increasing concern. The variation of ozone and total oxidant ($Ox=O_3+NO_2$) observed recently in the world were reviewed. And the high levels of Ox have been observed as a persistent feature of air pollutants in Beijing city, a typical Mega city in the world experiencing stringent air quality control in the processes of economic growth. To evaluate the effect of stringent air quality control in recent years, ground-base on-line measurements at one urban site were conducted in summer, and the variations of O_3 with simultaneous changes in NOx and the volatile organic compounds (VOCs) between 2005 and 2010 were analyzed. We combine the O_3 trend observed from commercial airlines over Beijing from 1995 to 2005 with ground-base measurements to explore a fifteen-year temporal trend of total oxidant in Beijing. Daytime high values of Ox concentrations in summer have been increasing significantly at an annual rate of about 2-4% yr-1. A simplified atmospheric model is used to evaluate effect of ozone precursor changes on the ozone production. It is shown that the greater increase in the emission of VOCs than that of NOx between 1995 and 2005 results in an increase of about 30% in the production of Ox. This agrees remarkably well with the 37% increase in O_3 observed in that period. Our recent measurements show that government's abatement efforts in controlling the emissions of ozone precursors have been effective. However the faster decrease of NOx than VOCs

reactivity has contributed to the continued increase of Ox. Our evaluation suggests that better coordinated reduction of NOx and VOCs emissions in the urban area is essential for ozone control in Beijing. The methods of evaluating the trends of total oxidants will also be useful for other cities for ground-level ozone control strategies.

S3-I-1 Anthropogenic land cover change effects on the short-lived climate forcers

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The short-lived climate forcers (SLCFs) ozone, methane and aerosol particulates and their indirect effects on cloud properties significantly impact regional and global climate but in complex ways involving both warming and cooling mechanisms. Selective reduction of the warming SLCFs is currently receiving attention as a way of mitigating near-term warming, reducing the rate of warming (important for adaptation of ecosystems) and simultaneously improving air quality. Feedbacks from anthropogenic land cover change have not been considered in previous assessments of historical and future SLCFs. Lack of quantitative information of these changing interactions, which perturb emissions of reactive carbon from vegetation, dry deposition rates of pollution to ecosystems and the underlying surface albedo, represents a major uncertainty in the ability to assess the climate and air quality benefits of reductions in the SLCFs. Here, a global Earth system model (NASA ModelE-Yale) with a new interactive vegetation biophysics module that incorporates photosynthesis-dependent isoprenoid emissions is applied to quantify the effects of historical (1850 – 2005) cropland expansion on the SLCFs. The resultant secondary organic aerosol direct radiative forcing entirely counteracts the biophysical (albedo) forcing from this human-induced land cover change (+0.16 versus -0.09 W/m^2). The land cover change alone implies a 15% longer methane lifetime in 1850 than 2005 while overall net effects on ozone radiative forcing are small. The analyses are extended to integrate the effects of future anthropogenic land cover change into projected aggressive air pollution emission abatement scenarios.

S3-I-2 Using Global Models to Unravel the Footprint of Human Activities on Clouds and Climate

NENES Athanasios, Karydis Vlassios, Capps Shannon, Henze Daven

Cloud droplets form upon pre-existing atmospheric aerosols, and their modulation has profound impacts on cloud radiative properties, the hydrological cycle and climate. Unraveling the complex relationship of cloud droplet number concentration (CDNC) to aerosol and aerosol precursor emissions is carried out with Global Climate Models (GCMs), often by activating/deactivating emissions. Although straightforward, this approach neglects the nonlinear dependence of aerosol concentrations on aerosol precursor emissions so that the resulting sensitivity reflects not only the actual CDNC response to an emissions change but also an altered size distribution and composition.

Adjoint modeling eliminates these issues by propagating an infinitesimal perturbation backwards through the model and, thus, elucidating the sensitivity of model output (e.g., CDNC) to the entire field of model parameters (e.g., emissions) without perturbing the model state. In this study, we reveal these relationships by developing the adjoint of the state-of-the-art aerosol-cloud interaction parameterization and coupling it with the adjoint of the GEOS-Chem chemical transport model. This model framework is then used to determine the impacts of emissions of black carbon and inorganic aerosol precursors on cloud droplet formation through adjoint gradients. Specifically, the adjoint model is applied to quantify the influences of global emissions on the predicted CDNC within the selected regions in the continental US and Europe. The resulting sensitivity maps display the range of spatial, sectoral and seasonal variability in the susceptibility of CDNC to emissions changes.

S4-I-1 Atmospheric Chemistry and Health

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Many studies have related adverse health effects to atmospheric particles and gases such as ozone and nitrogen dioxide. Air pollution mixtures are inherently complex, and we are only beginning to understand which mixtures or components are most harmful. Investigators have made many efforts expecially to establish which characteristics of airborne particles are most responsible for adverse health effects. These characteristics include size, surface area, chemical composition, presence of biological contaminants etc. It has, so far, been difficult to establish conclusively that some compoentns are more responsible for adverse health effects than others, or that other components are essentially harmless. The presentation will review recent studies in this area, and provide some perspectives for the way ahead.

S4-I-2 Spatial Analysis of Air Pollution and Mortality in California

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States Rationale

Chronic exposure to ambient air pollution may contribute to premature death, but the current evidence contains conflicting reports on the size and significance of this association. Some of the uncertainties may arise from inaccurate exposure assessment.

Objective

To assess the association of three criteria air pollutants (fine particulate matter, ozone and nitrogen dioxide) with death in a large cohort of California adults using individualized exposure assessments.

Methods

For fine particulate matter with aerodynamic diameter of 2.5 microns or less and nitrogen dioxide, we used generalized estimating equations and machine-learning model selection to derive predictive exposure models. For ozone we interpolated exposure with an inverse distance algorithm. Multilevel and standard Cox survival models were used to assess the association between death and air pollution exposure.

Measurements

Health data for 76,343 subjects were drawn from the American Cancer Society Cohort Cancer Prevention II survey with baseline ascertainment of individual characteristics in 1982 and follow up of vital status to 2000. Exposure data were drawn from hundreds of government monitoring sites throughout California.

Results

Fine particulate matter, ozone and nitrogen dioxide were all associated with death from cardiovascular causes and particularly ischemic heart disease in this cohort. Nitrogen dioxide and fine particulate matter were also associated with death from all causes. Only nitrogen dioxide was significantly associated with lung cancer deaths. In multipollutant models, nitrogen dioxide had the strongest and most consistent associations with death.

Conclusions

The results from this investigation indicate consistent and robust effects of fine particulate matter, ozone and nitrogen dioxide on premature death in California.

S5-I-1 Biofuel production, isoprene emissions and ground-level ozone

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Although our understanding of the fundamental processes controlling isoprene emissions from plants and their impacts in the atmosphere is still not complete, it is believed that in NOx-rich regions of the world isoprene emissions enhance the formation of surface ozone, a priority pollutant which causes oxidative damage to plants, reducing crop yields, and respiratory damage to humans and other animals. Many biofuel crops emit significantly different reactive volatile organic compounds to the atmosphere than traditional agricultural food crops, including, in the case of poplar, willow, eucalyptus, reed grass and oil palm, much higher emission rates of isoprene. Hence, large-scale land use change to deliver increased biofuel crop production may have impacts on ground-level ozone.

As an example of the continuing progress towards better quantification of the role of isoprene in the atmosphere, we show that accounting for the circadian control of isoprene emissions that we observed in the 2008 OP3 field campaigns in Borneo may improve estimates of landscape-scale isoprene emissions and hence improve model replication of ground-level ozone observations. Using data collected during the OP3 campaigns and a box model of atmospheric chemistry, we also show that increasing oil palm production in this region of the tropics may very substantially increase ground-level ozone concentrations in the future,

We then go on to show how the current world-wide expansion of bioenergy feedstock crop production, driven by concerns about climate change and energy security, may give rise to substantial air quality problems. We show that growing sufficient short rotation coppice (SRC) in Europe to replace 1% of the projected global fossil fuel demand in 2020, in place of agricultural crops or pastures, results in a global increase of isoprene emissions of around 1%. Using a global chemistry-transport model and a box model of atmospheric chemistry we estimate that this leads to increases of ~3 ppbv in the July average ground level ozone concentration over Europe. The maximum ozone increases coincide with the period of peak photochemical activity and main growing season, maximising both health and crop impacts. Using dose-response functions derived from the literature on epidemiological studies and field trials, we show that such cultivation of SRC for biofuel production could be responsible for 1,300 premature deaths and a 4% reduction in wheat yield each year in Europe as a result of the increase in ozone concentration.

Our studies show the need to consider more than simple carbon budgets when considering the cultivation of biofuel feedstock crops for greenhouse gas mitigation.

S5-I-2 BVOC exchange at the atmosphere-forest interface: Adding canopy complexity from the local to regional scale

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The balance between biogenic volatile organic compounds (BVOC), anthropogenic nitrogen oxides (NOx), and hydrogen oxide radicals (HOx) is key to determining photochemical regimes and aerosol formation. Field campaigns in forested regions have improved our understanding of the role of BVOC emissions on gas-phase tropospheric chemistry, and here we evaluate the role of canopy complexity to in-and above-canopy dynamics and atmospheric chemistry at two isoprene-emitting deciduous broadleaf forests in the continental United States (the University of Michigan Biological Station and Harvard Forest). We specifically focus on the role of canopy structure on the forest exchange dynamics and in-canopy and above-canopy chemistry of BVOC and its oxidation products.

This includes the contributions of understory species and light shading within the canopy to determine how canopy structure affects emissions and oxidation. Local, one-dimensional modeling experiments investigate the role of forest-atmosphere mixing versus atmospheric chemistry, and synthesize key processes needed for scaling forest-canopy exchange to regional scale photochemical models.

S6-I-1 On the physical state of aerosol particles: principal processes and atmospheric implications

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Knowledge of the physical state of aerosol particles is of fundamental importance for understanding many atmospheric processes including heterogeneous chemistry and cloud formation. In this overview presentation, different types of physical states will be discussed including normal liquids, ultraviscous liquids, amorphous semi-solids and solids, as well as crystals. Moreover, both thermodynamic as well as kinetic aspects of phase transitions between these states are examined and several examples for such processes will be presented. These include gas-particle interactions such as the uptake and release of water by aerosol particles, homogeneous and heterogeneous nucleation processes of ice and other crystalline solids, and the effects of physical state on the kinetics of heterogeneous chemical reactions. For particular emphasis, the formation and properties of amorphous semi-solid and glassy states in organic condensed matter are reviewed as well as their dependence upon changes in temperature or humidity. Implications of the glassy state of aerosol particles for atmospheric multi-phase processes will be discussed, for example regarding heterogeneous chemistry, aerosol lifetime, and cloud formation. In addition, established and new experimental techniques and modeling tools for studying such processes will be introduced. The talk concludes with an assessment of the current state of knowledge, open questions, and proposals for future studies to resolve existing uncertainties.

S6-I-2 Field measurements of tropospheric OH and HO_2 radicals: Comparisons with model calculations, novel chemical mechanisms, and technique development

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The hydroxyl radical, OH, initiates the removal of the majority of trace gases in the atmosphere, and together with the closely coupled species, the hydroperoxy radical, HO_2, is intimately involved in the oxidative chemistry of the atmosphere. Trace species removed by OH include greenhouse gases and substances harmful to health, and OH initiates the formation of a wide range of secondary products, many of which are implicated in poor air quality, for example ozone and aerosols. In this presentation I will present recent field measurements of local concentrations of OH and HO_2 radicals in the troposphere, as well as OH reactivity (the total rate of OH removal), and comparisons that have been made with numerical model calculations containing a detailed chemical mechanism, for example the Master Chemical Mechanism (6700 species, 17000 reactions). Owing to their short lifetime, the abundances of OH and HO_2 are determined solely by their rate of chemical production and loss, and not by transport. The level of agreement between field measurements of OH and HO_2 concentrations and model calculations provides an indication of how well we understand the underlying oxidation chemistry. Measurement-model comparisons for a range of different environments sampled from the ground and from aircraft will be presented, including the marine boundary layer, low-NOx regions influenced by biogenic emissions, for example forests, the polluted urban boundary layer (including aircraft measurements at night), and polar regions. Significant discrepancies remain unexplained, for example in forested regions where measured OH concentrations are up to an order of magnitude higher than model calculations. Combining field measurements of OH and OH reactivity suggest new OH sources are required during the oxidative degradation of biogenic emissions, for example of isoprene. Understanding these new sources is a highly active area of research. In the marine boundary layer and polar regions the concentrations of OH and HO_2 are significantly influenced by iodine and bromine species, and emissions from the snowpack enhance HOx levels in polar regions. The measurement of OH and HO_2 radicals in the troposphere remains extremely challenging, with only two methods used to measure their concentrations in the field: laser-induced fluorescence spectroscopy at low pressure (FAGE) and chemical ionisation mass spectrometry (CIMS). I will discuss potential interferences for OH and HO_2 radicals measured using the FAGE technique, which have recently been reported for some instruments.

Oral Presentation Abstracts

S1.1 Changing ozone at Europe's borders: Quantifying current and future impacts

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Despite relatively large reductions in emissions of ozone precursors over the past two decades, observation sites over Europe show little evidence for decreases in the number of days ozone exceeds the 120 μg/m^3 air quality threshold or in annual averaged surface ozone, which continues to rise at many locations. While a number of factors may contribute to this, including changes in meteorology, natural emissions and emissions distribution, there is also an increasing inflow of ozone from distant upwind sources and from long-lived precursors such as methane. This study reviews changes in long-term measurements of ozone at sites on the edge of the European continent, such as Mace Head, Ireland, which have in the past been used to provide boundary conditions for regional scale modelling informing development of policy objectives. We explore the extent to which these sites are representative of changes in ozone inflow to Europe. We use information on the ozone response to past emission changes from 14 independent chemistry-transport models participating in the HTAP studies to explore the extent to which models reproduce the observed trends and to estimate and attribute changes in ozone and its precursors at the boundaries of Europe. We then examine the likely changes by 2030 under a number of different emission scenarios,and explore the impacts of these changes on European ozone in light of the expected regional emission changes using the EMEP regional model. These ozone changes are contrasted with the changes expected due to changing climate over the same period. We conclude by highlighting the role that ozone from upwind sources and from methane play in contributing to surface ozone changes over Europe, demonstrating the value of analysis of ozone changes by season, latitude and altitude, and emphasizing the need for measurement stations at continental boundaries that are capable of gauging continental inflow and outflow.

S1.2 Historic atmospheric NMHC and alkyl nitrate trends from firn profiles at North GRIP and NEEM and the changing production efficiency of alkyl nitrates

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We present atmospheric histories for the past 50 years of two alkanes, butane and pentane, and four alkyl nitrates, 2-butyl, 2-pentyl, 3-pentyl and 2-methyl-3-butyl nitrate. These were derived based on analysis of firn air from the North GRIP and NEEM sites, Greenland (drilled in 2001 and 2008 respectively) using a direct firn model. The histories show that atmospheric mixing ratios of the alkanes peaked in the mid-1970s, whereas those of the alkyl nitrates peaked in the early 1990s.

Alkyl nitrates are formed in the atmosphere by reaction of an alkyl-peroxy radical with NO. We report the first simultaneous measurements of alkyl nitrates and their parent alkanes. Using the changing ratios of the alkyl nitrates to their parent alkanes we have investigated the changing efficiency of this alkyl nitrate production pathway. The results show a threefold increase in this ratio between the mid-1970s and the early-1990s, with results from the North GRIP and NEEM sites agreeing very well. The data from the NEEM site then shows the ratio decreasing again up to 2008 (the date the air was recovered) to almost pre 1975 levels. The explanation for this trend seems to be a changing production efficiency which appears to be related to changing NOx mixing ratios rather than changing hydrocarbon mixing ratios.

We conclude that Arctic firn air represents a consistent history of changing hydrocarbon and NOx emissions in the northern hemisphere.

S1.3 Long-term monitoring of volatile organic compounds at the background and suburban sites in the Czech Republic

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The purpose of Volatile organic compounds (VOCs) monitoring is primarily to identify and quantify pollution sources. As a follow-up, oxidation models, together with

meteorological parameters and information concerning the emission of individual substances, make possible to understand the processes in the atmosphere and to estimate the long-range transport of air pollution. In 1992, the Co-operative programme for monitoring and evaluation of the long-range transport of air pollutants in Europe (EMEP) started VOC monitoring at selected stations of the EMEP network including the Observatory Košetice (geographical co-ordinates: 49°35′ N; 15°05′E; 534 m a.s.l), operated by the Czech Hydrometeorological Institute (CHMI). In 1995, CHMI started the same type of measurement at the Praha-Libuš station (50°00′ N; 14°27′ E; 301 m a.s.l), which represents the suburban area of the capital Prague. The aim of presented study is to detect the long-term trends of VOCs in the Czech Republic and to compare the VOCs pollution levels at the background scale and at the suburban site of big city. The VOCs emissions in EU dropped in the period 1990-2008 by almost 50%. The descrease in the Czech Republic was even more rapid, the emissions 1990 decreased by more than 63% in the same period. Air samples are taken and put into steel cans for the later assay of VOC concentrations. The sampling has been implementing at 12 o′clock UTC every Monday and Thursday for the duration of 10 minutes. In the laboratory, a group of hydrocarbons is quantified by gas chromatography in a capillary column with a flame-ionisation detector. Following VOCs have been measured: ethane, ethene, propane, propene, i-butane, acetylene, n-butane, total butanes, i-pentane, n-pentane, total pentenes, i-hexane, n-hexane, isoprene, i-heptane, n-heptane, benzene, toluene, etylbenzene, m,p-xylene, o-xylene. Most of non-methane hydrocarbons follow an annual course that reflects their emission levels, i.e. with maximums in winter and minimums in summer. Isoprene is an exception. Despite it’s ranking among the most reactive VOCs, it is of natural origin (deciduous trees release it) and displays an inverse annual course. In general, the reduction of VOCs emissions in last 2 decades was reflected in decrease of concentrations at both regional and suburban level of the Czech Republic. The nonparametric Mann-Kendall method was used for statistical evaluation of VOCs trends significance (4 levels of trend significance). The most significant downward trend was found for almost all of measured VOCs at both stations. The ethane trend was more significant at suburban site than at the background level. The trend of isoprene concentrations is controlled first of all by natural conditions and shows different patterns as other VOCs. No significant trend was found under the period under review at Košetice observatory and slightly increasing trend was detected at Praha-Libuš. The concentrations of main VOCs at the suburb level were higher by 150-200% than at the background site in the nineties. In the last decade, the difference between suburban and background level was smaller, thanks to the considerable reduction of the VOCs anthropogenic emissions.

S1.4 A new perspective for the self-cleansing capability of the atmosphere

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The self-cleansing capability of the atmosphere is predominantly based on the reaction cycles of hydroxyl-radicals, OH, and on the catalysis of these reactions by nitrogen oxide, NO. Published observations in forested regions with high hydrocarbon and low nitrogen-oxide concentrations show unexplained enhancements of OH up to a factor of 10 when compared to the current theory of atmospheric chemistry. Here we develop a new photochemical coordinate system which uncovers a common feature in all of these data sets. In contradiction to current theory the observations show that the self-cleansing capability of the atmosphere is stabilized at its maximum efficiency in regions with a high natural burden of VOCs even without catalysis through nitrogen oxide.

S1.5 Observation of abnormal daytime N_2O_5 and NO_3 at a coastal urban site in southern China

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Dinitrogen pentoxide (N_2O_5) and nitrate radical (NO_3) are known as key reactive nitrogen oxides in the nocturnal troposphere. The daytime chemistry of N_2O_5 and NO_3, however, is far from being fully understood for various atmospheric environments. Here we present the recent observation of N_2O_5 and NO_3 using a chemical ionization mass spectrometer (CIMS) in urban Hong Kong during October–December 2010, a season with high photochemical activity and rich nitrogen oxides. Abnormal elevated concentrations of daytime $N_2O_5+NO_3$ were observed with the maximum hourly value of 1033 pptv. During about 1/3 of the measurement period (total 50 days), $N_2O_5 + NO_3$ exhibited a daytime peak in the afternoon with hourly mixing ratios over 200 pptv. Rising ozone and NO_2 in combination with reduced NO in the afternoon introduced the special atmospheric condition to form remarkable daytime N_2O_5 and NO_3. The measured daytime N_2O_5 and NO_3 were found to be highly dependent on their precursors – ozone and NO2, and would build up to shape a daytime peak with ascending ozone or total ozone ($Ox = O_3 + NO_2$). NO titration, the dominant removal process at this urban site, was observed tardy to deplete the daytime NO_3 and N_2O_5 if excess ozone was present. Photodecomposition, a key loss process in the presence of sunlight, however, was less important in urban Hong Kong compared to NO depletion, and thus exhibited rare suppression to the daytime NO_3 and N_2O_5. Steady-state prediction also showed daytime peaks of $N_2O_5+NO_3$ in the afternoon. However, the observed abundances were usually much higher than those predicted in cases of moderate or high NO, which suggests there were possibly additional physical or chemical source processes in certain atmospheric conditions. Vertical transport due to the concentration gradient of N_2O_5 in range of dozens of meters as well as intensive turbulence probably served as a significant physical source of NO_3 and N_2O_5 at this site especially during daytime. Important impacts of the elevated daytime N_2O_5 and NO_3 on atmospheric chemistry are discussed for a polluted scenario, including enhanced Ox destruction, accelerative conversion of NOx to nitric acid, increased oxidation of VOCs and chlorine activation.

S1.6 Understanding our current knowledge on organic aerosol modeling and comparison with measurements: an AEROCOM multi-model approach

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Comparisons of individual models with organic aerosol (OA) measurements have shown a large underestimation of the OA component by the models, especially during winter. The formulations used by individual models are very different, since OA simulations have many degrees of freedom due to the missing knowledge on the behavior and fate of both primary OA (POA) and secondary OA (SOA) in the troposphere. On top of that, several assumptions need to be made and are translated to model tuning parameters that vary greatly from one model to the other. Trying to bridge the gap between models and observations, several recent model developments account for SOA, intermediate volatility organics, multiphase chemistry, oceanic fluxes and semi-volatile POA. In addition to the different processes included in various models, the different emission inventories and meteorological fields adopted make a comparison with other models and measurements even more challenging.

The organic aerosol intercomparison AEROCOM exercise aims to evaluate the actual status of global modeling of the OA occurrence in the global troposphere and analyze discrepancies between models as well as between models and observations. More than 20 models participate in this exercise that aims to quantify the uncertainties and attribute them to major contributors. It will also try to identify and analyze potential model systematic biases. The ensemble of the simulations will be used to build an integrated and robust view of organic aerosol sources and sinks in the troposphere.

The year 2006 was selected as the base year for the study. Recent results from the intercomparison will be presented together with a compilation of field data that is being used for the validation of the models results. The similarities and differences between models will be highlighted, in an attempt to evaluate and understand the model-measurements comparison.

S1.7 Secondary Organic Formation in the Megacity of Beijing

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Fine particle ($PM_{2.5}$) has been focus for air pollution control. In order to investigate the potential sources of $PM_{2.5}$, Multiple state-of-the-art instruments were used to measure aerosol physical and chemical components at the urban atmospheric environmental monitoring station at the campus of Peking University, Beijing in the past few years. $PM_{2.5}$ and its chemical components vary seasonally due to meteorological conditions and source emissions. Combination of several methods to distinguish the primary and secondary formation was conducted, that is, EC tracer method to estimate the primary and secondary organic aerosol, PMF-AMS method to identify the evolution of secondary organic aerosol, filter based organic speciation and CMB to quantify the major approach of secondary organic formation. The results showed the primary emission and secondary formation is comparable in Beijing. SOC/OC ranged 1/5 in winter and nearly half in summer.

A high-resolution aerosol mass spectrometer (HR-AMS) was applied in the two campaigns conducted in Dec., 2010 (winter) and Aug., 2011 (summer), respectively. Positive Matrix Factorization (PMF) analysis of the high resolution organic mass spectral resolved four organic aerosol components in summer and six components in winter. The low-volatile oxygenated aerosol (LV-OOA) and semi-volatile oxygenated aerosol (SV-OOA) components correspond to fresh formed and aged secondary organic aerosol respectively. The total organic mass in Beijing summer dominated by LV-OOA (37.3%) and SV-OOA (28.4%), which was much higher than that of in winter (18.3% and 12.6%). Cooking emissions were found to be an important organic source in Beijing, as PMF resolved the cooking-related organic aerosol (COA) accounted for 21.3% (summer) and 19.2% (winter) of total organic mass. The COA mass spectra and diurnal patterns in summer and winter both showed concentration peaks in noon (12:00-13:00) and early evening (19:00-20:00). Hydrocarbon-like (HOA) was identified to be closely associated with primary combustion tracers (e.g. BC, CO). The HOA accounted for 13% of organics in summer. More specific primary sources (coal-combustion OA, vehicle OA and Biomass burning OA) were indentified clearly in the winter campaign with strong primary emissions, accounting for 24%, 14%, and 12% of total organics, respectively.

Totally 14 kinds of secondary organic tracers derived from isoprene, -pinene, -caryophyllene and toluene were quantified using gas chromatography-mass spectrometry. Tracer-yield method was used to estimate biogenic and anthropogenic SOA/SOC in the atmosphere at PKU. Toluene SOA was the major secondary contributor at PKU, followed by isoprene SOA. Compared with other studies, PKU had the highest anthropogenic SOA, and biogenic SOA concentration was between biogenic and anthropogenic SOA of other studies, indicating the high background in Beijing. The high concentration of anthropogenic SOA suggested the regional effects of anthropogenic pollution.

S1.8 Ambient Organic Aerosol Aging: Application of the 2-D Volatility Basis Set to Field Studies

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Multigenerational oxidation chemistry of atmospheric organic compounds and its effects on aerosol loadings and chemical composition is investigated by implementing the Two-Dimensional Volatility Basis Set (2D-VBS) in a Lagrangian host chemical transport model. Three model formulations were chosen to explore the complex interactions between functionalization and fragmentation processes during gas-phase oxidation of organic compounds by the hydroxyl radical. The base case model employs a conservative transformation by assuming a reduction of one order of magnitude in effective saturation concentration and an increase of oxygen content by one or two oxygen atoms per oxidation generation. A second scheme simulates functionalization in more detail using group contribution theory to estimate the effects of oxygen addition to the carbon backbone on the compound volatility. Finally, a fragmentation scheme is added to the detailed functionalization scheme to create a functionalization-fragmentation parameterization. Two condensed-phase chemistry pathways are also implemented as additional sensitivity tests to simulate 1) heterogeneous oxidation via OH uptake to the particle-phase and 2) aqueous-phase chemistry of glyoxal and methylglyoxal. The model is applied to summer and winter periods at three sites where observations of organic aerosol (OA) mass and O:C were obtained during the European Integrated Project on Aerosol Cloud Climate and Air Quality Interactions (EUCAARI) campaigns. The base case model reproduces observed mass concentrations and O:C well, with fractional errors (FE) lower than 55% and 25%, respectively. The detailed functionalization scheme tends to overpredict OA concentrations, especially in the summertime, and also underpredicts O:C by approximately a factor of 2. The detailed functionalization model with fragmentation agrees well with the observations for OA concentration, but still underpredicts O:C. Both heterogeneous oxidation and aqueous-phase processing have small effects on OA levels but heterogeneous oxidation, as implemented here, does enhance O:C by about 0.1. The different schemes result in very different fractional attribution for OA between anthropogenic and biogenic sources.

S1.9 Particle-resolved simulations to determine size-dependent aging time-scales of black carbon aerosol

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The composition of aerosol particles containing black carbon changes continuously after emission during their transport in the atmosphere. Coagulation, condensation, and photochemistry are contributing processes, collectively known as aging. This changes the particles' physico-chemical properties, in particular their hygroscopicity, from initially hydrophobic to hydrophilic, and hence their ability to act as cloud condensation nuclei. Parameterizations of black carbon aging are widely used in global aerosol models and are often based on a first-order model with the single parameter of aging rate or its inverse, the aging time-scale, which is applied to the whole population of hydrophobic black carbon particles. Sensitivity studies have shown that the black carbon burden in global models depends strongly on the assumed aging time-scale, hence constraining this value is important for predicting the black carbon climate impacts.In this study we present a method for explicitly calculating size-dependent aging time-scales of black carbon aerosol based on simulations with the particle-resolved aerosol model PartMC-MOSAIC. As criterion for aging we used the activation of the particles at a chosen supersaturation threshold, and we included coagulation and condensation as aging processes. The resulting aging time-scale is a function of the wet diameter of black-carbon-containing particles D_(wet,BC), the supersaturation threshold S, and the time of day t. We determined size-dependent aging time-scales for combustion-generated particles for about 300 scenarios, exploring a range of environmental parameters and pollution characteristics. The value of the time-scale varied from minutes to weeks, depending on the size of the fresh black-carbon-containing particles and the local plume dynamics. The tendency for a particle to age by condensation versus coagulation depended on its wet diameter prior to aging. The condensation aging time-scale was shortest (~10 minutes at S=0.3%) for the largest fresh particles (D_(wet,BC)>100 nm) and decreased with the secondary aerosol mass production rate per wet aerosol surface area (from ~1 week to ~1 hour for D_(wet,BC)≈100 nm at S=0.3%). The coagulation aging time-scale was shortest (~1 hour at S=0.3%) for the smallest (D_(wet,BC)<20 nm) fresh particles and decreased with the total number concentration of large (D_(wet)>200 nm) particles. Therefore, both condensation and coagulation play important roles in aging, and their relative impact depends on the particle size range.

S1.10 Nighttime chemistry: modelling and observations from RONOCO

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During the day, the hydroxyl radical (OH) is abundant and the nitrate radical (NO_3) is rapidly photolysed, so NO_3 is not an important daytime oxidant. At night, however, OH concentrations are low, and so NO_3 takes its place as the primary oxidant driving nighttime chemistry. The aims of the RONOCO project (ROle of Nighttime chemistry in controlling the Oxidising Capacity of the atmosphere) are to quantify the processes influencing nighttime chemistry, using UK-based aircraft observations in combination with atmospheric chemistry models covering different scales (including a box model, a high resolution regional model and a global model).

Here, observations and results from the global chemistry transport model pTOMCAT will be presented. Observations including O_3, CO, NOx, NO_3, N_2O_5, HNO_3, PAN, aerosol and VOCs were taken on board the FAAM (Facility for Airborne Atmospheric Measurement) BAe146 research aircraft during July-August 2010 and January 2011, around the UK.

The model is run with present day aerosol and future aerosol scenarios. Changing the aerosol directly affects nighttime chemistry, as it changes the amount of N_2O_5 lost through hydrolysis on aerosol surfaces. In this model, this is the only direct effect of changing the aerosol. However, changing the N_2O_5 loss rate causes changes to other species such as NOx, O_3, and OH. The impact of changing the aerosol on composition and oxidising capacity is evaluated at the regional and global scales. The impact of aerosol composition on the parameterisation of N_2O_5 hydrolysis is also investigated.

S1.11 Trends in atmospheric mercury and implications for past and future mercury accumulation in surface reservoirs

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Global mercury deposition has tripled since pre-industrial times, and global emission inventories indicate a persistent rising trend. Yet atmospheric observations show a decrease over the past 15 years. We find that this atmospheric trend mostly reflects observations in the North Atlantic region and show that it can be explained by a decline in mercury from the subsurface Atlantic, which was greatly enriched in mercury during past decades. We propose several explanations for this historical enrichment and recent decline of mercury in the North Atlantic including environmental release of mercury from commercial products, wastewater discharges, change in the Hg(II)/Hg(0) emission ratio, and increasing boundary layer ozone (decreasing Br atom concentrations and hence Hg(0) oxidation). We discuss constraints on atmospheric oxidation of Hg(0) from CARIBIC aircraft observations in the lowermost stratosphere. Atmospheric mercury observations in the Arctic show a summer peak, in contrast to the summer minimum at northern mid-latitudes, and we suggest that this reflects seasonal evasion from the shallow Arctic Ocean enriched by riverine inputs. Climate change may thus cause an increase of mercury in the Arctic due to increasing river discharge combined with mobilization of mercury from boreal soils by permafrost thaw and forest fires. Although current mercury emissions to the atmosphere are dominated by Asia, the historical loading of mercury in surface reservoirs such as the North Atlantic mostly reflects the legacy of past emissions in North America and Europe. The time scales for dissipation of this legacy mercury (by transfer to stable reservoirs) are of the order of decades. We conclude that limited policy action to hold mercury emissions at present-day levels would result in a

continued global increase of mercury in surface reservoirs as the system is presently out of steady state. However, vigorous curtailing of mercury emissions would not only provide an immediate gain by decreasing atmospheric deposition but also a longer-term gain by decreasing the surface reservoirs of mercury.

S1.12 Atmospheric ammonia: global distributions and trends from the IASI satellite mission

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Human activities have significantly perturbed the natural nitrogen cycle during the last decades, and this had major environmental consequences. The anthropogenic perturbation of the nitrogen cycle originates from our production of energy and food, which causes atmospheric emission of reactive nitrogen compounds, principally nitrogen dioxide (NO_2) and ammonia (NH_3). NH_3 is the principal alkaline component in the atmosphere. It rapidly forms aerosols and is therefore a species of high relevance for air quality and climate. Global NH_3 emissions have doubled in the last century and are expected to further increase in the coming decades, with huge implications on the Earth's environment and human health.

As a short-lived species, NH_3 is highly variable in time and space. Its global atmospheric budget is currently poorly understood. Satellite instruments, with their high temporal sampling and global coverage, provide new means to monitor NH_3 distributions and trends. Especially the Infrared Atmospheric Sounding Interferometer (IASI), on board of the MetOp platform, has demonstrated its capabilities for probing

NH3 daily and globally. An Optimal Estimation Method on Spectral Ensemble (OEMSE) has been developed recently to obtain global distributions of NH_3 total columns from IASI with improved sensitivity in comparison to previous studies; it also allows assessing more critically the retrievals errors. The method is presented here.

In this work we also provide a first validation of the new product using ground-based in situ and aircraft measurements as well as model data. We examine the capabilities of IASI to monitor local sources and transport of NH_3, and to contribute, by using regional model simulations from LOTOS-EUROS, in refining emission inventories above Europe. Global distributions, seasonal and inter-annual variations of NH_3 totals columns are presented for the period from 2008 to 2012. Finally, we also present preliminary results from an empirical analysis of NH_3 volatilization, performed by correlating the total columns retrieved globally to soil parameters (such as temperature, pH and relative humidity) and anthropogenic activity indicators (e.g. livestock density, intensity of fertilization and population density).

S1.13 Anthropogenic atmospheric source region plume characterisation of the interior of South Africa

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Although southern Africa is an important source region from a global atmospheric chemistry perspective, it is currently not well characterised. Atmospheric scientific studies in southern Africa have mainly been limited to short-term intensive campaigns e.g. SAFARI-92 and SAFARI 2000, or long-term campaigns with less comprehensive measurements. The Cape Point GAW station is the most comprehensive long-term station in this region. It is, however, not representative of continental southern Africa since it is far removed from the industrial hub of South Africa and is not influenced by meteorological patterns dominating the interior. To at least partially address the knowledge gap, i.e. need for longer term comprehensive data sets that are regionally representative of continental southern Africa, the University of Helsinki (Finland), the North-West University (South Africa) and the Finnish Meteorological Institute initiated various multiple year measurements campaigns and also led the South African part of the EUCAARI project measurement campaign. In mid 2010, measurement equipment utilised in the afore-mentioned campaigns, as well as additional equipment were unified into a permanent measurement station at Welgegund (www.welgegund.org, 26°34'10"S, 26°56'21"E), in the North-West Province. This led to the establishment of most likely the best equipped permanent atmospheric measurement station in southern Africa. In this paper, the first 15 months of data collected at Welgegund is utilised to characterise the air masses arriving at the station from all the major anthropogenic source regions of the interior of South Africa, in terms of trace gases, black carbon, aerosol optical properties, as well as PM1 chemical composition and new particle formation. The source regions include the:

- western Bushveld Igneous Complex containing eleven pyrometallurgical smelters occurring within a ca. 55 km radius.
- Johannesburg-Pretoria conurbation with a population of more than 10 million people, making it one of the 40 largest metropolitan areas in the world.
- Vaal Triangle, where most of the South African petrochemical and related chemical industries are concentrated, together with other large point sources.
- Mpumalanga Highveld with eleven coal-fired power stations (all without de-SOx and de-NOx), with a combined generation capacity of ca. 20 GW, as well as a very large petrochemical plant, several pyrometallurgical smelters and numerous coal mines - all within a ca. 50km radius.
- anticyclonic recirculation source region, which is a geographical area with lower overall anthropogenic point source density, but where air masses recirculate from the major anthropogenic source regions towards the Welgegund site, which allows the possible investigation of aging of the anthropogenic source region emissions.
- regional background, which was defined as a sector that contained no significant point sources. Results from the anthropogenic source regions could therefore be compared with the regional background.

The above-mentioned source region plume characterisation will enable modellers and other atmospheric scientists to more accurately represent the different source regions of South Africa. Consecutively, this will result in improved modelling accuracy for this very under sampled, but important region.

S1.14 Secondary organic aerosol formation from fossil fuel and biogenic sources dominate summertime organic mass at Bakersfield, California

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Secondary organic aerosols (SOA), known to form in the atmosphere from oxidation of volatile organic compounds (VOCs) emitted by anthropogenic and biogenic sources, are poorly understood. In this study, we examined the chemical and physical properties of SOA at Bakersfield, a site influenced by anthropogenic and biogenic emissions. Factor analysis was applied to the infrared and mass spectra of fine particles to identify constituents that contributed to the organic mass (OM). We found that OM accounted for 56% of submicron particles, with SOA components contributing 80% to 90% OM from 15 May to 29 June of 2010. SOA formed from alkane and aromatic compounds, the two major classes of vehicle-emitted hydrocarbons, accounted for 65% OM. Specifically, alkane SOA likely formed via ozone-driven reactions, while OH radicals played an important role in forming aromatic SOA. The alkane and aromatic SOA components were associated with 200- to 500-nm-sized particles, which was consistent with condensation of gas-phase photochemical products during daytime. In contrast, biogenic SOA likely formed from condensation of secondary organic vapors, produced from NO3 radical oxidation reactions during nighttime hours, on 400- to 700-nm-sized primary particles. Sources from local petroleum operations, negligible in the current source inventory for southern San Joaquin Valley, accounted for about 13% OM. Vegetative detritus (10%) and cooking activities (7%) accounted for other small yet nonnegligible sources. While the mass spectra of several linearly-independent SOA components were nearly identical and external source markers were needed to separate them, each component had distinct infrared spectrum, which was source-specific and can be used as reference spectrum when external measurements are not available.

S2.1 Megacity ozone air quality under four alternative future scenarios

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The impact of the megacities of the world on global tropospheric ozone, and conversely, the extent to which megacities are influenced by emissions of ozone precursors from outside of the megacities is examined under the four alternative RCP ("Representative Concentration Pathway") emissions scenarios. Despite accounting for about 6% of present-day anthropogenic emissions of ozone precursor species, the contribution of emissions from megacities to global tropospheric ozone is calculated to be 0.84%. By 2100 this contribution falls to between 0.18% and 0.62% depending on the scenario, with the lower value being for the most-polluting of the four future emissions scenarios due to stringent controls on ozone precursor emissions from highly populated areas combined with a stronger tropospheric background ozone field. The higher end of this range is from the least-polluting of the four emissions scenarios, due to lower background tropospheric ozone combined with the use of a simpler downscaling methodology in the construction of the scenario, which results in higher emissions from megacities. Although the absolute impact of megacities on global ozone is small, an important result of this study is that under all future scenarios, future air quality in megacities is expected to be less influenced by local emissions within the cities, but instead more influenced by emission sources outside of the cities, with mixing ratios of background ozone projected to play an increasing role in megacity air quality throughout the 21st century. Assumptions made when downscaling the emissions scenarios onto the grids used in such modelling studies can have a large influence on these results; future generations of emissions scenarios should include spatially explicit representations or urban development suitable for air quality studies using global chemical transport models.

S2.2 WMO GURME addressing air quality in Megacities

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Megacities need climate, weather and environmental services in order to be resilient in withstanding environmental hazards. The World Meteorological Organization’s (WMO) Global Atmosphere Watch (GAW) Urban Research Meteorology and Environment (GURME) addresses air pollution questions in urban areas, in particular megacities, building observational and modelling capabilities in developing countries through pilot projects and transfer of scientific expertise. GURME contributes to improving capabilities to handle meteorological and related features of air pollution by addressing end-to-end aspects of air quality, linking observational capabilities with the needs of chemical weather prediction, with the goal of providing high quality air quality services.

In Latin America, GURME has assisted addressing air quality problems mainly in Mexico City, Santiago and in Sao Paulo, and connecting to other Latin American cities. This has consisted largely of organizing expert and training workshops on air quality modelling from 2002 to 2011.

The Shanghai Pilot Project, part of the WMO Shanghai multi-hazard early warning system project, was initiated in 2007 and was successfully implemented/demonstrated during the 2010 Expo. Activities included enhancing the observing system; enhancing air quality & weather & heat wave forecasting; a field experiment, joint with NCAR, to provide data to evaluate air quality models; and a variety of workshops and training activities. In this project an important step has been taken towards providing several types of health related forecasts. The project is now extended with the goal that an enhanced network will provide improved meteorological and air chemistry measurements that would greatly increase the ability to monitor the current state of atmospheric (and related Earth surface) conditions in order to better define various atmospheric-related risks and hazards and improve their mitigation through improved forecasts and predictions.

The SAFAR (System for Air Quality Forecasting and Research) pilot project, focusing on air quality forecasting in support of the Commonwealth Games (CWG) held in New Delhi, India, in October 2010, was established as a follow-up activity from the air quality forecasting training workshop held in 2008 for South Asia. This is the first air quality forecasting system demonstrated broadly to the public in India and successfully operated during the CWG. It is currently being further expanded to seven other Indian cities.

S2.3 Representing the air quality impacts of megacities in a global chemistry-climate model

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The continuing growth of the world's urban population has led to an increasing number of cities with more than 10 million inhabitants. Understanding the effect that future climate changes may have on air quality in these megacities is a challenge, particularly as current global models are typically used at a coarse resolution (approx. 150km) to enable analysis of several different climate scenarios. We have performed global model integrations using “UM-UKCA”, an atmospheric chemistry model, coupled to the UK Met Office Unified Model, at typical climate resolution (approx. 150km). Using chemical and meteorological initial conditions from the climate resolution run, we have additionally performed shorter, time-slice integrations at a higher resolution (approx. 40km). This enables us to investigate the representation of megacities in a global model, the dependence of this on both model and emissions resolution, and to evaluate the benefits and limitations of running such a high resolution global model for megacity future air quality scenarios.

The ability of this state-of-the-art model to represent the diurnal variation and probability distribution of gaseous pollutants such as ozone, nitrogen oxides and carbon monoxide in megacities such as London and Paris, as well as background pollution levels in rural locations, has been evaluated by comparison with UK and European observational datasets for the present day. Both summer and winter months for the year 2005 are considered and in particular we explore changes to chemical regimes both in the megacities themselves and in surrounding areas.

On a regional scale, the climate model integration is found to overestimate ozone values, whereas the higher resolution has a tendency to underestimate them. Using the higher model resolution is found to capture lower ozone values and a more

realistic diurnal cycle in urban areas such as the megacities of London and Paris. An investigation into the cause of these differences and the limitations of each model setup allows us to assess megacity representation in the global model, and hence assess the suitability of long, climate integrations to predict changes in air quality under future climate conditions.

S2.4 Long-term trends in motor vehicle emissions of gaseous pollutants in the usa, 1990-2010

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Over the last two decades, large changes have been observed in gasoline and diesel-powered vehicle emissions of carbon dioxide (CO_2), carbon monoxide (CO), volatile organic compounds (VOC), nitrogen oxides (NOx), and ammonia (NH_3). In this study, a fuel-based approach was used to estimate national and state-level emissions of gaseous air pollutants from on-road gasoline and diesel engines. Emission factors were derived from measurements using roadside emission spectrometers (aka remote sensing) as well as measurements in highway tunnels. Vehicle activity was measured by taxable fuel sales reported at the state and national level. Trends in non-methane VOC emissions were inferred using ambient air ratios of VOC to CO measured in special field studies. We compare fuel-based emission estimates from the present study to inventory estimates based on EMFAC, MOVES, and EDGAR models. Between 1990 and 2010 in the United States, gasoline consumption increased by 25% nationally while on-road use of diesel fuel increased by ~75%. Over the same time period, control of NOx emissions from diesel engines has lagged, such that diesel engines have become the largest source of NOx emissions. We also make comparisons between urban and rural areas in California. We show that in rural areas, diesel-powered vehicles have dominated on-road emissions of NOx for the last two decades. In urban areas, gasoline engines were the dominant NOx source up until 2006, after which time diesel engines dominated.

S2.5 BC/OC ratios : a new metrics to mitigate Emissions, Air quality, Health and Radiative Impacts. Focus on African megacities.

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Fossil fuel and biofuel emissions of particles in Africa are expected to significantly increase in the near future, particularly due to rapid growth of African cities. Air quality degradation is then expected with important consequences on population health and climatic/radiative impact.

In our group, we are constructing a new integrated methodology to study the relations between emissions, air quality and their impacts. This approach includes: (1) African combustion emission characterizations; (2) joint experimental determination of aerosol chemistry from ultrafine to coarse fractions and health issues (toxicology and epidemiology). (3) integrated environmental, health and radiative modeling. In this work, we show the central role of black carbon to organic carbon ratios (BC/OC) to characterize African anthropogenic emissions and impacts:

- a new BC and OC African anthropogenic emission inventory adapted to regional specificities has been constructed for the years 2005 and 2030.
- BC and OC radiative impacts in Africa have been modeled with TM5 model and Penner et al. (2011) radiative code for these inventories for 2005 and 2030 and for two emission scenarios: a reference scenario, with no further emission controls beyond those achieved in 2003 and a ccc* scenario including planned policies according to Kyoto protocol and regulations as applied to African emission specificities. In this study we will show that enhanced heating is expected with the ccc* emissions scenario in which the OC fraction is relatively lower than in the reference scenario.
- results of short term POLCA intensive campaigns in Bamako and Dakar in terms of aerosol chemical characterization linked to specific emissions sources and their inflammatory impacts on the respiratory tract through in vitro studies. In this study, organic carbon particles appear more biologically active than BC particles.

Quite importantly, air quality improvement obtained through regulations in the ccc* scenario with higher BC/OC ratios are accompanied by stronger heating impact. BC/OC ratio variations may be considered as a standard reference index to study air quality, health and climatic impacts.

S2.6 The growing dryland city of Urumqi (Xinjiang, China): A long term study (2000-2006) of emission sources, air pollution, and meteorological processes

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Seven years (2000–2006) of monthly PM_{10}, SO_2 and NO_2 concentrations are reported for Urumqi, the capital of the Uighur Autonomous Province Xinjiang in North-West China. Considerably high mean annual concentrations have been observed which ranged between 150–240μg·m^{-3} (PM_{10}), 31–50μg·m^{-3} (NO_2), and 49–160μg·m^{-3} (SO_2). The shapes of seasonal variation of all pollutants were remarkably similar, however winter/summer ratios of concentrations were quite different for PM_{10} (2–3) and NO_2 (approx.4) compared to SO_2 (up to 30).

Very high consumption rates of fossil fuels (particularly coal) for energy generation and domestic heating (during a five months long winter) are mainly responsible for high annual pollution levels, as well as the (very) high winter/summer ratios. Detailed analysis of the 2000–2006 records of Urumqi's meteorological data (surface and radiosonde observations) resulted in inter-annual and seasonal frequency distributions of (a) (surface) inversion events, (b) heights of surface inversions, (c) stability classes of Urumqi's boundary layer (Pasquil/Turner), and (d) the Wang & Angell "Air Stagnation Index (ASI)". Urumqi's boundary layer is shown to be characterized by high mean annual and seasonal frequencies of (surface) inversions and by the dominance of stable dispersion classes. A further outcome of the meteorological analysis is the proof of Urumqi's strong diurnal wind system (mountain-valley breeze) which might have particularly contributed to the stabilization of the nocturnal boundary layer. Annual and seasonal variations of pollutant's concentrations are discussed in the context of corresponding variations of occurrences of inversions, boundary layer stability classes, and ASI.

For the period 2006 – 2011, SCIAMACHY tropospheric NO_2 retrieval data are available as monthly means of the tropospheric NO_2 vertical column density (VCDs; 30×60 km). By suitable interpolation, data could be downscaled to grid cells of 8.1×11.2 km. Under the justified assumption that the entire satellite NO_2 signal is due to NO_2 molecules accumulated in Urumqi's boundary layer, NO_2 VCDs have been transformed to boundary layer averaged NO_2 concentrations. For 2006, these data have been compared with Urumqi's (city-wide averaged) surface concentrations. Particularly for the winter months, the agreement is remarkably good. Corresponding relationships between NO_2-VCDs and surface NO_2 concentrations have been used to study the further development of Urumqi's air pollution (2006-2011).

S2.7 Changes in Emissions and the Effect on Regional Ozone Production from the London Megacity

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Elevated levels of ozone, which are harmful to human health, have been observed in urban and rural locations. The main precursors to ozone formation are volatile organic compounds (VOCs) and nitrogen oxides (NOx). National and European legislation has targeted ozone precursor emissions, resulting in significant reductions over the past two decades. A significant amount of this regulation has focused on emissions of VOCs and NOx from road transport and petrol evaporation, important sources in urban areas. Understanding ozone precursors and being able to model them accurately to derive the impact of emissions changes is important for policy-makers and the improvement of air quality, and will lead to better predictions of SOA formation and zone levels.

Data from long-term monitoring sites in London are used here to determine trends in urban emissions for the past 10-12 years. Decadal and longer trends in VOCs, carbon monoxide, and NOx will be presented. This data is then used to investigate the changes in emissions for the city of London and the effect this has on the oxidizing environment of the megacity, and thereby the ozone production and concentrations in the city and region. These trends show that the oxidizing balance of London has changed dramatically. This change in the relationship of VOCs-NOx emissions was applied in a regional model. Results from the regional modelling and the effect on regional ozone production will be presented. Implications for policy and population exposure to ozone will be discussed.

S2.8 The Paris MEGAPOLI campaign to better quantify fine aerosol sources and formation in a tertiary type mid-latitude Megacity

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Within the EU MEGAPOLI project, two intensive field campaigns have been conducted in the Greater Paris region during July 2009 and January/February 2010. The major aim was to quantify sources of primary and secondary aerosol, and the interaction with gaseous precursors, within a large agglomeration, and in its plume. Greater Paris has been chosen for such a campaign because it is a major and dense pollution source (more than 10 million inhabitants), surrounded by rural areas and relatively flat terrain. A particular focus was put on carbonaceous aerosol, for which primary emissions and secondary formation are still not well quantified. Detailed aerosol and gaseous precursor measurements have been conducted at an urban and two sub-urban sites, from five mobile platforms and from the French ATR-42 research aircraft (for plume characterization), and allowed a detailed documentation of fine aerosol and related gaseous species sources and formation processes.

In this paper, the campaign set-up and objectives, and an overview over the major results obtained so far will be given. First, the regional/ local share of sources of fine aerosol are analysed from a set of AMS and PILS measurements obtained at several urban and peri-urban sites (located up or downwind of the agglomeration as a function of wind direction), and from air quality modelling. Despite the fact that the campaign took place in a Megacity with nearly 12 millions of inhabitants, the regional impact through advection from other European sources turned out to be dominant for fine aerosol (PM_1 and $PM_{2.5}$). Second, different source apportionment methods (Positive matrix factorisation of AMS and PILS measurements, ^{14}C analysis, specific chemical tracer methods) made evident a dominant modern carbon origin of organic aerosol (OA), in summertime due to biogenic secondary and cooking related OA, in wintertime due to a local and continental residential woodburning source.

Third, from more than ten airborne primary pollutant measurements during the summer campaign it appeared that the pollution plume was still well defined at more than one hundred kilometres downwind of the agglomeration. This gives a “safe” framework for evaluating secondary inorganic and organic aerosol build-up in the plume, which was shown to be significant for OA and nitrate, while for sulphate also shipping and industrial emissions from outside the agglomeration led to distinct peaks. Airborne and ground based measurements made evident frequent nucleation events, at the edge of the pollution plume. Besides, a full radical closure experiment could also be conducted during the summer campaign at a periurban site and showed radical (OH and ROx) levels in agreement, within the errors bars, with detailed 0D modelling using the MCM master mechanism. In addition, black carbon and NOy measurements within the plume were used to constrain the local emission inventory. The large set of observations is used for a thorough evaluation of 3D air quality models.

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S2.9 Emissions and Chemistry of Volatile Organic Compounds in a North American Megacity: Los Angeles, California

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During the CalNex study in May-June of 2010, an extensive set of volatile organic compounds (VOCs) was measured in the Los Angeles basin and its in- and outflow areas. Measurements were made from the NOAA WP-3D research aircraft, the research vessel Atlantis and a ground site in Pasadena, California. Here, an overview of the measurements is presented and the main results are discussed.

The VOC data from CalNex in 2010 are put in perspective by comparing them with results from an earlier flight of the NOAA WP-3D in the Los Angeles basin in 2002, from a number of other earlier studies and from two different air quality monitoring networks. Strongly decreasing trends of ~7% per year are observed for most VOCs that are emitted from motor vehicles or photo-chemically produced from these emissions. Decreasing trends are less strong for small alkanes, which are mostly from natural gas related emissions, and oxygenated VOCs produced from them.

The composition of urban VOC emissions was determined using the data obtained by gas-chromatography mass spectrometry at the ground site in Pasadena. Emission ratios of hydrocarbons versus ethyne were obtained using two methods. First, emission ratios were determined from nighttime data only. Second, the degree of photochemical processing of the sampled air masses was estimated, and emission ratios were determined by extrapolating to a zero photochemical age. Both methods agreed within the combined uncertainties for most VOCs. The composition of urban VOC emissions in Los Angeles was compared with other urban regions including in the U.S. and Europe and found to be similar.

Measurements of ethanol at the ground site in Pasadena showed much higher mixing ratios than observed earlier in the northeastern U.S. The difference is attributed in part to the strongly increased use of fuel ethanol in the U.S.: in 2010, about 10% of gasoline consisted of ethanol, whereas that percentage was about 1% in the early

2000s. The data were examined for the effects of increased ethanol use on acetaldehyde, one of its main atmospheric oxidation products. Any effects on acetaldehyde are overwhelmed by the general decrease in VOC emissions that took place over the same years.

During the daytime, the VOCs in the LA basin were significantly more aged than at the ground site at night. The degree of photochemical processing generally increased from West to East along with the prevailing daytime wind direction. During weekends, when NOx emissions are lower due to a reduction in Diesel traffic, the processing of VOCs occurred significantly faster.

The relative importance of emissions and photochemical formation of oxygenated VOCs is also studied using data from the ground site in Pasadena. It is found that aldehydes, ketones and glyoxal have significant direct emissions in addition to secondary formation sources. The direct emissions are not well represented in emission inventories. Acids have no or small direct emissions but significant secondary formation that is difficult to account for using our best understanding of the gas-phase chemistry. Alcohols have direct emission sources but no or small secondary formation.

S2.10 The evolution of air quality and monitoring in Santiago, Chile since the mid 1980's

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Santiago, Chile is located at 33˚27'S and 70˚40'W at an altitude of 500 m.a.s.l in a semi-arid basin (annual rainfall <350 mm) in the central part of Chile bounded by the high Andes. Santiago 's population surpasses 6 million inhabitants, and the city concentrates roughly 50% of the Chile's gross internal product. The climate of Santiago is characterized by the quasi-permanent influence of the subtropical Pacific high, and the intrusion of occasional cold fronts, which bring precipitation in wintertime. The South Pacific high determines quasi-stagnant anti-cyclonic conditions that are further intensified, especially in fall and winter by the presence of sub-synoptic features known as coastal-lows. There is a characteristic radiatively driven circulation that defines up-slope southwesterly winds in the afternoon and down-slope northeasterly winds in the night and morning hours, more strongly so in the summer. Santiago shows a rather continuous record of criteria pollutants since the late 1980's. This work addresses, on the one hand, the evolution of Santiago's air quality in terms of concentrations of pollutants, and, on the other hand, the evolution of the monitoring network both regarding sampling ability and instrumentation. Sampling ability is approached by means of statistical and variational analysis. Instrumentation is described in a qualitative manner, and it emphasizes the need of integration of measurements from various platforms and models.

S2.11 Improving the view of air quality from space: An overview of DISCOVER-AQ observations over the Baltimore-DC area during July 2011

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DISCOVER-AQ is one of five NASA Earth Venture-1 airborne science projects scheduled to occur in the 2011-2015 timeframe. These five-year, targeted science investigations are part of NASA's Earth System Science Pathfinder program. The goal of DISCOVER-AQ is to improve the interpretation of satellite observations to diagnose near-surface conditions relating to air quality. Distinguishing between pollution at the surface and aloft is a particularly difficult problem for satellites. The strategy for studying this problem is articulated in the acronym which stands for Deriving Information on Surface conditions from COlumn and VERtically resolved observations relevant to Air Quality. This strategy dictates an observational approach that enables systematic and concurrent observation of column-integrated, surface, and vertically-resolved distributions of aerosols and trace gases relevant to air quality as they evolve throughout the day. The information gathered is expected to improve the interpretation of current satellites and contribute to the implementation of future observations from geostationary orbit. Initial flights were conducted over the Baltimore-Washington area during July 2011 as the project completed the first of four planned deployments. Two NASA aircraft were flown over six instrumented ground sites on fourteen days. The NASA UC-12 flew a circuit at high altitude (26 kft) passing over individual ground sites five to six times on each flight day, providing lidar observations of aerosols and passive remote sensing of trace gases. Concurrently, the NASA P-3B conducted in situ sampling of trace gases and aerosols along a circuit that allowed for three profiles of the lower atmosphere (typically from 10,500 to 1000 feet) over each ground site as well as low-altitude transects along the I-95 traffic corridor at 1000 feet. The six ground sites were chosen from the long-term monitoring network maintained and operated by the Maryland Department of the Environment. These sites also hosted additional instrumentation that included remote sensors (sunphotometers and trace gas spectrometers), instrumented trailers, ozonesondes, and tethered balloons. Partners also contributed additional airborne observations, ground lidars, ship-based measurements over the Chesapeake Bay, and satellite observations. Selected data and preliminary findings are presented as well as an introduction to the DISCOVER-AQ data archive (http://www-air.larc.nasa.gov/missions/discover-aq/discover-aq.html) which is open to anyone interested in using the data for their own research.

S2.12 Remote Sensing of Spatial Distributions of Greenhouse Gases in the Los Angles Basin

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The Los Angeles air basin is a significant anthropogenic source of major greenhouse gases (CO_2, CH_4, CO and N2O), contributing significantly to regional and global climate change. Recent legislation in California, the California Global Warming Solutions Act (AB32), established a statewide cap for greenhouse gas emissions for 2020 based on 1990 emissions. Verifying the effectiveness of regional greenhouse gas emission control requires high-precision, regional-scale measurement methods combined with models that capture the principal anthropogenic and biogenic sources and sinks. The existing database in the Los Angeles area is extremely sparse, and new methods are required to provide high spatial and temporal resolution.

We present a novel approach for monitoring the spatial distribution of greenhouse gases in the Los Angeles basin using high-resolution remote sensing spectroscopy. A new Fourier Transform Spectrometer called CLARS-FTS has been deployed at Mt. Wilson, California at JPL's California Laboratory for Atmospheric Remote Sensing for automated long-term measurements of greenhouse gases since May 2010. The CLARS-FTS points at ground sites in the Los Angeles basin from its location at an altitude of 1.7 km. It scans through the ground sites once per hour with an averaged ground pixel size about 200 meters in diameter. It records atmospheric absorption spectra over a broad spectral range (4000 – 14000 cm-1) in reflected sunlight. From these rotationally resolved spectra, the column densities of trace gases are retrieved along the line of sight.

Using the measured column densities of greenhouse gases and molecular oxygen, we computed the column-averaged dry-air mole fractions [XGAS] including XCO_2, XCH_4, XN_2O and XCO. The strong correlations between these species are observed which are attributed to co-emission from sources and to boundary layer effects. The seasonal cycles of CO_2 and CH_4 over LA basin have been captured by CLARS-FTS measurements since August 2011 till up to date. These correlations can be used to derive relative emission rate factors. Contour maps of [XGAS] from the CLARS-FTS measurements reveal the temporal and spatial variations of greenhouse gases in the LA basin.

S2.13 Megacity NOx emissions and lifetimes probed from space

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Megacity emission inventories, based on bottom-up estimates, are still highly uncertain, in particular in developing countries. Satellite observations have been demonstrated to allow regional and global top-down emission estimates of nitrogen oxides ($NOx=NO+NO_2$), but require poorly quantified a-priori information on the lifetime of NOx.

Here we present a new method for the determination of megacity NOx emissions and lifetimes from satellite measurements. Mean patterns of NO_2 tropospheric columns are analyzed separately for a set of different wind direction sectors. From the combined use of the observed total burden and the downwind evolution of NO_2, mean NOx photochemical lifetimes and total emissions are derived simultaneously.

Typical daytime lifetimes of about 4 hours are found for several megacities at low and mid-latitudes, corresponding to mean OH concentrations of ~6e6 molec/cm3. The derived emissions are generally in good agreement with bottom-up inventories, but are significantly higher in e.g. the case of Riyadh (Saudi Arabia).

The presented method works best for isolated “hot spots” of NOx emissions. For megacities in the vicinity (in terms of some hundred km) of other strong sources, like e.g. Paris, or for highly populated regions like East China, modified approaches are necessary. Different approaches are discussed, and the estimated emissions+uncertainties are set in relation to emission inventories.

S2.14 Economic crisis detected from space: Trends in air quality of Athens in Greece.

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Athens, the capital of Greece, gathers about 35% of the total population of the country. In the past, it has been identified to be among the most polluted capitals worldwide owing to: i) the extensive number of registered vehicles (2.7M private cars, 0.7M motorcycles and 0.3M trucks), ii) the existence of industrial regions close to the city and iii) the complex topography of the area, favouring pollutant accumulation. As a result, this densely populated area (up to 16 thousand citizens per square kilometre) suffers from high levels of pollutants emanating mainly from anthropogenic sources.

One of the major pollutants affecting air quality in Athens is nitrogen dioxide (NO_2). NO_2 belongs to the family of nitrogen oxides ($NOx=NO+NO_2$). Nitrogen oxides play a key role in determining air quality, with direct and indirect consequences on Earth's atmosphere and human health. Due to its environmental importance, the spatial and temporal variability of NO_2 over Athens has been thoroughly investigated with the use of satellite and in situ observations. Specifically, the spectral information recorded by the SCIAMACHY, GOME-2 and OMI spectrometers on board the ENVISAT, MetOP and Aura satellites, respectively has been used for retrieving the tropospheric NO_2 columns with the Differential Optical Absorption Spectroscopy (DOAS). Additionally, in-situ observations of surface NO_2, NO, carbon monoxide (CO) and sulfur dioxide (SO_2) mixing ratios, obtained from the Ministry of Environment, Energy and Climate Change at various locations in Athens, covering urban and suburban regions, were analyzed.

The studied period covers the years before the beginning of the economic crisis until 2010. The multiannual analysis of the tropospheric column of NO_2 over Athens shows that NO_2 has been significantly reduced by 32%, 38% and 40% according to the SCIAMACHY, GOME-2 and OMI analysis, respectively. This decline is supported by the in-situ NO2 mixing ratios of the period 2007-2010; on average a 30% reduction of surface NO_2 levels is recorded over the city-centre, mirrored by a decrease of the ambient NO_2 levels by 12 ppbv. Similarly, at suburban areas the observed mean reduction is 32%, and the respective decrease of the NO_2 levels ranges between 1.5 and 8 ppbv, depending on the site. NO2 is not the only pollutant significantly reduced; a decrease is also observed for CO (by 21%), SO_2 (by 55%), and aerosol optical depth levels, AOD (by 10%). In contrast, an average increase in ozone levels by 13% is recorded, arguably owing to the decreased nitrogen monoxide mixing ratios by 25%.

Trend analysis of basic pollutant levels over Athens in the last decade (2000-2010), revealed that the reduction of NO_2 ($0.25 ppbv \cdot y^{-1}$) and SO_2 ($0.33 ppbv \cdot y^{-1}$) during the period 2000-2007, has been significantly accelerated in the economic crisis period (2008-2010), reaching $2\ ppbv \cdot y^{-1}$ and $1\ ppbv \cdot y^{-1}$, respectively. Indeed lower incomes, in addition to the continuous increase of oil price, led citizens to minimize the use of cars, and contributed to an enhancement in the use of natural gas, for domestic heating purposes. Moreover, a large number (up to 30%) of small-scaled industries and enterprises around Athens ceased their activities potentially adding to the above mentioned SO_2 reduction.

S3.1 Air Quality in the US under Current and Future Climate Conditions

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Elevated concentrations of tropospheric ozone (O_3) and particulate matter with diameters less than 2.5 µm ($PM_{2.5}$) depend highly on local meteorological conditions and emissions. In the future, these conditions are expected to change as a result of climate change and economic growth. To better understand the implication of these changes to ambient O_3 and $PM_{2.5}$ concentrations, we conducted a comprehensive modeling effort that is briefly described below. In this modeling effort we explicitly evaluated the effect of; biogenic emissions, land use changes, wildfires, global/regional anthropogenic emissions on O_3 and $PM_{2.5}$ concentrations. In addition, we investigated the current and future background concentrations of O_3 and $PM_{2.5}$ in the absence of anthropogenic emissions in North America.

Results from a global climate model driven with the A1B emission scenario from the IPCC were downscaled using a regional climate model to provide the required meteorological fields. These meteorological fields were used to calculate current and future biogenic and wildfire emissions. Current global emissions of O_3 and $PM_{2.5}$ precursors from anthropogenic, natural, and biomass burning were estimated by the POET and EDGAR emission inventories, whereas regional anthropogenic emissions were obtained from the US EPA NEI2002 emission inventory. Global emissions for black and organic carbon were obtained from Bond et al (2004). Future global emissions of O_3 and $PM_{2.5}$ precursors were obtained by adjusting the global inventories to the IPCC A1B emission scenario, while regional emissions were projected to the future using the Market Allocation system dynamics model (MARKAL). We developed air quality simulations using the Community Multiscale

Air Quality (CMAQv4.7) for two nested domains with 220km and 36km horizontal grid cell resolution for a semi-hemispheric domain and a continental US domain, respectively. The coarse domain was used primarily to generate the chemical boundary conditions for the 36-km domain simulations.

Preliminary results suggest that O3 will increase in a range between 2 to 12 ppb across most of the domain, with the higher increase in the South, Central and Midwest regions of the US due to increase in temperature, enhanced biogenic emissions and changes in land use. On the other hand, simulations show localized increases of $PM_{2.5}$ between 2 and 4 μg/m3 at the Northeast, Southeast and South regions, mostly as a result of enhanced biogenic emissions and Land Use. On the contrary, the Northwest and Central regions experienced reductions in $PM_{2.5}$ due to regulatory curtailment and increase in precipitation.

In conclusion, our simulations show that under the A1B IPCC emission scenario, the considered changes in US emissions due to regulatory curtailment are not significant, and they don't result in substantial reduction in O_3 and $PM_{2.5}$ concentration. In addition, increased background concentrations of O_3 will complicate the effort of air quality managers to maintain air pollutants under safety standards.

S3.2 Long Term Trends in Ozone in the Southern Hemisphere Extra-tropical Boundary Layer and Free Troposphere

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Ozone in the troposphere has five important roles. Tropospheric ozone is a greenhouse gas, ranked third in anthropogenic radiative forcing after CO_2 and CH_4; ozone in the troposphere makes up an important fraction of the atmospheric shield against ultraviolet radiation particularly when ozone depletion events occur and when the total column abundance of ozone drops below 200DU, and tropospheric ozone drives tropospheric chemistry being the major precursor of the OH radical which is the sink for gases such as methane. In the boundary layer increases in background ozone have other impacts through urban and regional photochemical smog on human health and through plant damage on crop productivity and the sequestration of carbon into the biosphere.

There is very limited long-term monitoring and are few process studies of tropospheric ozone in the Southern Hemisphere. Due to the very different mixes of anthropogenic and natural sources, different regimes of tropospheric ozone may occur in each hemisphere.

We present here analyses of surface ozone from 6 long-term background stations spanning 34˚S to 90˚S and tropospheric ozone from 5 long-term ozonesonde stations spanning 38˚S to 69˚S.

The measurements in surface air reflect a NOx depleted environment were there is net ozone loss at most locations. This is reflected in the free troposphere where the annual ozone maximum is in winter/spring and minimum in summer compared with the Northern Hemisphere where the tropospheric ozone annual maximum is in spring/summer at many locations. Long-term trends in the mid-latitudes of the Southern Hemisphere and at the South Pole appear to be decoupled.

Analyses of the contributions of various processes to the observed Southern Hemisphere tropospheric ozone trends will be discussed. These include (a) dynamical effects including ENSO, SAM and (b) chemical processes such as changes in biomass burning and methane trends in the Southern Hemisphere.

Tropospheric ozone in the extra-tropical regions of the two hemispheres is significantly different and the southern hemisphere should give greater insight into pre-industrial tropospheric ozone and the effect on tropospheric ozone of the global methane perturbation.

S3.3 Development of a mechanism generator and its application for the CAPRAM mechanism extension

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Organic compounds are ubiquitous in the tropospheric multiphase system. With either large biogenic and anthropogenic sources, they play an important role and have thus become a major research topic within the last decades.

The oxidation of volatile organic compounds (VOCs) leads to a great variety of multifunctional compounds with a decreased vapour pressure, which will partition into deliquescent particles and cloud droplets. There, they get further oxidised leading to new products, which are released to the gas phase changing the chemical composition in return.

Modelling can provide a useful tool to explore complex multiphase chemical systems and their interactions with the cloud microphysics. Therefore, the Chemical Aqueous Phase RAdical Mechanism CAPRAM 3.0i has been developed comprising 777 reactions and 52 species undergoing phase transfer. It is coupled to the Master Chemical Mechanism MCM with 11381 reactions in the gas phase.

The current study was aimed at expanding the organic C3 and C4 chemistry in the aqueous phase. However, the oxidation of larger compounds will lead to a strongly increased number of species and reactions, which results in the necessity of automated mechanism construction. Due to missing experimental kinetic data, estimation methods are needed. An evaluation of suitable prediction methods such as structure-activity relationships (SARs) has been performed. On the basis of the results of this evaluation and kinetic data from the literature, a protocol for automated mechanism has been designed and used to advance the CAPRAM mechanism. This protocol was implemented in the framework of the Generator for Explicit Chemistry and Kinetics of Organics in the Atmosphere (GECKO-A). A mechanism with more than 2500 additional aqueous phase reactions and equilibria was constructed. Several sensitivity studies accompanied the mechanism construction to refine the protocol.

Box model studies have been performed with the extended CAPRAM/GECKO-A mechanism to investigate important key issues of tropospheric multiphase chemistry. A scenario with non-permanent clouds was used to investigate the processing of tropospheric particles and their modification by clouds as well as the cloud chemistry itself. The investigation of the concentration profiles of important organic species was accompanied by detailed time-resolved flux analyses, which led to a better understanding of the production and destruction pathways of the species considered.

With the advanced mechanism several improvements were achieved. The use of SARs easily allows for branched OH attacks, while in the previous CAPRAM version only the main channel was implemented. Also, a new hydration routine has been adopted, which described e.g. the hydrolysis of oxo-succinic acid. According to the implemented SARs, the main channel is surprisingly the H-abstraction at the hydroxyl group although the bond dissociation energy is very high. The alkoxy radical formed will decay to malonic acid, raising its concentration to about 80 $ng\cdot m^{-3}$. These results are more consistent with measurements from field studies than previous model results with only a few ng m 3 malonic acid in the aqueous phase.

S3.4 Subseasonal Variability in the Transport of Continental Aerosol to the Remote Indian Ocean: Possible Relation to the MJO

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The Dynamics of the Madden-Julian Oscillation (DYNAMO) field campaign took place in the equatorial Indian Ocean during the fall of 2011. The goal of the field mission was to increase the scientific understanding and prediction of the forces behind the Madden-Julian Oscillation (MJO), a period of intraseasonal weather instability that propagates eastward starting in the Indian Ocean and is the largest source of sub-seasonal weather variability in the tropics. As part of this field mission the R/V Revelle (Scripps Oceanographic Institute) was stationed at 80 degrees E on the equator, a remote marine location in the Indian Ocean. The instrumentation on board the R/V Revelle was chosen to obtain a detailed profile of the local ocean and atmosphere from 200 m above the ocean surface to 200 m below the ocean surface before, during, and after MJO events. One subset of these measurements was continuous measurements of the chemical, physical, and optical properties of sub and supermicron aerosols in the local atmosphere. These measurements included real-time and filter-based analysis of aerosol chemical composition, number size distributions from the Aitken to the coarse, particle number concentrations, aerosol scattering and absorption, cloud condensation nuclei (CCN) concentrations, and total sub- and supermicrometer aerosol mass.

Aerosols can perturb the earth’s radiation balance directly through the scattering (e.g. by sea salt) and absorption (e.g. by soot) of incoming solar radiation and indirectly through aerosol-cloud interactions that can change the microphysical properties, lifetime, and extent of clouds. The aerosol source affects the chemical, physical, and optical properties of the aerosol, thus controlling its climate forcing capability and influence on local weather patterns. The positioning of the R/V Revelle allowed the measurement of both clean marine and aged continental aerosol. Over the course of DYNAMO we observed a distinct change in the circulation pattern around the research station and a parallel change in the aerosol’s chemical, microphysical and optical properties. During the first half of the field mission in October, long periods of low aerosol concentration were interspersed with increased sulfate concentrations introduced from the upper troposphere by downward mixing. Changes in circulation patterns during November and December, possibly tied to a strong MJO event and a tropical cyclone observed in late November, drew more aged continental aerosol to the field site. Increased absorption and measurable elemental carbon in several aerosol events during this time period suggest an anthropogenic influence. HYSPLIT back trajectories and radon measurements confirm a continental aerosol source.

Here we compare the background marine aerosol properties in the early fall to the aged continental aerosol properties during the late fall and discuss the transport of anthropogenic pollutants to marine environments far from primary emission sources. From this work, anthropogenic emissions are shown to have a far-reaching impact that varies from season to season in remote marine environments. Seasonal circulation patterns, the MJO, and cyclone activity may increase anthropogenic aerosol transport. These factors, coupled with increased aerosol during the Asian dry season, may introduce significant anthropogenic pollutants into the equatorial Indian Ocean.

S3.5 Bounding the role of black carbon in climate: A scientific assessment

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Bounding the role of black carbon in the climate system" is an assessment conducted as part of the IGAC Atmospheric Chemistry and Climate Initiative. Black carbon aerosol plays a unique and important role in Earth's climate system due to its unique combination of physical properties. It has both natural (biomass burning) and anthropogenic (biofuel and fossil fuel combustion) sources. Measurement and modeling studies have provided a range of estimates of climate warming due to light absorption by black carbon in the atmosphere and in surface snow and ice. Our assessment provides an evaluation of black-carbon climate forcing that is comprehensive in its inclusion of all known and relevant processes and that is quantitative in providing best estimates and uncertainties of the main forcing terms: direct solar absorption, influence on liquid, mixed-phase, and ice clouds, and deposition on snow and ice. These effects are calculated with models, but when possible, they are evaluated with both microphysical measurements and field observations. Direct radiative forcing alone does not capture important rapid adjustment mechanisms that contribute to forcing. A framework is described and used for quantifying climate forcings and their rapid responses and feedbacks. Sources that emit black carbon also emit other short- lived species that may either cool or warm climate. Climate forcings from co-emitted species are estimated and used in the framework described herein. Net forcing is calculated for the principal effects of BC plus co-emissions, including cooling agents such as sulfur dioxide for source categories as well as for specific BC-rich sources within those categories. The uncertainties in net climate forcing from black-carbon-rich sources are substantial, largely due to lack of knowledge about cloud interactions with both black carbon and co-emitted organic carbon. In prioritizing potential black-carbon mitigation actions, non-science factors, such as technical feasibility, costs, policy design, and implementation feasibility play important roles. The major sources of black carbon are presently in different stages with regard to the feasibility for near-term mitigation. This assessment, by evaluating the large number and complexity of the associated physical and radiative processes in black-carbon climate forcing, sets a baseline from which to improve future climate forcing estimates.

S3.6 Black Carbon Deposition and High Asia Glaciers: An Interaction between Human and Nature Revealed by Pollutants

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Black carbon (BC) is now concerned as one of the most significant factors impacting environment and climate mainly from anthropogenic activities. One aspect of its impacts is that the accelerated snow and ice in nature when BC is deposited in their surfaces. Since 2000, 18 High Asia glaciers have been surveyed for black carbon deposition 22 times, and numerous snow samples and ice cores have been collected by researchers. However, most of the results were interpreted individually in papers. Here, we assemble the data and discuss the distribution of BC deposition and its impacts on the melting of the glaciers through radiative forcing. We find that BC distribution on the surfaces of High Asia glaciers primarily depends upon their elevations (i.e., higher sites have lower concentrations) and then on regional BC emissions associated with surface melting conditions. BC concentrations in High Asia glaciers are similar to the Arctic and western American mountains but are significantly less than heavy industrialized areas such as northern China. Although Himalayan glaciers, which are important due to their water resources, are directly facing the strong emissions from South Asia, their mean BC is the lowest due to high elevations. A new finding indicated by ice core records suggested great valleys in the eastern Himalayan section are effective pathways for BC entering the Tibetan Plateau and makes increasing BC trends in the local glaciers. On average, BC deposition causes a mean forcing of ~6 W m-2 (roughly estimated 5% of the total forcing) in High Asia glaciers and therefore may not be a major factor impacting the melting of most glaciers.

S3.7 Multi-decadal trends of atmospheric aerosols and their effect on surface radiation

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We present an investigation on multi-decadal changes of atmospheric aerosols and their effects on surface radiation using a global chemistry transport model along with the near-term to long-term data records. We focus on a 30-year time period of satellite era from 1980 to 2009, during which a suite of aerosol data from satellite observations, ground-based measurements, and intensive field experiments have become available. We analyze the long-term global and regional aerosol trends and their relationship to the changes of aerosol and precursor emissions and assess the role aerosols play in the multi-decadal change of solar radiation reaching the surface (known as "dimming" or "brightening") at different regions of the world, including the major anthropogenic source regions (North America, Europe, Asia) that have been experiencing considerable changes of emissions, dust and biomass burning regions that have large interannual variabilities, downwind regions that are directly affected by the changes in the source area, and remote regions that are considered to representing "background" conditions.

S3.8 Wet removal of black carbon in Asian outflow: Aerosol Radiative Forcing in East Asia (A-FORCE) aircraft campaign

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The Aerosol Radiative Forcing in East Asia (A-FORCE) aircraft campaign was conducted over East Asia in March-April 2009. During the A-FORCE campaign, 120 vertical profiles of black carbon (BC) and carbon monoxide (CO) were obtained in the planetary boundary layer (PBL) and the free troposphere. This study examines the wet removal of BC in Asian outflow using the A-FORCE data. The concentrations of BC and CO were greatly enhanced in air parcels sampled at 3-6 km in altitude over the Yellow Sea on 30 March 2009, associated with upward transport due to a cyclone with modest amounts of precipitation over northern China. In contrast, high CO concentrations without substantial enhancements of BC concentrations were observed in air parcels sampled at 5-6 km over the East China Sea on 23 April 2009, caused by uplifting due to cumulus convection with large amounts of precipitation over central China. The transport efficiency of BC (TEBC, namely the fraction of BC particles not removed during transport) in air parcels sampled above 2 km during the entire A-FORCE period decreased primarily with the increase in the precipitation amount that air parcels experienced during vertical transport, although their correlation was modest. TEBC also depended on the altitude to which air parcels were transported from the PBL and the latitude where they were uplifted locally over source regions. The median values of TEBC for air parcels originating from northern China (north of 33N) and sampled at 2-4 km and 4-9 km levels were 86% and 49%, respectively, during the A-FORCE period. These median values were systematically greater than the corresponding median values (69% and 32%, respectively) for air parcels originating from southern China (south of 33N). Use of the A-FORCE data set will contribute to the reduction of large uncertainties in wet removal process of BC in global- and regional-scale models.

S3.9 Quantifying above-cloud aerosols through integrating multi-sensor measurements from A-Train satellites

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Quantifying above-cloud aerosols can help improve the assessment of aerosol intercontinental transport and climate impacts. Large-scale measurements of aerosol above low-level clouds had been generally unexplored until very recently when CALIPSO lidar started to acquire aerosol and cloud profiles in June 2006. Despite

CALIPSO's unique capability of measuring above-cloud aerosol optical depth (AOD), such observations are substantially limited in spatial coverage because of the lidar's near-zero swath. We developed an approach that integrates measurements from A-Train satellite sensors (including CALIPSO lidar, OMI, and MODIS) to extend CALIPSO above-cloud AOD observations to substantially larger areas. We first examine relationships between collocated CALIPSO above-cloud AOD and OMI absorbing aerosol index (AI, a qualitative measure of AOD for elevated dust and smoke aerosol) as a function of MODIS cloud optical depth (COD) by using 8-month data in the Saharan dust outflow and southwest African smoke outflow regions. The analysis shows that for a given cloud albedo, above-cloud AOD correlates positively with AI in a linear manner. We then apply the derived relationships with MODIS COD and OMI AI measurements to derive above-cloud AOD over the whole outflow regions. In this talk, we will present spatial and day-to-day variations of the above-cloud AOD and the estimated direct radiative forcing by the above-cloud aerosols.

S3.10 Pole-to-Pole Observations of Black Carbon Aerosol in the Remote Pacific

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Aerosol and trace gas measurements from the HIAPER Pole-to-Pole Observations (HIPPO) study provide insight into the role of synoptic-scale variability on the intercontinental transport of pollutants between Asia and North America. Five HIPPO campaigns with the NSF/NCAR G-V aircraft have been completed over all four seasons and include over 600 vertical profiles from 0.3 to 14 km altitude between 85°N and 67°S latitude in the remote Pacific and Arctic regions. The aerosol observations in the northern hemisphere (NH) Pacific exhibit large variability between and also within each season. Very polluted conditions were encountered during NH spring over a large depth of the troposphere between 0.3 and 8 km altitude associated with large-scale pollution plumes advected across the Pacific at midlatitudes and in the subtropics from anthropogenic and biomass-burning sources in Asia. Observations of black carbon (BC) mass loadings across the intertropical convergence zone show large interhemispheric gradients in all seasons but especially in NH spring. The NH BC mass loadings account for over 90% of the pole-to-pole burden of BC mass in the remote Pacific during this time of year. One of the goals of this analysis is to examine aerosol-cloud-precipitation interactions and identify and characterize the removal of aerosols in major precipitation events. Here we first present the HIPPO observations and then analyze these data in the context of the large-scale meteorological flow and satellite-derived precipitation patterns to address this and other potentially important impacts of high loadings of anthropogenic and biomass-burning aerosol exported from Asia.

S4.1 Science in support of decision-making concerning particulate matter air quality legislation: the European example

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The European Commission (EC) is preparing a comprehensive review of the existing air quality legislation, which should be finalised by 2013. In this context, the EC is coordinating a consultation process in the course of which EC-funded projects in the field of air quality, climate change and health aspects included, are asked to provide science-based evidences relevant for the review process. Within this comprehensive effort, this communication concerns the aspects connected to particulate matter (PM) effects on air quality and human health, including the synergies between air quality and climate change policy. In fact, there is now sufficient evidence that certain PM constituents (e.g. black carbon) have a high radiative forcing potential, thereby contributing to the warming up of the atmosphere in the short term. The exercise is organised around the answers of the science community to five key questions: i) which PM size fraction should be the focus of new regulatory effort? ii) do we know the link between aerosol composition and health effects? iii) what are the most important PM sources to control and in what order? iv) can the control of PM sources be regulated to maximise the benefits for health and climate effects? e) hot spots or the background, which should be the priority for control of PM sources and precursors? The answers to these five questions will be discussed in the context of the results provided by the most recent European and international scientific achievements.

The authors of this communication are coordinating the effort to provide policy makers with science-based answers to the above five key questions on PM, with the crucial support from several relevant EC-funded research projects.

S4.2 Cardio-Pulmonary Responses of Healthy Young Adults to Source-Specific Particulate Constituents during the Beijing Olympics

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Rational: Unprecedented pollution control actions during the Beijing Olympics provided a quasi-experimental opportunity to examine biological responses to source-specific particulate level changes in air pollution levels.

Objectives: To determine whether changes in levels of cardio-pulmonary biomarkers reflecting autonomic and vascular function, respiratory pulmonary inflammation and oxidative stress burden in both respiratory tract and in the whole body, were associated with changes in source-specific particulate levels in healthy young adults.

Methods: We measured heart rate variability (HRV), blood pressure (BP), fractional exhaled NO (FeNO), a number of exhaled breath condensate (EBC) markers (H^+, nitrite, nitrate, and 8-isoprostane), and urinary 8-OHdG in 125 participants twice in each of the pre- (high pollution), during- (low pollution), and post-Olympic (high pollution) periods. We measured concentrations of particulate and its constituents, and gaseous pollutants near where the participants lived and worked. We used mixed-effects models to estimate changes in biomarker levels across the three periods and to examine whether changes in biomarker levels were associated with changes in particulate and its constituents concentrations, adjusting for meteorological parameters. We used factor analyses to determine the primary sources of the particles.

Preliminary Results: Concentrations of air pollution decreased substantially from the pre- to the during-Olympic period, except O_3. Concurrently, we observed decreases in mean levels of BP, FeNO, EBC nitrate, nitrite, nitrate plus nitrite, hydrogen ion, and decreases in the fraction of above-detection-limit for EBC 8-isoprostane and urine concentrations of 8-OHdG, but not in HRV. However, after adjustments for covariates, the changes in all the biomarker levels were significantly associated with changes in the ambient particulates and the constituents from industrial combustion and transportation sources.

Summary: Our findings suggest that source-specific constituents in particles may play important role mediating adverse cardio-pulmonary and systemic oxidative responses in general population without chronic diseases. The underlying pathophysiologic mechanisms should be further investigated.

S4.3 SiO_2 nanoparticles stimulate inflammatory reaction in j774.4 macrophages cell line and basal insulin secretion in Min 6 cell line

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This study examines the cytotoxicity of 20nm SiO_2 nanoparticles (NPs) towards macrophage cell lines and insulin secreting cells. Exposure of macrophage cell line (j774) to SiO_2 NPs leads to expression of IL-1β, IL-6 and Il-8. Once secreted to the blood stream, cytokines may cause beta cell stress and eventually death. Direct exposure of insulin secreting cells (Min6) to SiO_2 NPs for 2.5h induces basal insulin secretion independent of glucose presence. To revoke the possibility that insulin secretion was caused by membrane damage and consequent leakage, we performed MTT and cytotoxicity assays for 2, 4 and 24h. A decrease in cell viability occurred only after 24h, suggesting that the observed insulin secretion was not caused by membrane leakage. To understand the molecular mechanism for the observed basal insulin secretion, we checked different components of the endoplasmic reticulum (ER) stress signaling pathway. After 2h of exposure we found a minor elevation in XBP, CHOP and ATF-4 genes, and a more significant increase after exposure for 4h. These results suggest that silica nanoparticles can induce inflammatory responses in macrophages and perturb basal insulin secretion that may be caused by ER stress. Possible implications for diabetic conditions are discussed.

S4.4 Systemic inflammatory changes, oxidative stress and increased cardiovascular risk in rural Indian women cooking with biomass fuels

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Objectives: The study was undertaken to investigate whether cumulative exposure to biomass smoke aggravates systemic inflammation and oxidative stress that might result in increase in the risk of developing cardiovascular disease (CVD) in rural Indian women compared to those cooking with a cleaner fuel like liquefied petroleum gas (LPG).

Methods: A total of 244 women (median age 34 years) who cooked with biomass and 236 age-matched control women who cooked with LPG were enrolled. Serum interleukin-6 (IL-6), C-reactive protein (CRP), tumor necrosis factor-alpha (TNF-α) and interleukin-8 (IL-8) were measured by ELISA as markers of systemic inflammation. Oxidative stress was assessed as generation of reactive oxygen species (ROS) by leukocytes (measured by flow cytometry) and depletion of erythrocytic superoxide dismutase (SOD; measured by spectrophotometry). Hypertension (diagnosed following the Seventh Report of the Joint Committee), plasma level of oxidized low-density lipoprotein (oxLDL; measured by ELISA), platelet hyperactivity (platelet P-selectin expression measured by flow cytometry; platelet aggregation measured by aggregometer), and serum autoantibody against cardiolipin (aCL IgG and aCL IgM measured by ELISA) were examined to evaluate risk of CVD. Particulate matter of diameter less than 10 and 2.5µm (PM_{10} and $PM_{2.5}$, respectively) in cooking areas was measured using real-time aerosol monitor.

Results: Compared with control, biomass users had 3-times more particulate pollution in indoor air, significantly elevated levels of IL-6, IL-8, TNF-α, CRP, oxLDL, aCL IgM, aCL IgG, increased platelet P-selectin expression and platelet aggregation, greater prevalence of hypertension, increased ROS generation and depleted SOD compared to their LPG-using neighbors. After controlling potential confounders, the changes were positively associated with PM_{10} and $PM_{2.5}$ in indoor air, suggesting a positive association between indoor air pollution (IAP) and increased cardiovascular risk.

Discussion: Our study has shown that systemic inflammation, oxidative stress, and known risk factors of CVD are exacerbated in poor women cooking with biomass fuel and hence, they are predispose to increased risk of CVD development compared to the women cooking using LPG. Systemic inflammation and oxidative stress may be the mechanistic factors involved in the development of CVD.

Conclusion: The study showing high risk of developing CVD among poor, underprivileged women in rural India is important from public health perspectives. It may motivate the government and the regulatory agencies of the country to take a serious note of the IAP from biomass fuel use as it threatens the health of millions of women, children and the elderly who mostly stay indoors. We hope the findings will strengthen the demand for setting up a standard for indoor air quality in the country in the line of national ambient air quality standard. The findings may also inspire the authorities to take measures for reduction of IAP by improving housing, kitchen ventilation and cook stoves. Moreover, the parameters used in this study can be utilized for large, population-based studies to identify women at a higher risk of developing CVD so that medical intervention can be taken at the formative stage of a disease.

S4.5 Production of Reactive Oxygen Species Mediated by Humic-like Substances in Atmospheric Aerosols

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Particulate matter (PM)-mediated reactive oxygen species (ROS) generation has been implicated in health effects posed by PM. A number of studies show that ambient PM and diesel exhaust PM can catalyze the generation of ROS. However, knowledge on the range of chemical species in PM responsible for ROS generation is very inadequate, due to our limited knowledge of the chemical composition of PM, especially the myriad of organic compounds that constitute the organic fraction of the PM. Humic-like substances (HULIS) are a mixture of water-extracted organic compounds from atmospheric samples that comprise of hydrophilic polycyclic ring structures or hydrocarbon side chains, and polar functional groups (e.g., hydroxyl, carboxyl, carbonyl or organosulfate groups). In this study, we use a cell-free dithiothreitol (DTT) assay to measure ROS production mediated by HULIS. The DTT assay is a chemical based redox-active assay that quantifies catalytically active redox species. DTT plays the role that NADPH or ascorbate plays in cells. The HULIS samples are isolated from aerosols collected at a rural location and a suburban location in the Pearl River Delta, China. In our experiments, ROS activities by residue metal ions in the HULIS fraction are suppressed by including a strong chelating agent in the DTT assay. Under conditions of DTT consumption not exceeding 90%, the HULIS-catalyzed oxidation of DTT follows the zero-order kinetics with respect to DTT concentration and the rate of DTT oxidation is proportional to the dose of HULIS.

The ROS activity of the aerosol HULIS, on a per unit mass basis is 2% of the ROS activity by a reference quinone compound, 1,4-naphthoquinone and exceeds that of two aquatic fulvic acids. The HULIS fraction in the ambient samples tested exhibits comparable ROS activities to the organic solvent extractable fraction, which would contain compounds such as quinones, a known organic compound class capable of catalyzing generation of ROS in cells. HULIS was found to be the major redox active constituent of the water-extractable organic fraction in PM. It is possible that the reversible redox sites in HULIS could transfer electrons from the reducing equivalents like NADPH or ascorbate in cells to O_2 and lead to continuous production of ROS accompanied with cellular aerobic respiration. However, the health effects of HULIS (so as PM) on human body may be too complex to be addressed by a single method due to its complicated constituents and the diversity of biological endpoints/mechanisms. Our work suggests that HULIS could be an active PM component in generating ROS and further work is warranted to characterize its redox properties.

S4.6 Hybrid Chemical Transport- Receptor-Geostatistical Modeling for Spatial and Temporal Source Impact Assessment in Health Studies

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Background: An integral part of air quality management is knowledge of the impact of pollutant sources on ambient concentrations of particulate matter and there is a growing trend to directly use source impacts in health studies. However, source impacts cannot be directly measured. Traditionally, observed concentrations have been utilized in source apportionment methods based on receptor-oriented modeling (RM). Several limitations are inherent in such approaches, leading to the development of a novel hybrid approach that is used to determine source impacts by combining the capabilities of RM and chemical transport modeling (CTM). The hybrid method calculates an adjustment factor (R) for the estimated impact of each source at each monitor location using observations and the results of CTM sensitivity analysis. R is a scaling factor applied to the original CTM source impacts and is obtained using a chemical mass balance approach that incorporates measurement, modeling, and emission uncertainties. Previously, R was calculated only at monitoring locations. That approach is extended to produce an R value for every grid cell in the CTM domain for source impact adjustments at locations beyond observation locations. The interpolation can also be done in time.

Methods: In this study, kriging is the primary method used to spatially interpolate R values, which are calculated using data from the Speciation Trends Network (STN). An additional analysis is completed using data from the IMPROVE monitoring network in order to determine the performance of the hybrid-kriging (HK) method. Results from each method are used to reproduce concentrations for 42 species measured by STN and 30 measured by IMPROVE. All data used were reported on 9 reporting days in January of 2004. Typically, STN and IMPROVE data are limited temporally, as the monitoring networks report data one in every three or six days, so temporal interpolation will be assessed in the future.

Results: For the spatial analysis of STN data, the mean concentrations of total $PM_{2.5}$ were 12.1, 19.3, 14.9, and 17.2 $\mu g/m^3$ for the observation and CTM, hybrid, and HK predictions, respectively. For the spatial analysis of IMPROVE data, the monthly means for total $PM_{2.5}$ concentration were 6.0, 10.9, and 12.5$\mu g/m^3$ for the observation and CTM and hybrid predictions, respectively. Trends were similar for other species.

Discussion: For STN data, the hybrid method predicted concentrations closest to observations, but HK predictions were closer to observations than CTM predictions. For IMPROVE data, CTM predictions were closer to the observations than HK predictions. It is possible that this resulted because IMPROVE results were generated based on the variability of urban-based adjustment factors and the rural variability of IMPROVE data was not incorporated.

Conclusion: The HK method was developed to provide improved source apportionment at all locations in the CTM domain. While the hybrid method may be more precise in its prediction of concentrations, the method is time consuming and is currently limited by data availability. HK significantly reduces the computing time and capacity needed to perform large-scale source apportionments.

S4.7 Quantification of source-specific contributions to ozone mortality through adjoint sensitivity analysis

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Introduction

Air pollution decision-making is streamlined if air quality impacts can be traced back to individual emission sources. Adjoint sensitivity analysis of numerical models allows for calculation of spatiotemporal influences of individual emission sources on health metrics (e.g. mortality) as a whole. In the adjoint approach, source specificity is conserved, making it a viable option for evaluating the relative importance of emission reduction policies in a straightforward manner.

Methods

In this work, we attribute short-term mortality (valuated as an overall “health benefit") in Canada and the U.S. to anthropogenic NOx and VOC emissions across North America. We integrate epidemiological data with the adjoint model to calculate derivatives of nationwide health benefits with respect to anthropogenic emissions of each species at each location.

Results

Our results show significant spatial and temporal variability in health benefit influences of NOx and VOC emissions reduction on Canada-wide and U.S. mortality. We note the largest influences on Canada come from south-eastern Ontario and Quebec, where population centres are concentrated. Health benefit derivatives for Canada reach upwards of $250K/day (NOx) and $50K/day (VOCs) for 10% reduction is emissions, and are highest just upwind of Toronto. For U.S. mortality, we observe that major American cities tend to have higher-magnitude influences due to both larger emissions and populations, with some cities showing strongly negatives benefits/dis-benefits (e.g. -$680K/day from New York for NOx).

Discussion

In calculating the influences of NOx emissions across North America on Canada and U.S. mortality, we find that long-range transport is a significant contributor to mortality in Canada but not necessarily the U.S. Our results show that a 10% reduction in all NOx emissions in the U.S. would benefit Canada by $3.8M/day, more than twice the $1.7M/day influence the same reduction in Canadian emissions would have on the U.S. For all sources, we notice significant day-to-day variability in health benefit influences related to O_3 mortality. Major Canadian cities such as Toronto and Montreal exhibit some days with negative NOx sensitivities despite having an overall positive impact (or sensitivity) for the modeling period. Major American cities generally tend to have larger variability in health benefit influences. Finally, we examine the effect of epidemiological averaging period on these derivative estimates by comparing results generated using a daily 1-hr maximum vs. 24-hr average O_3 concentration for the U.S. We observe that health benefit influences calculated using a 24-hr average O_3 concentration are significantly lower in magnitude than those calculated using a daily 1-hr maximum O_3 concentration.

Conclusions

Our work suggests that potential health benefits are vastly under-represented in the current benefit-cost analysis framework that lacks source specificity. With these results scaled to a marginal change in emissions (e.g. the benefit from a unit reduction of 1 tonne/yr), we can evaluate the benefits of emission control policies in any North American location (e.g. health benefits of public transportation systems or stationary source controls). By comparing source-specific health benefits with sectoral abatement costs, benefit-cost analysis of control options becomes a straightforward task.

S5.1 Marine submicron aerosol sources, sinks and chemical fluxes

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The flux of sea-spray aerosol has been studied previously as sea salt mass fluxes or aerosol size and number flux. Apart from one or two studies, the flux experiments have typically focused on super micron sized particles. When eddy covariance method has been introduced it allowed studying submicron particle fluxes. None of the above techniques were suitable for studying chemically resolved fluxes either, because chemical analysis typically requires long sampling time (off-line analysis) or does not have enough resolution in time (aerosol mass spectrometers) to be used with the eddy covariance system. After the discovery of a significant fraction or marine particles composed of organic matter compounds, it became ever more pressing issue of principal sources and sinks of marine organic matter. A preliminary evaluation of the principal and a pilot study of marine chemistry gradients has been performed by Ceburnis et al. (2008) to reveal primary and secondary components of marine aerosol specifically distinguishing marine organic matter components (water soluble (WSOM) and water insoluble organic matter (WIOM)). The aforementioned study revealed that the WSOM is largely produced by secondary processes while WIOM is of primary origin.

The current study was aimed at running eddy covariance system in parallel to provide essential micrometeorological parameters for calculating chemical fluxes and their relationship with wind speed. The range of chemical species studied was expanded substantially to include recently discovered organic nitrogen species (amines) in marine aerosol. The undertaken strategy allowed a complete analysis on the source and sink fluxes as a function of wind speed and oceanic biological activity and provided a quantification of both primary and secondary inorganic and organic aerosol cycling in the marine boundary layer.

The marine aerosol sources, sinks and chemical fluxes were studied over the entire year by the gradient method. The chemical fluxes of primary species, such as sea salt, WIOM and sea-spray were found to show strong power law relationship with the wind speed. The power law exponent of sea salt mass source function was 2.53 which was slightly lower than the generally considered cubic power law relationship. The flux versus wind speed relationship of WIOM was found to be even stronger than that of sea salt (4.41 versus 2.53) confirming a strong dependence on the biological activity in oceanic waters as supported by the strong organic matter enrichment dependence on chlorophyll-a concentration. The study of certain secondary species (nitrate, oxalate, MSA, WSON) was performed for the first time which showed a potential to estimate their production or removal rate in the marine boundary layer.

The boundary layer filling time by sea spray was found to be approximately 2 days. The removal rate of WSOM at the surface was found to be in the range of 1-2 days which is the important parameter considering aerosol life time and the fate of pollutants in the marine boundary layer.

S5.2 Studies of the propagation of bromine chemistry in the Arctic: from the sea ice to open leads and across the tundra

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During the BROMEX (Bromine, Ozone and Mercury Experiment) in the spring of 2012, we flew the Airborne Laboratory for atmospheric Research (ALAR) in the vicinity of Barrow, AK and around the North Slope region, while measuring temperature, 3D winds, aerosols, ozone, and BrO, using a multi-axis DOAS instrument. We were able to measure these species from the surface to 3.5km, above multi-year ice, first year ice, open leads, refrozen leads, and the snowpack over the inland North Slope tundra of Alaska. At the same time, BrO measurements were made from the surface via CIMS and MAXDOAS, at Barrow, and on both sides of an open lead. We found that, often, there was as much or more BrO over the tundra. Here we report initial results, and discuss the implications with respect to our understanding of how bromine chemistry is initiated and propagated.

S5.3 Isoprene and monoterpenes at a mixed suburban site in the Indo-Gangetic Plain: Implications for regional ozone chemistry and seasonal biosphere-atmosphere exchange

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Introduction

Annual global emission fluxes of isoprene and monoterpenes by vegetation are circa 550 Tg /yr and 100 Tg/yr, respectively. Yet the contribution to these global budgets from India is very uncertain as no in-situ measurements reporting on both diel and seasonal variability are available. In this work, we investigate the diel trends and seasonal variability of isoprene and monoterpenes from a site in the North Indo-Gangetic plain focusing on possible implications for regional ozone chemistry.

Methods

The dataset reported in this work is from surface air measurements(15m agl) performed between July-2011 to April-2012 at a suburban site (30.6794°N, 76.7289°E, 310 m asl) in the Indo-Gangetic plain covering the monsoon, post monsoon, winter and spring periods. Both isoprene and sum of monoterpenes were measured using a proton transfer reaction mass spectrometer (PTR-MS) which detects species based on the principle of soft chemical ionization. The signal due to isoprene was measured at m/z = 69 while the signal due to the sum of monoterpenes was measured at the sum of m/z =137 and m/z = 81.

Results and Discussion

During the monsoon and post monsoon, diel profiles of m/z=69 showed characteristic isoprene emission profiles with hourly average diel values increasing steadily after sunrise until late afternoon. Peak values reached levels of about 3 ppb. The timing of the peaks was different in the two seasons and advanced from 16:00 hours during the monsoon to 12:00 hours during the post monsoon, possibly due to water stress. From winter till the onset of spring, the diel profile of m/z=69 showed a bimodal profile with peaks in the early morning and evening hours but no distinct diurnal variation suggesting that emissions of isoprene are very low in this season and most of the m/z=69 signal is due to the contribution of anthropogenic VOCs like furan. The diel profile of the m/z=81 and m/z=137 signals followed the diel temperature trend with peak average hourly values of up to 2 ppb. The strongest emission was observed in the post monsoon period during the day between September 17 and October 5. Correlation with other specific tracers was also used to infer and interpret the strength of isoprene and monoterpenes.

Conclusion

The current work report a unique new dataset on the levels and seasonal trends of isoprene and monoterpenes derived from high frequency in-situ measurements of air in the Indo Gangetic plain. Peak average values of 1-3 ppb in certain seasons are comparable with values observed near forested regions of South America and Europe. When mixed with nitrogen oxides, these emissions can cause large increases in the production potential of both ozone and secondary organic aerosol suggesting that the role of isoprene and monoterpene chemistry in the Indo-Gangetic plain cannot be neglected. The results highlight the need for similar studies in other parts of the India.

S5.4 Aircraft-scale Eddy Covariance Fluxes of Biogenic Volatile Organic Compounds in California during CABERNET

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An aircraft flux study was conducted to assess BVOC emissions from California ecosystems with a focus on oak woodlands. The direct eddy covariance (vDEC approach) approach used PTR-MS onboard a CIRPAS Twin Otter aircraft during June 2011 as part of the CABERNET (California Airborne BVOC Emission Research in Natural Ecosystem Transects) project. Oaks are assumed to be the dominant isoprene emitters in this landscape, and they grow in specific elevations which makes them an ideal exploratory subject to study from an aircraft. Oak woodlands surround the polluted Central Valley. Since the distribution of oaks is relatively sparse, their LAI is not as high as that of densely forested regions in the Sierra Nevada Mountains which are dominated by conifers emitting mainly methanol, methylbutenol, and terpenes. During the flights over Oaks isoprene flux was very high, but was small over all other landscapes. Therefore, accurate species composition and aircraft derived emission factors are critical for the improvement of the accuracy of modeled predictions for isoprene and other important ozone and aerosol precursor compounds. We succeeded in measuring emissions with the largest coverage with respect to biogenic sources, so far conducted, of isoprene, MVK+MAC, methanol, monoterpenes, and MBO. In addition, acetaldehyde emission was measured from rice fields in the delta region. Isoprene emissions from agricultural crop regions and shrublands were generally low, but high methanol and monoterpenes were found above some of these regions. MEGAN and BEIGIS models are commonly used in California to predict emissions of BVOC and these direct measurements are used to improve their predictions. This has been the most precise determination of direct fluxes and emission factors from aircraft transects over vegetative areas and “race-track” profiles over homogenous and nonhomogeneous oak landscapes allowing accurate measurements at high spatial resolution (less than 2 km).

S5.5 Quantifying the uncertainty in simulating global tropospheric composition due to the variability in global emission estimates of Biogenic Volatile Organic Compounds.

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The emission of C1 to C3 organic compounds from biogenic sources is an important source of trace gases in remote regions away from urban conurbations, and needs to be better known, notwithstanding the ongoing mitigation of anthropogenic emissions. For certain organics such as ethene, biogenic emissions act as the dominant sources at global scale. In this study we examine the contribution of biogenic volatile organic compounds (BVOCs) towards global tropospheric composition using the global 3D chemistry transport model TM5 and applying the recently developed modified CB05 chemical mechanism. We find that the contribution of C1 to C3 BVOC species (excluding CH_4) is approximately a third of the cumulative effect introduced by isoprene and the monoterpenes. For tropospheric O_3 and CO they contribute ~3% and ~10% to the global burdens, respectively. By examining an ensemble of simulations which adopt different global BVOC emission inventories we determine the uncertainty introduced with respect to simulating the composition of the troposphere and find that the variability introduced is of the order of 2-5% and 10-20% for O_3 and CO, respectively. It is found that the relative uncertainty is largest in the northern hemisphere during summer, introducing regional variations in resident O_3 and CO mixing ratios of ±5% and 20-40%, respectively. Although the uncertainty in tropospheric O_3 mainly pertains to the lower troposphere, the substantial uncertainty in CO mixing ratios propagates up to the upper troposphere. This subsequently perturbs the oxidative capacity of the troposphere and influences atmospheric chemical lifetimes. It is found that the relative uncertainty is largest in the northern hemisphere during summer, introducing regional variations in resident O_3 and CO mixing ratios of ±5% and 20-40%, respectively.

S5.6 Global constraints on methanol and formic acid sources based on measurements from the Tropospheric Emission Spectrometer

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Methanol is the most abundant non-methane organic compound in the atmosphere and an important precursor of CO, HCHO, and O_3. Formic acid is one of the largest sources of acidity to the global atmosphere. New space-borne measurements of these species from TES, onboard EOS Aura, offer powerful new information for quantifying methanol and formic acid sources on a global scale. Here we apply an ensemble of recent field measurements and a 3D chemical transport model (GEOS-Chem) to evaluate the TES retrievals and interpret them in terms of the constraints they imply for the budgets of methanol and formic acid. The observed seasonal cycle of methanol in temperate regions is inconsistent with present understanding, with the timing of the seasonal peak in model emissions 1-2 months too late. The discrepancy reflects a current underestimate of emissions from new leaves, and we apply the satellite data to better quantify the seasonal change in methanol emissions for midlatitude ecosystems. Implementing this new understanding into GEOS-Chem, we carry out an adjoint inversion of the TES data to better quantify emissions from plant functional types globally.

S5.7 Characterising African & South American Biomass Burning Emissions using Total Column Measurements from the East Coast of Australia

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Bushfires release large quantities of particulates and trace gases into the atmosphere, impacting upon greenhouse warming and the chemistry of the lower atmosphere at local to global scales. Biomass burning is the second largest source of trace gases to the atmosphere (behind fossil-fuel burning) and the largest primary source of fine carbonaceous aerosols. Vegetation fires release an average of 2100 mega-tonnes of carbon to the atmosphere per year, (or approximately one quarter of the total global carbon emissions). Estimated global carbon emissions from fires during 1997-2009 show that the largest contributors are the most poorly characterised regions, namely Africa (~50% of global carbon emissions from fires), South America (~14%), Indonesia (~10%) and Australia (~7%)

Prevailing westerly winds transport smoke plumes from African and South American fires over Australia on typical time-scales on 1-2 weeks. A remote sensing Fourier transform spectrometer which is located at Wollongong (34S, 151E) therefore represents a potential untapped source of information to help characterise these fires and evaluate current estimates of the resulting carbon emissions.

We present an analysis of existing spectra from the 16 year time-series at Wollongong (combined with satellite measurements and back-trajectory analysis) that identifies and characterises transported smoke from Africa and South America. Measured total column amounts of gases emitted by biomass burning are compared to those predicted by atmospheric chemical transport models to assess whether there is evidence that total emissions from fires in Africa and South America are underestimated (or overestimated) at present.

S5.8 Global simulations of organic and inorganic nitrogen atmospheric deposition: Past and Future changes.

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Nitrogen is important nutrient that controls the productivity of terrestrial and marine ecosystems. Emissions of reactive nitrogen into the atmosphere are increasing due to human activities, affecting also nitrogen deposition to the surface. There is also growing evidence that a significant fraction of nitrogen deposition occurs in the form of organic nitrogen, although the chemical characterization of this fraction remains a challenge.

The present study uses a global chemistry transport model (TM4-ECPL) to calculate the global distribution of nitrogen deposition – accounting for both its inorganic and organic fractions in gaseous and particulate phases. TM4-ECPL accounts for all major aerosol components, for oxidants and volatile organic chemistry and for secondary organic aerosol formation. The global distributions of organic and inorganic fractions of nitrogen deposition are computed and evaluated against recent observations. Present-day simulations suggest that the global organic nitrogen cycle has a strong anthropogenic component with ~45% of the overall atmospheric source (primary and secondary) associated with anthropogenic activities. Total present-day nitrogen deposition amounts about 122 Tg-N/yr globally from which about 42% is deposited over the oceans. Organic nitrogen carried by particles is estimated to contribute by 70% to the global atmospheric soluble organic nitrogen deposition that is estimated to be about 16Tg-N/yr with an order of magnitude of uncertainty. Significant past and future changes in these depositions due to human activities are also evaluated by TM4-ECPL driven by changes in the emissions of nitrogen deposition precursors.

S6.1 Recent Multiphase Atmospheric Chemistry Investigations: HCCT-2010 and CAPRAM Developments

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We have undertaken a complex ground-based chemical aerosol-cloud interaction experiment in autumn of 2010 with international participation. The experiment idea is outlined and first results are described from chemical offline (aerosol impactor and filter samplers, one, three and fives stage cloudwater collectors) as well as from four AMSs deployed in a Lagrangian set-up, consisting of one upwind station, one in-cloud station, and one downwind station in the area of the Schmücke, Germany. During appropriate cloud events, the upwind aerosol composition serves as a reference to study cloud processes at the in-cloud site, as well as possible aerosol modifications at the downwind site. A large number of compounds was analyzed from the offline samples, including inorganic ions, carbon sum parameters, monoand dicarboxylic acids, sugars and sugar-related compounds, organic carbonyl compounds, metals and metal ions. The AMS measurements allowed for highly time-resolved insights into changes of the aerosol chemical composition during a cloud passage.

In the second part, a number of recent developments for CAPRAM are described: A coupling to MCM is underway, there is a new, more complex CAPRAM halogen module (CAPRAM HM2), and, in collaboration with LISA, Paris, a mechanism generator for organic aqueous phase chemistry (GECKO-A) was developed and is operative now. For the development of GECKO-A, a comprehensive database with kinetic data for more than 600 reactions of organic compounds with hydroxyl and nitrate radicals has been created. For missing kinetic data, suitable estimation methods such as structure-activity relationships have been evaluated and implemented in a protocol for automated mechanism construction. The generator has been used to expand the organic chemistry in CAPRAM up to C4 chemistry in the current study. Together with the MCM, a multiphase mechanism with more than 15000 reactions (over 11000 reactions in the gas phase and over 3600 in the aqueous phase) was used in the current studies. Results from these developments are described. Besides investigations of the concentration profiles of key compounds, detailed time-resolved flux analyses of the source and sink fluxes of those compounds have been performed. The study provided a broader knowledge on the production and destruction of organic compounds, such as dicarboxylic acids, in the tropospheric multiphase system.

S6.2 Heterogeneous Processes between mineral dust and gases at ambient condition

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In the strategic plan for the U.S. Climate Change Science Program (CCSP), the composition of the atmosphere, especially its particles, were identified to be one of the largest uncertainties about the impact on climate. The Earth’s atmosphere is composed of gases and particles that vary with spatial scale and with time, influencing climate, air quality, the stratospheric ozone layer, and weather. In order to understand the complicated change process on the molecular level, and the reactivity of the chemical reactive species, laboratory investigations will play a crucial role. Therefore, we have performed laboratory studies on heterogeneous reactions between various trace gas and particles at different conditions.

Photoelectron and photoionization mass spectrometer (PES-PIMS), diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS), and ion chromatography were used to investigate the heterogeneous reactions in laboratory.

Mineral dust has been by mass one of the largest sources to the global budge of airborne particulate matter. Recent modeling studies have predicted that mineral aerosol could also have a significant influence on atmospheric chemistry by promoting heterogeneous reactions. The heterogeneous reactions of low molecular weight aliphatic acids (formic acid, acetic acid, and propionic acid) on dust particles were performed at ambient condition. The adsorptions of these compounds were found to be mostly irreversible on dust particles. In addition, the impact of relative humidity was studied. The loss of atmospheric aliphatic acids due to reactive uptake on available mineral dust particles may be an important loss pathway, especially in dusty urban and desertified environments. Furthermore, the temperature at the troposphere can reach values much lower than 220K, down to 180K above the Antarctic in winter. Low temperature measurement of reaction rate constants can provide information that can be used directly in atmospheric models. The heterogeneous oxidation of SO_2 by O_3 on $CaCO_3$ particle was investigated at temperature from -43℃ to 25℃. The main products detected by DRIFTS were sulfate at all temperatures. The experiments suggested that the heterogeneous oxidation is sensitive to temperature. The amount of sulfate forming on the surface increase as the temperature decreases from 25℃ to -23℃, and decrease as the temperature from -23℃ to -43℃. Heterogenenous reaction of NO_2 on alumina at low temperatures (from 298 to 230 K) was also investigated. It was found that the relative abundance of different structures of surface nitrate species with changing temperature. Our results suggested the condition factor should be considered when the reactive uptake coefficients used in the modeling studies.

S6.3 A crucial oxidation mechanism to form sulphuric acid in VOC rich environments

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It is crucial to fully understand the atmospheric sulphuric acid (H_2SO_4) budget in order to predict aerosol formation and growth and hence predict climate change. Traditionally it is thought that H_2SO_4 is produced by OH oxidation of sulphur dioxide (SO_2), and that the sinks of H_2SO_4 in the troposphere are mainly by condensation to the particle phase. We have tested a new oxidationmechanism of SO_2 to form H_2SO_4 that will enhance H_2SO_4 production and hence aerosol formation in VOC (volatile organic compound) rich areas.The suggested mechanism is that stabilized Criegee intermediate (sCI) or its derivative can oxidise SO_2 and form H_2SO_4. For our model simulations, we use sCI from ozone (O_3) oxidation of the three tree emitted monoterpenes; alpha-pinene, beta-pinene and limonene with the rate constants $6*10^{-13}$ $cm^3 s^{-1}$, $6*10^{-13}$ $cm^3 s^{-1}$ and $8*10^{-13}$ $cm^3 s^{-1}$, respectively based on new laboratory experiments. We use the zero dimensional model MALTE-BOX with near-explicit chemistry from MCM. The simulations are done for high and low VOC concentrations at two measurement stations: SMEARII, Hyytiälä (data from HUMPPA-COPEC-10) and Hohenpeissenberg (measurement from spring2000).At both stations the differences between measured and modelled H_2SO_4 concentrations, based on the traditional production mechanism, reaches values of more than 50 % when the monoterpene concentrations are above 200 ppt. On days when the monoterpene concentrations are low, the measure/modelled H_2SO_4 concentration difference is around 10-20 %. However, if we include the sCI oxidation of SO_2, the measured/modelled ratio of H_2SO_4 approaches unity independently of the magnitude of the measured VOC concentrations. Especially in night time when OH concentrations are low (close to detection limit), the contribution of the new oxidation mechanism is crucial in the simulation of H_2SO_4 concentrations.We conclude that the traditional mechanism to form H_2SO_4 can only explain ~50% of the observed H_2SO_4 concentration in VOC rich environments, but 80-90% in VOC poor environments. This new mechanism should be included in global and regional models in order to simulate aerosol formation and growth more accurate and hence improve the prediction of climate change.

S6.4 Insights for SOA formation mechanisms from measured gas/particle partitioning of specific organic tracer compounds

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Semi-volatile and intermediate-volatility organic compounds (S/IVOCs) in both gas and particle phases were measured using a Thermal desorption Aerosol Gas chromatograph (TAG) instrument during the CALifornia at the NEXus of Air Quality and Climate (CALNEX) campaign in Bakersfield, CA from May 31st through June 27th, 2010. The gas/particle partitioning of phthalic acid, pinonaldehyde and 6, 10, 14-trimethyl-2-pentadecanone is discussed in detail to explore secondary organic aerosol (SOA) formation mechanisms. Measured fractions in the particle phase (fpart) of 6, 10, 14-trimethyl-2-pentadecanone were similar to those expected from gas/particle partitioning theory, suggesting that its partitioning is dominated by absorption processes. However, fpart of phthalic acid and pinonaldehyde were significantly higher than predicted. The formation of low-volatility products from reactions of phthalic acid with ammonia is proposed as one possible mechanism to explain the enhancement of particle-phase phthalic acid. This type of SOA formation mechanisms is expected to lead to high O/C ratios of SOA because it favors the partitioning of gas-phase organic acids into particles by forming ammonium salts. Particle-phase pinonaldehyde was present when inorganic acids were fully neutralized, showing that strong acids are not necessary for reactive uptake of pinonaldehyde on particles. It is found that fpart of pinonaldehyde increased with relative humidity (RH) without an observed increase in the aerosol acidity, suggesting the likely importance of aerosol water content in the formation of particle-phase pinonaldehyde. As the gas/particle partitioning of each of these SOA tracers represents a distinct SOA formation mechanism, this work shows that multiple tracers of SOA are needed to reconstruct SOA mass in source apportionment models.

S6.5 Comparison of Spectroscopic Signatures of Smog Chamber and Atmospheric Aerosols

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The three largest types of sources that emit vapors that form secondary organic aerosol (SOA) are fossil fuel combustion, biomass and biofuel burning, and biogenic vapor fluxes. Each type of vapor emission has a different characteristic mixture of molecular structures – from the long-chain or aromatic-ringed hydrocarbons of fossil fuels to the branched or non-aromatic rings of biogenic vapors, with biomass and biofuel burning vapors having branched, aromatic, and other ringed compounds with some oxidized groups. As these compounds are oxidized by combustion or photochemistry, they add oxygenated groups but retain some of the carbon backbone that characterized the original molecule. Spectroscopic analysis can be used to track the initial hydrocarbon backbones to distinguish different types of oxygenated SOA. In this study, we compare both NEXAFS and FTIR spectra from smog chamber studies with those collected from spectra isolated from atmospheric particles dominated by one of these three SOA source types. Atmospheric SOA associated with combustion sources is consistent with both alkane and aromatic precursors. The remote forest observations have ratios of carboxylic acid, organic hydroxyl, and non-acid carbonyl groups similar to those observed for isoprene and monoterpene chamber studies. Forest fires include biogenic emissions that produce SOA with organic components similar to isoprene and monoterpene chamber studies, also resulting in non-acid carbonyl groups in SOA.

S6.6 Change of morphology, hygroscopisity and optical properties during soot aging under ambient atmospheric conditions

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A Quasi-Ambient Atmosphere Chamber （QAC） was designed to study soot aging process under ambient atmospheric conditions. Two individual sections separated by a semipermeable membrane made up the chamber as a whole; the lower section consisting of the flow region of the chamber where ambient air was pulled through continuously, and the upper section where experimental procedures were studied. Measurement was conducted in the urban area of Houston, Texas. Mono-dispersed soot particles were introduced into the upper section in which gas concentration and oxidation condition was proved similar to that in ambient air. Significant growth in particle size and density, hygroscopic growth factor, as well as singe scattering albedo (SSA) was observed during all ten experiment days. Soot Aging normally happened between 12:00 and 16:00 when photochemical reactions are active, indicated by high O_3 concentration. However no obvious change of soot properties was found in the nighttime. The particles experienced large mass growth (about 4-fold) in some days, together with density increasing before mobility diameter. Dynamic shape factor (DSF) of soot particles decrease from 1.5 to nearly 1.0 in most experiments, suggesting the fast morphology change of soot particle under ambient atmospheric conditions. Coating with secondary aerosol components enhanced extinction of soot particles by 2-fold, taking no account of the effect of hygroscopic growth on optical properties. Large variations of the soot property change were found in different experiment days, indicating the importance of the precursor concentrations and oxidation condition on soot aging. Parameters of soot aging process in ambient air measured in this study can be used in the optical and climate models, which will have a profound implication on visibility, direct and indirect of radiate forcing.

S6.7 Stable carbon isotope ratio analysis of anhydrosugars in biomass burning source aerosol

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Compound specific isotopic measurements of levoglucosan, mannosan and galactosan were performed by employing ThermoDesorption – Two Dimensional Gas Chromatography – Isotopic Ratio Mass Spectrometry (TD-2DGC-IRMS). The d13C measured in a standard mixture showed good agreement with isotopic measurements of the bulk anhydrosugars, carried out by Elemental Analysis - Isotope Ratio Mass Spectrometry (EA-IRMS). Isotope ratios of levoglucosan, mannosan and galactosan from source samples, collected during combustion of hard wood, softwood and crop residues, were determined. d13C values of levoglucosan were found to vary between -25.6 and -22.2 ‰, being higher for the softwood samples. In this presentation, the observed d13C for levoglucosan will be compared to the carbon isotopic composition of the parent fuel holocellulose. The potential of using compound specific d13C measurements of anhydrosugars for improved source apportionment will be discussed together with results from ambient aerosol samples.

Poster Presentations, Session 1

P-1-001 Supply of Soluble Iron from Anthropogenic Sources to the Ocean

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Key words
iron ship

Iron (Fe) is an essential element for phytoplankton. Global models have been used to deduce atmospheric iron supply to the ocean, but uncertainty in the deposition flux remains large, which can influence the air-sea CO_2 fluxes and thus radiative forcing significantly. Here we demonstrate the impact of the combustion iron on soluble iron deposition in a three-dimensional aerosol chemistry transport model. The emission data sets for combustion-generated aerosols such as those from biomass and fossil fuel burnings are taken from the emission inventory. The iron from combustion sources such as biomass and fossil fuels burning is readily released into solutions in aerosols assuming constant iron solubility (i.e., the mass fraction of dissolved to total iron). In contrast, the emissions of dust are calculated on-line, based on the surface wind speed and soil wetness from the GMAO assimilated meteorological fields. Further, the iron solubility dynamically changes from that in the originally emitted dust aerosols (which is 0.45%) due to reactions with acidic species. The model response of soluble iron deposition to perturbation by combustion sources suggests that these sources may supply a potentially important source of soluble iron to the open ocean, compared to dust. These results have important implications for the parameterization of iron dissolution and highlight the necessity to improve the source chemical composition.

P-1-002 Regional Impacts of Biomass Burning Activities in South East Asia on Southern China

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Key words
biomass burning impact South East Asia

Introduction
Biomass burning is one of the significant sources of trace gases and aerosols in the troposphere. The trace gases include CO, CH_4, NMHC, NOx and O_3 which can affect the tropospheric oxidation capacity, while CH_4 and O_3 are greenhouse gases. The aerosols such as black carbon can affect the atmospheric radiative budget and thus have climate change implications. Although biomass burning activities in Southeast (SE) Asia are peaked in spring every year, their regional impacts are not

well-documented, except studies undertaken during the TRACE-P and ACE-Asia campaigns.

Methods

In the present study, the impacts of burning activities were investigated by a combination of tools: (1) Hysplit4 back-trajectory model; (2) remote sensing products from the MODIS, OMI and AERONET; and (3) a regional chemical transport model.

Results and Discussion

Evident by the back-trajectory calculation, the burning impacts on Hong Kong in 2008 were found from 2000m to 3000m above ground. The events were seldom detected near ground levels of higher latitudes (> 20°N) because of the dominant effects of the East Asian Winter Monsoon at the boundary layer (Wai and Tanner, 2010). Signature of the biomass burning has also been detected at an elevated remote site (2860m a.s.l.) in Taiwan (Wai et al., 2008). The MODIS imagery for thermal anomaly confirmed intense burnings in SE Asia such as Myanmar. High Aerosol Index was also observed within the same areas by OMI deployed in the Aura satellite platform. The volume medium radius, obtained from the measurements of AERONET sun-photometry at a remote site in Hong Kong, showed peaks in the range from 0.20 to 0.27 micron. These sizes were larger than those for aged and regional smoke in Africa and South America. The enhancement of CO due to the burning activities was captured by both MOPITT observations and the regional transport model. Transport of the burning plumes was studied in detail.

Conclusion

The biomass burning activities in SE Asia occur in every year. They influence the regional tropospheric composition which is capable to be characterized by the mentioned tools.

P-1-003 Large scale tarnsport of aerosol observed in East China Sea area

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Key words
aerosol mass spectrometer sulfate

Transport of gas and aerosol from the Asian continent is frequently observed in East China Sea area in spring. To monitor it, we have set up Aerodyne aerosol mass spectrometers (Q-AMS and ACSM) and TEOMs at three sites in west part of Japan, Fukue (128.7E, 32.8N), Fukuoka (130.4E, 33.6N) and Cape Hedo (128.3E, 26.9N) in spring time. These three sites cover the centeral part of East China Sea.

On March 11 in 2012, the high mass concentration of $PM_{2.5}$ (about 70 microgram m^{-3}) was observed at both Fukuoka and Fukue. The main species were sulfate, niterate, ammonium and organics. The mass concentration started to increase at 5am at Fukue and 8am at Fukuoka. Since the distance between Fukue and Fukuoka is about 200km, the size of air pollution is large enough to cover the both places. According to the CFORS simulation, the high sulfate region spreads from Shanghai to Sea of Japan covering the west part of Japan. About 12 hours later, the high mass concentration of $PM_{2.5}$ (about 70 microgram m^{-3}) was also observed at Cape Hedo. The main species were sulfate, ammonium and organics. Nitrate fraction was less compaered to those observed in Fukue and Fukuoka. According to the CFORS simulation, the large scale air pollution observed at Fukue was transported from northen to southern part of East China Sea. The large scale transport was occurred in the East China Sea and it was observed at three different sites in the same day.

On March 14-17, the high mass concentration of $PM_{2.5}$ was observed at three sites, too. The concentration was about 50 microgram m^{-3} at Fukuoka and Fukue and was about 40 microgram m^{-3} at Cape hedo measured by TEOM. The main species were sulfate, niterate, ammonium and organics for Fukuoka and Fukue. At Cape Hedo, sulfate, ammonium and organics were the main species though little nitrate was observed. According to the CFORS simulation, the high sulfate region spreads from Sandong to Osaka, which is 500km east to Fukuoka. Again, the large scale air pollution covers the west part of Japan. At the same time, the wind came from the Pacific Ocean at Cape Hedo, Okinawa. The relatively high sulfate region spread over the Pacific Ocean since the large scale air pollution was pushed out to the Pacific Ocean in the previous days. This is why the high mass concentration was observed at Cape Hedo though the wind came from the ocean side.

These observation and simulation show that the large scale of air pollutants were transported in East Asia in spring.

P-1-004 20 Years' aerial observations of atmospheric pollutants over the seas between Japan and Asian continent

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Key words
Long-range trans-boundary air pollution aerial observations

[Introduction] Aerial observations over the seas between Asian continent and Japan were conducted for twenty years in order to analyze the transport and transformation of log-range transported atmospheric pollutants. Covered areas are the East China Sea, the Sea of Japan, and the Yellow Sea. During this period the economic growth in East Asia, particularly in China, was remarkable and as a result emission of atmospheric pollutants increased tremendously. Here we will show the results of our analyses on the long term trend of gaseous pollutants such as SO_2 and ozone as well as ionic components of aerosols.

[Analyzed Data] The data used for the analyses were 11 data sets, i.e., those from experiments carried out in October, 1991, November, 1992, March and December, 1994, January and December, 1997, February, 1999, March, 2001, March, 2008, October, 2009, and December, 2010. Aircrafts employed were Cessna 404, and Fairchild Swearingen, and Beachcraft Kingair 200T. Atmospheric pollutants transported in lower troposphere (below 3000 m) were analyzed.

Pollutants commonly observed in 20 years experiments were ozone, SO_2, and NOx (NOy) as well as ionic species in aerosols collected with a high-volume tape sampler. Sulfate, nitrate, ammonium, and calcium were the main ionic species analyzed.

[Results and Discussion] Anthropogenic species in the gas phase (SO_2 and ozone) and aerosols (sulfate, nitrate, and ammonium hereafter referred to as SO_4, NO_3, and NH_4) were discussed in this presentation. SO_4 and NH_4 showed a similar trend. That means SO_4 and NH_4 existed in the form of ammonium sulfate and/or ammonium bisulfate. Slightly decreasing trend for SO_4 could be seen from 2001. Unfortunately, since the data between 2002 and 2007 are lacking (we carried out aerial observations over the main land China in that period), it is not very clear. However, since the emission of SO_2 in China is decreasing from 2006, our results seem in accordance with the trend of the emission.

In contrast, $NO_3$was low before December, 1997 (maximum 1.5 ug/m^3), but it is increasing after 1999 having maximum concentrations exceeding 7 ug/m^3. It seems to show an increasing trend. Emission of NOx, which is a precursor of NO_3, is increasing in China, and that can cause increase of NO_3 over the East China Sea. More clearly, the ratio of NO_3/SO_4 increases reflecting the trends of sulfate and nitrate.

Ozone was analyzed using histograms for former period (1991-1999) and latter period (2001-2010). In 1990s ozone in the range of $40<O_3$ conc.<45ppb appeared most frequently, whereas ozone in the range of $65<O_3$ conc.<70ppb appeared most frequently in 2000s. This feature was more remarkable in the lower troposphere below 1500 m. Ozone in the boundary layer is increasing because of anthropogenic emission of NOx in China.

[Conclusion] Long term trend of atmospheric pollutants over the East China Sea was discussed on the basis of aerial observation data obtained since 1991. Clear upward trend of NO_3/SO_4 ratio can be seen which indicates increase of NO_3 or decrease of SO_4 or both. Ozone at lower altitude showed clear increase.

P-1-005 Long-term, comprehensive monitoring of non-CO_2 greenhouse gases and short-lived climate forcers in Asia and Oceania using voluntary observing ships

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Key words
SLCF voluntary observing ships trend emissions Asia and Oceania

The National Institute for Environmental Studies (NIES) has been operating a long-term program for monitoring trace gases of atmospheric importance over the Pacific Ocean since 1995. The NIES Voluntary Observing Ships (NIES-VOS) program currently makes use of four commercial cargo vessels that are in regular operation in constant routes for long periods and sail over a wide area between various ports (e.g., between Japan and the United States, between Japan and Australia/New Zealand, and between Japan and southeast Asia). These routine platforms offer the advantage of systematic measurements of trace gases and aerosols, providing long-term datasets for pristine background air over the Pacific Ocean and regionally polluted air around east Asia. Ambient measurements are made by combination of continuous instruments onboard ships and flask sampling apparatus followed by laboratory analysis. We observe both long-lived greenhouse gases (LLGHGs, e.g., carbon dioxide, methane, nitrous oxide, etc) and short-lived air pollutants (or short-lived climate forcers: SLCFs, e.g., tropospheric ozone, black carbon, etc) on a continuous basis. Flask samples are analyzed for carbon dioxide, methane, nitrous oxide, halocarbons, and carbon monoxide by using gas chromatographic techniques. In addition, we recently installed cavity ringdown spectrometers for high-resolution measurement of methane and carbon dioxide to capture their highly variable features in regionally polluted air around southeast Asia (e.g., Hong Kong, Thailand, Singapore, Malaysia, Indonesia and Philippine), which is now thought to be a large source due to expanding socioeconomic activities as well as biomass burnings. Comparison between in situ measurements and flask analyses enabled us to check data quality and thus improve accuracy and precision of overall measurements. Contrasting the Japan-Australia/New Zealand and Japan-southeast Asia cruises revealed remarkable characteristics of spatial and temporal variations that reflect regional characteristics of emissions, suggesting additional sources for methane, nitrous oxides, and carbon monoxide in this tropical Asian region. We will overview long-term trends and interannual variations of multiple species at different latitudinal bands and different geographic

regions. In addition, we will highlight the impact of large-scale biomass burning events in El Nino year 2006 on trace species based on the analysis with AIRS satellite data, GFED emissions inventory, and FLEXPART model.

P-1-006 Haze Particles over a coal-burning region in the China Loess Plateau in winter: three flight missions in December 2010

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Key words
coal-burning Brown haze Flight research Transmission electron microscopy

Brown hazes have far-reaching effects on both regional and global scales, influencing climate changes and human health. During the past decades, the atmospheric air quality in whole East China was deteriorated for a long time because of the brown hazes. Regional haze layers in East China, both in large cities and rural areas, exhibit surprisingly high loadings of atmospheric pollutants. To evaluate the regional brown haze in northern China, a transmission electron microscopy (TEM) was employed to study individual haze particles in different kinds of haze episodes. We classified three kinds of brown hazes smoke-haze, industrial-urban haze, and coal-burning-haze.

Coal-burning haze: Heavy haze frequently occurs in winter over a coal-burning region, the Taiyuan Basin in northwestern China. One research flight was employed to collect aerosol particles and monitor SO_2 concentration in hazes from the ground (780 m asl) up to 4110 m during 17-18 December, 2010. Meteorological records reveal that the whole layer was stable and further divided into three layers by two shifts at about 1500 m and 3000 m. SO_2 concentration was 16 - 116 ppb with an average of 58 ppb from surface to 1500 m (Layer-1), 2 - 45 ppb with an average of 9 ppb from 1500 to 3000 m (Layer-2), and 2 - 10 ppb with an average of 4 ppb above 3000 m (Layer-3). The accumulation of SO_2 in the Layer-1 was due to the stable meteorological conditions and the strong anthropogenic emissions in addition to the basin topography. Analyses of the collected particles using a transmission electron microscope revealed the dominance of organic particles and fly ash in the Layer-1 and -2 and sulfate particles in the Layer-3. The organic aerosols frequently contained certain amounts of Si and Cl. Fly ash particles consisted of O and Si with minor Fe, Mn, Zn, Ti, Pb, As, Co, and Cr. These two types of aerosol particles are typically emitted from coal burning. Therefore, the hazes were characterized in principle by aerosols from primary emissions of coal burning, which is different from the hazes over the North China Plain where secondary sulfate particles are a dominant component.

Although regional hazes frequently occur in China, their aerosol sources are different in different areas and seasons because of various new sources following the economy developing and reformation in the whole China. These regional hazes not only cause diverse health problems in continental China but also influence the regional or global climate. Compared with other areas in the world, there are many different atmospheric chemical mechanisms or haze form in the heavy polluted region because of the extremely high fine particle loading in the atmosphere. In addition, we do not know how haze aerosols influence the cloud formation and its precipitation. Therefore, the study about the haze aerosols in upper level in the atmosphere is necessary in the future.

P-1-007 Atmospheric conditions associated with high and low summer ozone levels in the lower troposphere over the eastern mediterranean

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Key words
Ozone Troposphere Mediterranean

In order to evaluate the observed high rural ozone levels in the Eastern Mediterranean area during summertime, vertical summer ozone profiles measured in the period 1994–2008 in the framework of the MOZAIC project (Measurement of Ozone and Water Vapor by Airbus in Service Aircraft) over the Eastern Mediterranean basin (Cairo, Tel-Aviv, Heraklion, Rhodes, Antalya) were analyzed, focusing in the lower troposphere (1-5 km). At first, vertical profiles collected during extreme days with very high or very low tropospheric ozone mixing ratios have been examined. Also, the average profiles of ozone, relative humidity, carbon monoxide, vertical temperature gradient and wind speed corresponding to the 7% highest and the 7% lowest ozone mixing ratios for three height layers (1000-1500m, 1500-3000m, 3000-5000m) for Cairo and Tel-Aviv have been examined and the corresponding composite maps of geopotential heights 850 hPa, 700 hPa and 500 hPa have been plotted.

In addition analysis of backward trajectories, using the FLEXPART model, in the lower troposphere over the examined aiports has been performed for the highest and lowest ozone days in combination with the calculation of the CAPE (CIN) criteria of instability (stability) for the selected vertical profiles.

From the above analysis the main concluding remarks are the following:

The highest ozone levels in the lower troposphere (60-80 ppb) over all the examined Eastern Mediterranean airports are associated with low relative humidity, low vertical temperature gradient and low wind speed. They occur under high-pressure (anticyclonic) meteorological conditions and in particular the strengthening of the north African anticyclone and its extension towards Central Europe and the Balkans, creating large scale subsidence conditions resulting to high ozone levels in the lower troposphere and influencing the boundary layer through the process of thermal vertical mixing. These anticyclonic conditions create a northern, lower troposphere and boundary layer, flow transporting ozone and precursors from Central and Eastern Europe and/or the Balkans towards the Eastern Mediterranean.

The lowest ozone levels are associated with high relative humidity, high vertical temperature gradient and high wind speed. They are observed under low pressure (cyclonic) meteorological conditions prevailing over Eastern Europe and the Middle East creating uplifting of boundary layer air towards the lower troposphere, enhanced by the significant in the region summer thermal vertical instability and occasionally by sea-breeze cells on the coastline.

The CO levels do not differ substantially between the two situations, which shows that high differences in tropospheric ozone mixing ratios (up to 50 ppb) might exist despite the comparable primary pollution levels in both examined air masses, indicating that local photochemistry could not be the main and only responsible for the observed high differences in ozone levels between the two examined conditions and atmospheric transport seems to play a major role.

Combining the above observations, it comes out that the lower tropospheric ozone variability over the Eastern Mediterranean area is mainly controlled by the mesoscale synoptic meteorological conditions and also from the local topographical and meteorological characteristics of each site.

P-1-008 Improving global fire emission estimates using source inversion constrained by two years of GOME-2 formaldehyde columns

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Key words
fire emissions satellite formaldehyde inverse modeling

Formaldehyde columns retrieved from the Global Ozone Monitoring Experiment-2 (GOME-2) instrument onboard MetOp-A platform in 2009 and 2010 are used to constrain a source inversion scheme to derive improved global biomass burning emission estimates for non-methane volatile organic compounds (NMVOCs) precursors of formaldehyde.

The GOME-2 retrievals benefit from a new scheme including a two-step fitting procedure that strongly reduces the interferences between formaldehyde and bromine oxide spectral structures, and from a new approach to better handle the strong ozone absorption effects, improving the fit at large solar zenith angles. Further, the better Earth sampling of GOME-2 compared to its predecessors guarantees a significant noise reduction and contributes to a better identification of the emission sources.

We use the IMAGESv2 global CTM driven by the Global Fire Emissions Database (GFED) version 3 for biomass burning. The agreement between modeled and observed formaldehyde columns is minimized using the adjoint of the model in a grid-cell-based setup run at a resolution of 2x2.5 degrees. Intense fire burning in the boreal forests of northern Russia and Alaska in July 2009, and the exceptional wildfires that broke out across Russia in late July and August 2010 are well captured by the space observations of HCHO, but are found to be underestimated in GFEDv3, by up to 60% depending on the precise location. Lower NMVOC emissions from fires than estimated by the a priori emission inventory are inferred by the inversion over the Amazon basin, during the severe fire events that occurred in September 2010, the underprediction being more significant in the Brazilian state of Mato Grosso and in the Rondonia region close to the Brazil-Bolivian border. On the global scale, the inferred emissions are found to be lower than in the a priori, by 17% in 2009 and by ca. 35% in 2010. The resulting monthly biomass burning emission fluxes will be evaluated through comparisons with (i) monthly estimates of the Fire INventory from NCAR (FINN) available for 2009-2010, and (ii) CO columns from the Infrared Atmospheric Sounding Interferometer (IASI).

P-1-009 Development and Evaluation of a regional African anthropogenic emission inventory for gases and particles for 2005 and 2030

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Key words
emission inventory gas and particles fossil fuel and biofuel africa anthropogenic

A regional fossil fuel and biofuel emission inventory for gases and particulates has been developed for Africa at a resolution of 0.25° x 0.25° for the year 2005. The original database of Junker and Liousse (2008) was used after modification for updated regional fuel consumption and emission factors. Consumption data were corrected after direct inquiries conducted in Africa, including a new emitter category (i.e. two-wheel vehicles including “zemidjans”) and a new activity sector (i.e. power plants) since both were not considered in the previous emission inventory. Emission factors were measured during the 2005 AMMA campaign (Assamoi and Liousse, 2010) and combustion chamber experiments. Two prospective inventories for 2030 are derived based on this new regional inventory and two energy consumption forecasts by the Prospective Outlook on Long-term Energy Systems (POLES) model (Criqui, 2001). The first is a reference scenario (2030ref), where no emission controls beyond those achieved in 2003 are taken into account, and the second is for a "clean" scenario (2030ccc) where possible and planned policies within Kyoto protocole for emission control are assumed to be effective. A ccc* scenario has been also constructed from african specific emission control (change of two-wheel vehicle park, change of biofuel type …).
Carbon monoxide, nitrogen oxides, sulfur dioxide, black and organic carbon, and volatile organic compounds emission budgets for these new inventories will be discussed for present and future scenarios according to the different sectors (traffic, biofuel, power plant, industries), fuels and african regions. Finally, we have compared the budgets of such chemical compounds to the previous existing global dataset dealing with Africa including ACCMIP inventory.

P-1-010 On the observed response of ozone to NOx and VOC reactivity reductions in San Joaquin Valley California 1995–present

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Key words
ozone NOx and VOC emissions changes

We present a statistical approach to describe the effects of nitrogen oxide (NOx) and organic reactivity reductions on the frequency of high ozone days. We use sixteen years of observations of ozone, nitrogen oxides, and temperature at sites upwind, within, and downwind of three cities in California’s San Joaquin Valley to assess the probability of exceeding of the California 8-hour average ozone standard of 70.4 ppb at each location. We demonstrate that the comprehensive data records

in the region and the steep decreases in emissions over the last decade are sufficient to constrain the relative import of NOx and organic reactivity reductions on the frequency of violations. We show that high ozone has a large component that is due to local production, as the probability of exceeding the state standard is lowest for each city at the upwind site, increases across the city center, is highest at downwind locations, and then decreases at the receptor city to the south. We see that reductions in organic reactivity have been very effective in the central and northern regions of the San Joaquin but not in the southern portion of the Valley. We find evidence for two distinct categories of reactivity sources: one source that has decreased and dominates at moderate temperatures, and a second source that dominates at high temperatures in the southern San Joaquin and has not changed over the last twelve years. We show that NOx reductions are already effective or are poised to become so in the southern and central Valley, where violations are most frequent, as conditions in these regions have or are transitioning to NOx-limited chemistry when temperatures are hottest and high ozone most probable.

P-1-011 Space based inverse modeling of anthropogenic and natural emissions of nitrogen oxides over China and systematic evaluation of effects of model uncertainties

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Key words

inverse modeling of NOx emissions model uncertainties anthropogenic and natural source attribution

Nitrogen oxides ($NOx \equiv NO + NO_2$) are important atmospheric constituents affecting the tropospheric chemistry and climate. They are emitted both from anthropogenic and from natural (soil, lightning, biomass burning, etc.) sources, and China has become a major region of increasing importance for anthropogenic sources.

Here satellite remote sensing for the vertical column densities (VCDs) of tropospheric nitrogen dioxide (NO_2) is used to estimate anthropogenic, lightning and soil emissions of NOx over East China. A multi-step regression approach is employed to separate different emission sources for 2006, taking advantage of their different seasonality. The global chemical transport model (CTM) GEOS-Chem is used to establish the relationship between VCDs of NO_2 and emissions of NOx for individual sources. The inverse estimate suggests annual budgets of about 7.1 TgN (39%), 0.21 TgN (61%), and 0.38 TgN (65%) for the a posteriori anthropogenic, lightning and soil emissions, respectively, about 18-23% higher than the respective a priori values. The enhancements in anthropogenic emissions are largest in cities and areas with extensive use of coal, particularly in the north in winter, as evident on the high-resolution grid. Derived soil emissions are consistent with recent bottom-up estimates. They are less than 6% of anthropogenic emissions annually,

increasing to about 13% for July. Derived lightning emissions are about 3% of anthropogenic emissions annually and about 10% in July. Overall, anthropogenic emissions are found to be the dominant source of NOx over East China with important implications for nitrogen control.

For the first time, a systematic evaluation is conducted to quantify uncertainties in various aspects of model meteorology and chemistry affecting emission inversion for East China. Modifications accounting for quantified errors in 10 selected meteorological/chemical parameters increase the NO_2 columns in most areas with an average positive impact of 22% in July and 10% in January, suggesting a possible systematic model bias with important implications for the inverse modeling. They however cannot account for the large model underestimates in extremely polluted areas as compared to satellite retrievals, likely pointing to underestimates of the a priori emission inventory in these places. Post-model modifications also have large impacts on surface ozone concentrations with the peak values in July over North China decreasing by about 15 ppb. Individually, modification for the uptake of the hydroperoxyl radical on aerosols has the largest impact for both NO_2 and ozone, followed by various other parameters important for some species in some seasons. It is thus evident that model uncertainties need to be better understood for emission inversion and other applications.

P-1-012 GlobEmission

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Key words
top-down emissions global and regional inventories NMVOC, NOx,, SO_2, CO

Emissions of pollutants to the ambient environment are at the origin of atmospheric pollution issues. Emission inventories provide important information on magnitude, type of activity, time evolution and the spatial coverage of the estimated emissions. These inventories are developed for use in scientific applications as input in urban, regional, continental or global scale models, and for use by the policy makers in order to evaluate progress towards emission abatement measures, and decide on future strategies.

Within the GlobEmission project (part of the Data User Element programme of ESA) emission estimates will be developed from satellite observations of air constituents. The main advantage of these emission estimates are the spatial consistency, high temporal resolution and the rapid availability of these estimates to the user. Based on the requirements of several dedicated users the following services will be developed:

- Global biogenic emissions
- Regional emissions of nitrogen oxides and sulfur dioxide over China, South Africa, and India
- European emission estimates of nitrogen oxides, sulfur dioxide and carbon monoxide
- Regional aerosol-related emissions
- Global emissions from forest fires

P-1-013 Evaluation of surface emissions of atmospheric pollutants at the global and regional scales

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Key words

surface emissions atmospheric pollutants

An accurate knowledge of the spatial and temporal distribution of surface emissions and of their evolution is essential to address air quality and climate change issues. Over the past few years, several inventories providing the distributions of surface emissions of different chemical compounds were developed, at both the global and regional scales. We will review the most recent developments in surface emissions inventories, and focus more particularly on the ACCMIP, MACCity, and EDGAR 4.2 global emissions datasets, and on the TNO inventory for Europe, which covers the 2003-2007 time period. We will also consider different set of scenarios, such as the RCPs or the GEA future scenarios,

or other available scenarios.

The quality of emissions inventories is very difficult to assess, since the methodology, input data and assumptions vary strongly between the inventories. This presentation will focus on anthropogenic emissions. We will discuss an evaluation of emissions distributions for the 1980-2010 period, for the following chemical compounds: CH_4, CO, NOx, SO_2, black and organic carbon, total VOCs and ammonia. The consistency between global and regional datasets for Western and Central Europe, the USA and Canada, South America, Africa, China, India and Oceania will be discussed.

This analysis will help identify the species and regions for which a high degree of consistency exist on emission levels and their trends during the past three decades, and the species and regions for which large uncertainties remain.

P-1-014 Uncertainty associated with global carbon dioxide emissions from fossil fuel consumption

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Key words
carbon dioxide uncertainty

A new evaluation of the uncertainty associated with the global total of carbon dioxide emissions from fossil fuel consumption is based upon approaching the data from a different perspective than that used by Marland and Rotty (1984, Tellus 36B: 232-261). This new evaluation is based upon quantifying the qualitative national error classes shown in Andres et al. (1996, GBC 10:419-429). The national error classes grouped countries by relative data quality.

Global total emissions for the years 1950 to 2010 were calculated from the sum of emissions from individual nations plus an additional term which accounts for emissions not tabulated by each nation (e.g., bunker fuels, Andres et al., 2012, Biogeosci. (in review)). The uncertainty on this global total was then calculated from square root [summation((emissions*uncertainty)^2)]
where the summation is done over all nations. The result of this calculation is the mass of uncertain emissions in a given year. Repeating this calculation gives an uncertainty on global fossil fuel emissions of 2.6 to 4.8% (2 sigma) for the years 1950 to 2010.

To examine an implication of this new uncertainty estimate, it is propagated into the rest of the global carbon cycle. For simplicity, this uncertainty is accommodated entirely within the global terrestrial biosphere flux. The resulting conclusion is a global terrestrial biosphere flux that may act as a source to or sink from the atmosphere.

P-1-015 Satellite Observations of Tropospheric Ammonia and Carbon Monoxide: Global Distributions and Correlations and Comparisons to Model Simulations

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Key words
satellite observations ammonia and carbon monoxide

The Tropospheric Emission Spectrometer (TES) instrument on NASA Aura satellite launched in 2004 provides global observations of tropospheric species that are highly relevant to studies of air quality, atmospheric chemistry and transport. We present TES observed seasonal and global distributions of ammonia (NH_3) and carbon monoxide (CO). These two species are primary pollutants emitted to the Earth's atmosphere from both common and distinct sources associated with human activities. They are precursors of tropospheric aerosol formation and ozone production. We also examine the distributions and correlations of NH_3 and CO from GEOS-Chem model simulations. The comparisons between satellite observations and model resultsare used to help demonstrate how well the global and seasonal pollutant sourcesare prescribed in the model. Performing the retrieval-model comparisons and distinguishing theinformation gained in satellite species retrievals from the a priori knowledge are challenging tasks. We will describe the characteristics of TES NH_3 and CO retrievalsand the method that we applied for TES and model data analyses in order to illustrate these challenges.

P-1-016 Chemical composition of precipitation and sources of acid rain in Jiuzhaigou Nature Reserve, eastern rim of Qinghai-Tibet Plateau

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Key words
HYSPLIT acid rain nitrogen deposition acid neutralization

Acid rain, which may cause acidification and nutrient enrichment in water environment, has become a serious problem in China since 1980s due to the huge emission of SO_2 and the increasing emission of NOx. Located in the eastern rim of Qinghai-Tibet Plateau, Jiuzhaigou is famous for its spectacular lakes, shoals and travertine landscapes. However, acid rain may lead to the travertine loss and increase in algae biomass that observed in Jiuzhaigou's water environments in recent decade. Thus, we measured the chemical composition of precipitation and determined possible sources of the acids in precipitation. The results indicate that although the acidity is well buffered by alkaline ions most of time, Jiuzhaigou can be suffered from acid rain during the wet season. SO_4^{2-} is the major source of acidity, reflecting the intensive use of coal combustion in China. Local limestone weathering is an important source of alkaline dust in Jiuzhaigou air, resulting in Ca^{2+} had the highest acidity neutralization capacity in the precipitation. If all the SO_4^{2-} and NO_3^- in Jiuzhaigou precipitation were assumed to occur as free acids, the corresponding pH (Estimated pH) would be 3.08~4.43, similar to that in Xi'an and Chongqing, but significantly higher than that in Yellowstone National Park. Therefore, we believe that Jiuzhaigou is suffering from acid pollutants transported from other regions. HYSPLIT backward-trajectories demonstrate that some cities in western China and northern India are the possible sources of acid pollutants. In order to better understand the impacts of acid rain on Jiuzhaigou's water environments, additional studies are needed.

P-1-017 Tropospheric Ozone Variations over the Northern India: Balloon-borne and Surface-based Measurements

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Key words

Ozonesonde South Asia Trace Gases

Tropospheric ozone is a key trace gas in the context of air quality, atmospheric chemistry and climate change. However, our understanding of its role in aforementioned atmospheric processes and its budget is limited by lack of observations with sufficient spatial and temporal density. The measurements are sparse over the Asian regions, especially over South Asia where rapidly increasing anthropogenic emissions in conjunction with higher photochemical activity are anticipated to enhance tropospheric ozone levels in the near future. In the above perspective, surface ozone measurements are initiated at two sites, Pantnagar (79.5oE, 29.0oN, 231 m amsl) and Dehradun (78.1oE, 30.3oN, 640 m amsl), located in the Indo-Gangetic Plain (IGP) region in 2009, where higher levels of ozone and related species have been seen in different satellite-based observations. Surface ozone measurements are based on UV light absorption technique. In addition, weekly balloon flights are being conducted to measure vertical distributions of ozone (EN-SCI Ozonesonde) and meteorological parameters (Imet-1 Radiosonde) at a high altitude site Nainital (79.5oE, 29.4oN, 1958 m amsl) in the central Himalayas since January 2011.

Ozone mixing ratios at Pantnagar and Dehradun show daytime build-up with levels sometimes as high as 100 ppbv. Ozone seasonal variations at these sites exhibit distinct spring maxima and lower values during summer/monsoon and winter. Ozone seasonality over IGP region is different from the southern India. The observed ozone seasonality shows good agreement with variations in meteorological parameters and satellite observations of tropospheric NO_2 and CO (681 hPa) over this region. Daytime ozone levels at Nainital show strong positive correlation with the nearby IGP site (Pantnagar) in spring ($r^2 = 0.96$ in May) suggesting transport of pollution from IGP to the cleaner Himalayas via boundary layer processes. Chemical box model simulations indicate that ozone production is NOx-limited in the IGP region. 3-monthly AOT40 index (ppbv*hr) is significantly higher during spring months (9756 at Pantnagar and 16289 at Dehradun), indicating threat for vegetations in this region.

Analysis of balloon-borne observations shows a strong seasonal cycle in the lower tropospheric ozone with highest values during spring (~100 ppbv in May) and lowest during summer monsoon (20-40 ppbv) which is consistent with surface observations over this region. Springtime biomass burning over the northern India is estimated to enhance ozone levels in lower-middle troposphere by 20-30 ppbv. Elevated ozone levels (~120 ppbv) are observed in the middle-upper troposphere along with very high wind speed (~50 m/s) indicating the role of dynamics in bringing ozone rich air above Nainital. Occasionally, signatures of ozone downward transport have also been observed in winter-spring months and are corroborated with enhancements in potential vorticity (WRF simulated) and reductions in water vapor (TES). Tropopause height estimated from radiosonde observations varies between 16 and 18 km (~100 hPa), and agrees well with the AIRS satellite data. Relative Humidity in lower-middle troposphere is highest

(80-100 %) during summer/monsoon. Further analysis of these measurements using a regional chemistry transport model (WRF-Chem), back air trajectories and satellite data is under progress and will be presented.

P-1-018 Ozone production efficiency by air pollutants transported from East Asia

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Key words
Ozone Long-range transport East Asia

Emissions of various air pollutants increase in East Asia due to the recent economic progress, so that high concentrations of ozone are observed frequently in Western Japan in spring. We have been observing ozone and its precursors at Fukue Island, Nagasaki, Japan and evaluated photochemical ozone production for the transport process from the Asian continent in terms of the observation. We observed total odd nitrogen species (NOy), total nitrate (TN), NOx (= NO and NO_2), CO, ozone and non-methane hydrocarbons (NMHCs). NOy was observed by a molybdenum catalyst followed by NO-O_3 chemiluminescent detection. TN was measured by the scrubber difference / NO-O_3 chemiluminescence method. NOx was measured by the near-UV LED photolytic converter recently developed by ourselves, followed by ozone chemiluminescent detector. NOx concentrations can be observed continuously using the photolytic converter. Indeed, the conversion efficiency of NO_2 to NO keeps the constant value (about 70%) since March 2010. CO, O_3 and NMHCs were observed using a nondispersive infrared photometer, UV absorption and a gas chromatograph / frame ionization detector, respectively.

We evaluated photochemical ozone production by using the "ozone production efficiency" (OPE). OPE is defined as the number of molecules of oxidant ($Ox = O_3 + NO_2$) produced photochemically when a molecule of NOx is oxidized. Actually, OPE is calculated by the ratio of the Ox to NOz (= NOy - NOx) production rates. In this abstract, the results between May 19th and 20th, 2010, were described. The

relationship between Ox and NOz is as follows. When the NOz concentration is low (less than 4 ppbv in this case), the Ox concentration increased involving rising the NOz concentration. On the other hand, the increase of the Ox concentration peaked out when the NOz concentration is high (more than 4 ppbv in this case). The increase of the NMHCs concentration became moderate and the NMHCs/NOy ratio when the NOz concentration is high. These indicate that the OPE decreases in the high-NOz region. These results can be explained as follows. The photochemical ozone production is activated by increasing the NOx concentrations when the NOx concentration during the transport process is low. On the other hand, the formation reaction of nitric acid (i.e. $OH + NO_2 + M \rightarrow HNO_3 + M$) becomes predominant and the photochemical ozone production is inhibited whtn the NOx concentration is high.

P-1-019 The study of cloud condensation nuclei activity for suburban aerosols

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Key words
cloud condensation nuclei activation diameter

The physical chemical properties of aerosols play a significant role affecting radiative energy budgets and the activity of cloud condensation nuclei (CCN). In this study, how the suburban aerosols affecting the cloud formation was investigated over Northern Taiwan for August 2011. The CCN concentration was monitored using a thermal-gradient diffusion chamber (cloud condensation nuclei counter, CCNc, DMT) at supersaturation (SS) ratios ranging from 0.1% to 1%, whereas the aerosol size distribution and condensation nuclei (CN) concentration were measured simultaneously using a Scanning Mobility Particle Sizer (SMPS, TSI) and a condensation particles counter (CPC, TSI), respectively. Both CN and CCN concentrations show a distinct diurnal pattern peaked around noontime, while our previous study in Taipei city was peaked in the early morning and late afternoon. The new particle formation was observed as the sun radiation is high enough, which might be associated to the oxidation or photolysis of volatile organic compounds (VOCs) are mainly emitted by terrestrial vegetation. The major abundant VOCs emitted by plants are isoprene and other plant derivatives, such as monoterpenes and sesquiterpenes which are reactive to atmospheric oxidants such as light, ozone and OH radicals. Secondary organic aerosols (SOA) contributed through the photooxidation of VOCs, which can change the physical chemical properties of aerosols to turn into CCN. With further activation diameter analysis, the results indicate that the hygroscopic material during the noontime has the lowest fraction. The impact of ambient aerosols through aging processes to CCN activation between urban and suburban regions will be discussed in this presentation.

P-1-020 Measurements of atmospheric VOCs by SIFT-MS at Wollongong, NSW, Australia

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Key words
trace gases SIFT-MS boundary layer

Volatile organic compounds (VOCs) are important atmospheric species that play a role in both tropospheric ozone (O_3) formation and secondary organic aerosol (SOA) production, and as such have considerable climate implications, especially in terms of radiation and climate forcing. Tropospheric O_3 and SOA also play crucial roles in tropospheric chemistry as well as in human, animal and plant health. Currently, our understanding of aerosol formation and of changing tropospheric ozone concentrations over Australia is limited, in part, by a lack of measurements of typical concentrations of a number of VOCs.

A Selected Ion Flow Tube Mass Spectrometer (SIFT-MS) has recently been acquired by the University of Wollongong and commissioned for measurements of VOCs in ambient air with the aims of characterising the VOC composition of the boundary layer at Wollongong as well as some of the important VOC sources in the area.

We present our first measurements of selected atmospheric VOCs in the boundary layer at Wollongong, NSW (34.106 S, 150.879 E), as well as emission ratios for several trace gases emitted in smoke from prescribed forest fires. These measurements contribute to improve our knowledge of tropospheric composition.

P-1-021 Global monitoring and forecasting of reactive gases in the MACC project

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Key words
MACC project carbon monoxide forecast reanalysis

The EU F7 project MACC (Monitoring Atmospheric Chemistry and Climate) aims to develop an operational system for analysis and monitoring the global atmospheric composition. Within the sub-project on global reactive gases, the chemistry transport models MOZART and TM5 are coupled to the integrated forecasting system (IFS) of the European Centre for Medium-range Weather Forecasts (ECMWF) in order to provide daily analyses and forecasts and reanalysis of various atmospheric constituents. In this study we discuss the performance of the MACC system based on comparison with several datasets. The performance of the forecasting system is analysed through comparison with MOPITT and IASI CO observations. The seasonal variability of CO over the past decade is investigated using MOPITT data and NOAA/GMD ground based measurements. The goal of this study is also to investigate the changes in atmospheric chemical composition due to anthropogenic and biomass burning emissions in the northern mid-latitudes. We focus more particularly on the underestimation of CO concentrations in the northern hemisphere during wintertime noticed from global model studies. For this purpose, an analysis is performed over several regions including industrialized areas such as Europe, USA and Asia. MOZART off-line simulations performed within MACC allows to investigate the impact of assimilation of satellite CO data on the performance of the MACC system. Sensitivity of the system to changes in anthropogenic emissions will also be addressed.

P-1-022 Impact of geoengineered aerosols on stratospheric chemistry

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Key words
geoengineering solar radiation management stratospheric particle injection

The deliberate injection of aerosol into the stratosphere is a solar radiation management scheme that would cool the Earth's surface by reflecting solar radiation back to space. Such a scheme is believed to be both affordable and have a high effectiveness when compared to other geoengineering schemes (Shepherd et al., 2009). The lower stratosphere already contains a sulfate aerosol layer, produced from both biogenic and volcanic sources, which has previously been shown to have a global cooling effect.

Most research has focused on the deliberate injection of sulfate aerosol (or precursors: SO_2 and H_2S) into the stratosphere because of the natural analogue. However, the use of aerosol material other than sulfate could be cheaper to produce and be more effective at scattering radiation by virtue of having a greater refractive index. The aerosol could also be designed to have lower absorption characteristics, and would therefore result in a smaller warming of the stratosphere. In addition to containing a sulfate aerosol layer, the lower stratosphere also contains the ozone layer which is crucial for protecting the Earth's biosphere from harmful UV radiation. In addition, the ozone layer also is a key component of the coupled atmospheric chemistry system, impacting temperatures and dynamics. International regulations are already in place to protect the ozone layer from depletion by man-made halocarbons. Therefore the deliberate injection of aerosols into the stratosphere must be assessed with respect to the possible interactions between the injected aerosol and the ozone layer.

This talk will review the possible effects of the use of geoengineered aerosol (sulfate or not) and will provide a framework with which to assess the suitability of different aerosol candidates for use in geoengineering (Pope et al., 2012). In addition I will discuss laboratory work, being carried out in the Universities of Birmingham and Cambridge, and at the Rutherford Appleton Laboratory, to investigate the chemical reactivity of non-sulfate aerosol. In particular two techniques are utilized: the laser tweezers technique is used to probe the changing chemistry of the aerosol bulk phase after interaction with gas phase species. An aerosol flow tube (AFT) with gas phase species detection via chemical ionization mass spectrometry (CIMS) is used to investigate the uptake coefficients of different gas species upon the particle surface (e.g. Pradhan et al., 2010).

Shepherd, J. (2009) Report: Geoengineering the climate; science governance and uncertainty, The Royal Society.

Pope et al. (2012) Nature Climate Change, Accepted.

Pradhan, M. et al., (2010) Environmental Science and Technology, 44(4), 1360.

P-1-023 Interannual variability in a global to regional nested modeling system

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Key words

interannual variability global to regional nesting

There is increasing awareness that air pollution as a trans-continental problem. Several estimates have been made quantifying the trans-continental contributions to air pollution and its consequences for health and the environment. Most of the estimates are based on single year calculations, and with relatively coarse global models.

Global scale calculations with the EMEP/MSC-W model with a 1 x 1 degree resolution is made estimating the trans-continental contributions to air pollution from major pollution regions such as North America, and East Asia, mainly focusing on the effects on Europe. Calculations are made for the meteorological years 2008 - 2010, to get an estimate of the potential for inter-annual variability. 2009 was a moderate el Nino year, and 2010 was a la Nina year. In year 2010 there were also major forest fire

events in Russia. The global model calculations are nested to the EMEP/MSC-W model on a regional scale with a resolution of 0.2 x 0.2 degrees. Both the global and regional model results are validated against measurements, and the effects of trans continental air pollution arecompared for the global and regional model results. By using the same model in both the global and the regional scale calculations, any differences can be attributed purely to model resolution. The role of boundary conditions, meteorology and model resolution for air quality assessment is discussed.

P-1-024 Modelling methane in the Arctic: emissions, seasonality and trends

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Key words
methane arctic

The Arctic is a major source of atmospheric methane, with both natural and anthropogenic emissions. Arctic methane has the potential to be important globally, changing radiative forcing and atmospheric oxidizing capacity. Both palaeo-records and present-day studies suggest that some Arctic sources, such as wetlands and methane hydrates, show strong positive feedbacks that may lead to enhancement of climate warming. It is important that Arctic greenhouse gas sources are quantified, by strength, geographic location, character (e.g. wetland, gasfield, clathrate), and by temporal variation (summer, winter, day, night), and their vulnerability to change be assessed. An integrated programme of measurements and modelling, 'Methane and other greenhouse gases in the Arctic: Measurements, process studies and Modelling' (MAMM), has been funded to address these issues as part of the NERC Arctic programme.

Here we present results from initial MAMM global model simulations analyzing the impact of different sources and sinks on the Arctic methane budget. The simulations include a set of methane tracers coloured by source, $^{13}C\text{-}CH_4$, and a simple chemistry scheme including reaction with OH in the troposphere and Cl in the marine boundary layer. Including a $^{13}C\text{-}CH_4$ tracer provides a further constraint

on the source mix in the model, distinguishing between isotopically light and heavy sources (e.g. wetlands and gas fields). We focus on using variations in CH_4 and ^{13}C-CH_4 , and the coloured methane source tracers to determine the causes of seasonal and recent interannual variations in Arctic methane.

P-1-025 Short-lived Atmospheric Halogens: measurements, emissions and transformations – a data/modelling study

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Key words
Short-lived halogens Emission estimates

We discuss the use of halocarbon data, collected at surface sites in SE Asia, in determining emission source strengths for short-lived gases. We report a geographically expanding halocarbon data set ($CHBr_3$, CH_2Br_2, etc), near-continuous since 2008, based on purpose-built, autonomous gas chromatographs. Using a CTM and a Lagrangian air parcel trajectory model, we have used the data to determine bromoform emissions; the new data lead to a downward revision of our previous bromoform emission estimates. The measurements show good agreement with the revised emissions when used in the pTOMCAT CTM. The emissions, and ocean and snow sources of inorganic bromine, have now been included in a troposphere-stratosphere CCM, whose preliminary results with a whole-atmosphere chemistry scheme are reported. Using the models, we discuss the significance of the measurements for composition structure and variability in the TTL.

P-1-026 How would external solar radiation management affect meteorological and composition variability of the winter time stratosphere?

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Key words
geoengineering stratospheric structure and composition

Solar radiation management (SRM) has been proposed as a means to mitigate the surface warming that arises from increasing concentrations of long-lived

greenhouse gases. However, a SRM strategy based on reducing surface global mean temperature does not ensure that the atmospheric state at regional scales or in the upper atmosphere will be necessarily close to the state before greenhouse gases started to increase. We have investigated three model integrations of a coupled atmosphere-ocean chemistry-climate model (UMUKCA) following the GeoMIP recommended experimental procedure: a pre-industrial control run, a four times CO_2 run and a run with four times CO_2 mitigated by decreased solar irradiance. Firstly, we demonstrate the changes of the stratospheric winter vortex dynamics and ozone in the northern hemisphere when CO_2 is increased. Secondly, we investigate the extent to which external solar radiation management (e.g. space mirrors) affects those changes. We explore changes in troposphere-stratosphere interactions and that role of ozone feedbacks. These runs inform a general understanding of ozone-dynamics feedbacks.

P-1-027 Long term measurements of sulfur dioxide, nitrogen dioxide, ammonia, nitric acid and ozone in Africa using passive samplers

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Key words
atmospheric deposition gases concentrations passive samplers dry deposition

Atmospheric deposition of emitted chemical species and secondary products to the earth's surface through wet and dry processes plays an important role in controlling the concentrations of pollutants present in the troposphere. Deposited species can provide essential nutrients to ecosystems or can cause disturbances. The chemical content of atmospheric deposition is the signature of several interacting physical and chemical mechanisms such as: emission and source amplitude; transport in and dynamics of the atmosphere; atmospheric chemical reactions; and removal processes. The study of deposition thus allows for tracing the temporal and spatial evolution of atmospheric chemistry and is a pertinent indicator for evaluating natural and anthropogenic influences.

The international IGBP/IGAC/DEBITS (Deposition of Biogeochemically Important Trace Species) program was initiated in 1994 in Africa. IDAF (IGAC/DEBITS/AFrica) is a co-ordinated network of observing stations and related facilities whose purpose and long term goals are to provide data on changes in the chemical composition and related physical characteristics of the background atmosphere on the African continent. The main IDAF/DEBITS objectives deal

with the study of wet and dry atmospheric deposition processes, to improve understanding of the behaviour of the atmosphere and its interactions with the oceans and the biosphere. The scientific activities of DEBITS are mainly based on long term quality-controlled measurements of precipitation chemistry to quantify wet deposition, as well as aerosol and gas concentrations to estimate dry deposition fluxes and are also supported by the WMO/GAW initiative. A major challenge has been to accomplish long term monitoring activities for each principal climatic zone and each major biome in Africa. Through the IDAF/DEBITS programme, a measurement network has been successfully established and maintained since 1994.

In this contribution, results of the gaseous measurements performed from 1995 to 2010 in the IDAF network at 10 regional sites: 7 in West and Central Africa and 3 in South Africa will be reported. The monthly mean gaseous concentrations of NO_2, HNO_3, NH_3, SO_2, and O_3, were measured using passive samplers and we report the major findings in the changes in surface gaseous chemical composition over the 15 years period. The mean annual concentrations, seasonal and inter annual variability according a latitudinal transect of ecosystems in West and Central Africa and in South African (3 sites) will be discussed. It is evident from the results at most of the sites that seasonal and strong inter annual variations occur. In South Africa, the influence of environmental pressures due to an increase in population density and industrialization is emphasized. In West and Central Africa, the inter-annual variability does not exhibit strong trends and reflects more of the variations of anthropogenic (e.g. biomass burning) and natural sources (e.g. biogeogenic emissions) of the region. Anthropogenic influences are most evident in the developing regions due to the rapid increase in population growth and associated energy demands. The comparison of the results between the different sites clearly reflects the anthropogenic and natural influences on atmospheric composition at a continental scale.

P-1-028 Long term trends in the chemical composition of precipitation measured at DEBITS sites in Africa

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Key words
atmospheric deposition precipitation chemistry wet-only samplers wet deposition acidity

Wet and dry deposition of chemical species to the earth's surface plays an essential role in controlling the concentration of gases and aerosols in the troposphere. Considering the importance of the tropics in atmospheric chemistry and that limited information on atmospheric deposition was available, DEBITS (Deposition of Biogeochemically Important Trace Species) an IGAC/IGBP « Core Project », was designed to study wet- and dry- atmospheric deposition in tropical regions.

The IDAF (IGAC/DEBITS/Africa) project has been initiated in 1994 to cover the African continent. The IDAF network presently consists of ten atmospheric monitoring stations in regions representative of the three main Africa ecosystems: i.e., dry savannas, wet savannas, and forests in tropical African. The scientific activities of IDAF are mainly based upon long term high-standard measurement of precipitation chemistry to quantify wet deposition, as well as aerosol and gas concentrations to estimate dry deposition fluxes. IDAF adhere to protocols, guidelines and quality control processes of the WMO/GAW program by which it is also supported.

This paper presents an overview of results over a decade period on precipitation chemistry collected from all the IDAF sites with automatic wet-only samplers. Inorganic (Ca^{2+}, Mg^{2+}, Na^+, K^+, NH_4^+, Cl^-, SO_4^{2-}, NO_3^-) and organic ($HCOO^-$, $CH3COO^-$) ions content were determined using Ion Chromatography, as well as pH and Conductivity. The results presented are abridged from a comprehensive global precipitation assessment review being prepared by the Science Advisory Group on Precipitation of the World Meteorological Organization for publication during 2012.

The objectives are: (1) to determine the mean annual and seasonal precipitation chemistry and wet deposition fluxes characteristic at the different African ecosystems; (2) to examine the influence of atmospheric gases and particles sources on the precipitation chemical content and the associated deposition of chemical species and (3) to identify the relative contribution of natural versus anthropogenic sources.

In West and Central Africa, the precipitation chemistry is influenced by four main sources: natural biogenic emissions from savanna soils (NOx and NH_3), biomass burning (savanna and domestic fires), terrigeneous particles emissions from dry savanna soils, and marine compounds embedded in the summer monsoon. The inter-annual variability of the weighted volume mean concentration of chemical species linked with wet deposition fluctuates by ~20% over the period. Nitrogenous compounds (Ammonium and Nitrate) are found to be high in remote West and Central African sites comparable to South African dry savannas. In South African sites, in addition to the previously mentioned sources, wet deposition fluxes are also influenced by SOx and NOx industrial emissions from the

Highveld. The acidity in dry savannas in western Africa is low and indicates the strong alkaline nature of the precipitation in this region whereas the acidity of precipitation increased and is rather high in industrialized regions of southern Africa. The results prove that the characterization of rainwater helps to evaluate the influence of different sources and enhance the understanding and identification of potential impacts on ecosystems.

P-1-029 Atmospheric Nitrogen Deposition Budget in Africa

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Key words
atmospheric deposition nitrogen wet and dry deposition precipitation and aerosols chemistry gases concentrations

The international IGBP/IGAC/DEBITS (Deposition of Biogeochemically Important Trace Species) program was initiated in 1994 in Africa. IDAF (IGAC/DEBITS/AFrica) is a co-ordinated network of observing stations and related facilities whose purpose and long term goals are to provide data on changes in the chemical composition in Africa. The scientific activities of DEBITS/IDAF are mainly based on high quality measurements of atmospheric chemical data (gaseous, precipitation and aerosols chemical composition) on the basis of a multi-year monitoring performed at 10 measurement sites representative of major African ecosystems. The objectives of the programme are to study wet and dry deposition fluxes, to identify the relative contribution of natural and anthropogenic sources and factors regulating these fluxes.

This work present a first estimation of the atmospheric nitrogen deposition budget in Africa based on experimental measurements over a 10 year period. To estimate atmospheric nitrogen deposition fluxes, including both wet and dry processes, we compiled the IDAF nitrogen data (gas, particles, rain) obtained from the measurement network for the period 1997-2007. In western and central Africa, we studied a latitudinal transect from dry savannas to forested ecosystems. Results will be compared to South African IDAF sites representative of a rural and an

industrialized semi-arid savanna. Presenting the different components of the nitrogen atmospheric deposition on these sites, i.e., dry deposition in gaseous (NO_2, NH_3, HNO_3) and particulate forms (pNH_4^+, pNO^{3-}) associated with wet deposition (NH_4^+, NO^{3-}), this study allows to estimate (i) the relative contribution of dry and wet deposition processes and (ii) the total nitrogen deposition at regional representatives sites in Africa.

The nitrogen atmospheric deposition presented for all the IDAF sites of the African continent range from 6 to 13 kgN.ha-1.yr-1. Sites from dry savanna in South Africa and West Africa have similar values (around 6-9 kgN.ha-1.yr-1) which are found in the lower part of the range. Wet zones from savanna and forests give values in the upper range (10 to 13 kgN.ha-1.yr-1). The Amersfoort site in South Africa, representative of an industrially impacted savanna region gives values also in the upper range (15 kgN.ha-1.yr-1). Moreover, this study indicates that dry deposition in all the ecosystems dominates and represents about 60-70% of the total nitrogen atmospheric deposition. This study on atmospheric nitrogen budget on the African continent is part a comprehensive global precipitation assessment review being prepared by the Science Advisory Group on Precipitation Chemistry of the World Meteorological Organization for publication during 2012. One of the major findings in the estimation of the atmospheric nitrogen budget in Africa (6 to 13 kgN.ha-1.yr-1) is to show that Nitrogen deposition fluxes are of the same order of magnitude than those of US or Europe industrialized countries (10 to 25 kgN.ha-1.yr-1).

P-1-030 Simulation and evaluation of ozone and its precursor gases over Indian region using the high resolution CHIMERE chemical transport model

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Key words
Tropospheric NO_2 Edgar emission Surface Ozone, CO, NOx

This study presents annual simulations of ozone and its precursors made for the first time using CHIMERE regional chemistry transport model over the Indian domain. The model simulations are made using EDGAR emissions (v4.1) on 50 km by 50 km grid resolution for the year 2005. The model simulated ozone and its precursors are evaluated against ground-bashed and space-borne observations. The comparison of model results with seasonal variation of surface ozone observations from eleven sites and diurnal variation of ozone, CO and NOx observations from two sites are presented. In addition, surface ozone measurements aboard 'Sagar

Kanya' over the Bay of Bengal during 2009 are compared with the model simulations. The spatial and temporal variabilities of ozone, CO and NOx are simulated very well by the model. The model in general compare well with the OMI observations during pre-monsoon and monsoon season, but underestimates tropospheric column NO_2 retrievals during post-monsoon and winter season. Model results indicate regional differences in surface ozone seasonality over Indian region. This study shows that CHIMERE model captures many important features of the observations and lends confidence to the use of model for understanding spatio-temporal variability of ozone over India. However, improvement of India emission inventories is also essential for accurately quantifying ozone production in this region.

P-1-031 Modelling future changes in surface ozone: A parameterised approach

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Key words
surface ozone intercontinental transport ozone trends chemistry transport modeling

Changing anthropogenic emissions of ozone precursors are likely to lead to future changes in surface ozone over many parts of the globe. Regional differences in these precursor emission changes lead to differing regional ozone responses, and these are superimposed on a changing background of ozone driven by increases in atmospheric methane. The chemical and transport processes governing surface ozone are reasonably well understood, but there is considerable uncertainty in our ability to represent them and the interactions between them in current atmospheric models. This study uses results from sensitivity studies with 14 global chemistry transport models contributing to the UNECE Hemispheric Transport of Air Pollution (HTAP) assessment to estimate global and regionally averaged changes in surface ozone due to future changes in anthropogenic emissions. This parameterised approach reproduces the results of full model simulations, and additionally allows a regional source attribution and an estimate of the associated uncertainty represented by the variation between models. Using the Representative Concentration Pathway (RCP) emission scenarios, we show how regional surface ozone is expected to respond to emission changes by 2050 and how changes in precursor emissions and atmospheric methane contribute to this. Surface ozone changes are substantially smaller than expected with the SRES emission scenarios, reflecting the more stringent precursor emission controls assumed

under the RCPs. By 2050 there is a 5 ppb difference in surface ozone between the outlying RCP 2.6 and RCP 8.5 scenarios, about 75% of which can be attributed to differences in methane abundance. The study reveals the increasing importance of limiting atmospheric methane growth as emissions of other precursors are controlled, but highlights differences in modelled ozone responses to methane changes of as much as a factor of two, indicating that this remains a major weakness in current models.

P-1-032 Ten years of observations of ozone depleting substances at Monte Cimone (Italy) for deriving trends and regional emissions.

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Key words
Chlorofluorocarbons Ozone depletion Emissions Long term observations

Introduction: Man-made halogenated gases that are potentially harmful for the stratospheric ozone layer are regulated under the Montreal Protocol, a good example of how international agreements can be effective in tackling environmental issues of global significance. In fact, as a consequence of the implementation of the Protocol, tropospheric abundances and emissions of most ozone depleting substances (ODSs) are decreasing. However, the enhancement polar vortex strength and decrease in stratospheric temperatures in the Arctic, likely to be due to change in global climate have been recognised as responsible of an unprecedented ozone loss recently observed over the Arctic. Therefore, the attention on tropospheric levels of ODSs is still high.

Methods: A long-term programme for continuous measurements of ODSs is being carried out since 2001 at the WMO-GAW Italian Climate Observatory on the top of Mt. Cimone (2165 m a.s.l.), the highest peak of the northern Italian Apennines. The observatory is an AGAGE affiliated station, linked to the AGAGE calibration scale and using the same calibration protocol. For estimating atmospheric trends and, consequently, annual growth rates, but also for emission evaluation, a clear identification of the background data is necessary. However, being the station surrounded by quite complex meteorological and emission fields the determination of the baseline is not trivial. Here is applied a statistical method recently developed specifically for this purpose.

Results: Time series of four chlorofluorocarbons, three hydrochlorofluorocarbons, two halons, two chlorocarbons, and two methyl halides are reported, along with their trends, accelerations in the trend, seasonal changes and annual growth rates

derived through the application of a proper polynomial. The influence of emissions from developing countries is discussed. Moreover, the elevations above the baseline are used to detect unreported emissions for the already phased out substances, and source regions are identified.

Conclusion: Direct long term and high-precision observations are necessary in order to correctly evaluate the global atmospheric budget of ODSs. Such observations are the most effective tool for ascertaining the compliance to the International Protocols of the countries pertaining to the site domain.

P-1-033 Global budget of black carbon: model results constrained by HIPPO

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Key words
black carbon wet deposition

We use a global three-dimensional model (GEOS-Chem) to interpret aircraft observations of black carbon (BC) containing aerosol over the Pacific during the HIPPO campaign from 2009 to 2011. The model generally reproduces the observed vertical and latitudinal gradients of BC. This level of agreement was achieved by improving the parameterization of wet deposition of BC, which strongly affects the modeled free tropospheric BC load in this region. Model source attribution shows that the BC originated mainly from anthropogenic sources (biofuel and fossil fuel). Asian emissions dominated anthropogenic BC at all altitudes in the Northern Hemisphere except those near the Arctic surface, where emissions from North America, Europe and Russia were also important. Open fire emissions contributed strongly only over the tropics in Mar-Apr and in Southern Hemisphere during the fire season (Jun-Nov). We will discuss the seasonal trend of BC transport over the Pacific and their implications for climate effect.

P-1-034 Laboratory studies on secondary organic aerosol formation from crude oil vapors

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Key words
Crude oil evaporation secondary organic aerosol VOC oxidation

Introduction

One class of volatile organic compounds (VOCs) that is emerging as important precursors of secondary organic aerosols (SOA) is organic compounds of intermediate volatility (IVOCs), which are not routinely measured nor taken into account as precursors in many models. For example, the Deepwater Horizon (DWH) oil spill in the Gulf of Mexico in April-August of 2010 resulted in a significant release of IVOCs to the atmosphere, which rapidly formed SOA downwind with a very high yield [de Gouw et al., 2011]. As these same compounds are also expected to be present in vehicle exhaust, these findings have implications for urban atmospheres. In this work, we conducted laboratory studies on the SOA formation from crude oil vapors to better understand which classes of compounds in crude oil are the most efficient SOA precursors.

Methods

Proton-transfer-reaction Ion Trap Mass Spectrometry (PIT-MS) is used to measure the vapors released from various fuel samples, including crude oil from South Louisiana, which has a composition similar to crude oil spilled at DWH. We simulate the formation of SOA from the evaporated hydrocarbons at a strongly accelerated rate in a Potential Aerosol Mass (PAM) photooxidation reactor [Kang et al., 2007]. SOA is analyzed by both Aerodyne High Resolution Time-of-Flight Aerosol Mass Spectrometry (HR-ToF-AMS) and a TSI Scanning Mobility Particle Sizer (SMPS). Oxidized gas-phase products are also measured by PIT-MS.

Results and discussion

A number of experiments for South Louisiana crude oil were conducted as a function of UV light intensity in the PAM reactor and as a function of relative humidity (RH). The vapors from the crude oil evaporation show decays with time at different rates depending on their vapor pressure. VOCs (C6-C9 hydrocarbons) with high volatility start at high mixing ratios and decay within ~30 minutes. IVOC (C10-C16 hydrocarbons) mixing ratios start at low values and slowly increase to a maximum after ~30 minutes evaporation because their relative concentration in the liquid crude oil increases due to the decreasing fraction in mixing ratios of high volatile VOCs, then gradually decay to zero. A typical experiment under moderate light intensity and low RH (~10%) conditions shows that SOA mass loadings reach a maximum value also after ~30 minutes evaporation, and then decay slowly over the next 20 hours. SOA mass loadings show a temporal pattern similar to IVOC concentrations.

By changing the UV light intensity and RH, the OH concentration in PAM changes and so does the oxidation rate and aerosol aging process. It is shown that the higher the UV light intensity, the more efficiently the precursors are oxidized and the more SOA is formed. Further investigation of the effect of RH on SOA formation will be performed.

More work will be performed to calculate aerosol yields and verify carbon mass closure by considering the mass of the crude oil, the evaporated precursors, and their gas-phase and aerosol products.

Conclusion

In agreement with the observed aerosol formation downwind from the DWH oil spill, our laboratory experiment showed a strong formation of SOA from IVOCs.

P-1-035 Australasian total column CO and HCHO investigated with FTIR measurements, a Chemical Transport Model (GEOS-Chem) and an Earth-System Model (ACCESS).

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Key words

Southern Hemisphere column measurements atmospheric models

Tropospheric pollutants, such as carbon monoxide (CO) and formaldehyde (HCHO) have a major interaction with hydroxyl radicals, the main cleansing species of the atmosphere. Increased concentrations of tropospheric pollutants can also have detrimental effects on human health and vegetation, so are a priority for study. Enhanced pollution is usually found over urban and industrial areas, such as Wollongong, NSW, Australia, or in regions affected by biomass burning such as Darwin, NT, Australia. There is still much uncertainty in the behaviour and distribution of tropospheric pollutants, particularly in the Southern Hemisphere.

In this study, CO and HCHO over sites in the Australasian region (10°S to 50°S, 105°E to 180°E) are investigated with measurements and model simulations. Remote sensing total column retrievals from ground based Fourier Transform Infrared Spectrophotometers (FTIR) for CO and HCHO are used. The global Chemical Transport Model (CTM) GEOS-Chem has been utilised to simulate atmospheric trace gases over the Australian region from 2004-2009. Subsequent comparison of model output with measurements allows analysis of CTM validity and also helps to improve our understanding of seasonal variability and background concentrations of target trace gases in the Australasian region.

Additionally, total column values are compared with the Australian Community Climate Earth-System Simulator (ACCESS), being developed in Australia as a joint project between the Commonwealth Scientific and Industrial Research

Organisation (CSIRO) and the Australian Bureau of Meteorology. Earth-System models, such as ACCESS, aim to integrate simulation of climate processes, atmospheric chemistry, oceans and land-surface interactions. This combination of systems produces a powerful tool for testing our knowledge of process interaction. Specifically, preliminary results from the chemistry module of ACCESS are analysed here.

Results indicate GEOS-Chem reproduces the magnitude and seasonal cycle of total column measurements within one standard deviation. In contrast, ACCESS consistently underestimates the magnitude of total column values and also misrepresents annual cycle peaks, suggesting emission inventory inconsistencies. Furthermore, a lack of isoprene and related chemistry in ACCESS is likely to impact total column values. Future simulations of ACCESS aim to include updated emission inventories and isoprene chemistry.

Comparison of results from a new Earth-System model with a well established CTM and measurements gives insight into modelling strengths, indicating processes accurately captured by each of the models. Model processes identified as weaknesses brings focus to areas requiring further development. Overall, the study increases our knowledge of atmospheric trace gases in the Southern Hemisphere.

P-1-036 Atmospheric concentrations of particulate sulfate and nitrate in Hong Kong during 1995-2008: impact of local emission and super-regional transport

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Key words
Sulfate Nitrate PM_{10} Wet deposition acidity Emissions Long-distance source

The release of large amounts of sulfur dioxide (SO_2) and nitrogen oxides (NOx) from the burning of fossil fuel leads to regional air pollution phenomena such as haze and acidic deposition. Despite longstanding recognition of the severity of these problems and the numerous studies conducted in China, little is known of long-term trends in particulate sulfate and nitrate and their association with changes in precursor emissions. In this study, we analyze records covering a 14-year period (1995-2008) of PM_{10} composition in the subtropical city of Hong Kong, situated in the rapidly developing Pearl River Delta region of southern China. A linear regression method and a Regional Kendall test are employed for trend calculations. In contrast to the decreased levels of SO_2 and NOx emissions in Hong Kong, there are increasing overall trends in ambient concentrations of PM_{10} sulfate and PM_{10} nitrate, with the most obvious rise seen during 2001-2005. The percentages of sulfate and nitrate in the PM_{10} mass and rainwater acidity also increased. Backward trajectories are computed to help identify the origin of large-scale air masses arriving in Hong Kong. In air masses dominated by Hong Kong urban sources and ship emissions, there was no statistically significant trend

for PM_{10} sulfate and a small increase for PM_{10} nitrate; however, the evident increases in PM_{10} sulfate and PM_{10} nitrate concentrations were observed in air masses originating from eastern China and are generally consistent with changes in emissions of their precursors in eastern China. Examination of PM_{10} mass data recorded at a pair of upwind-urban sites also indicates that long-range transport makes a large contribution (> 80%) to PM_{10} loadings in Hong Kong. Together with our previous study on the ozone trend, these results demonstrate the important impact exerted by long-distance sources and suggest a need to consider the impact of super-regional transport when formulating air-quality management strategy in Hong Kong in future.

P-1-037 Uncertainty in biogenic isoprene emissions in East Asia and their impacts on the tropospheric chemistry

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Key words
Isoprene HCHO column CMAQ SCIAMACHY

As a major precursor of the ozone and SOAs (secondary organic aerosols) formations, biogenic isoprene is of primary importance in the atmospheric chemistry. Isoprene also influences the level of inorganic aerosols (i.e., sulfate and nitrate) by controlling OH radicals. However, isoprene emission fluxes are highly uncertain in East Asia. While isoprene fluxes from the GEIA (Global Emissions Inventory Activity) and POET (Precursors of Ozone and their Effects in the Troposphere) inventories estimate approximately 5 Tg per month in East Asia for the summer seasons, those from the MEGAN (Model of Emissions of Gases and Aerosols from Nature) and MOHYCAN (MOdel for Hydrocarbon emissions by the CANopy) estimate approximately half of the GEIA and POET inventories for the same periods. In order to evaluate and/or quantify the magnitude of the isoprene fluxes over East Asia, the HCHO columns obtained from the SCIAMACHY (SCaning Imaging Absorption spectroMeter for Atmospheric CHartographY) observations were compared with the HCHO columns from the CMAQ (Community Multi-scale Air Quality) simulations over East Asia. In this study, US EPA Models-3/CMAQ v4.7.1 modeling using the INTEX-B (Intercontinental Chemical Transport Experiment-Phase B) and REAS (Regional Emission inventory in ASia) emission inventories for anthropogenic pollutants and GEIA, MEGAN, and MOHYCAN data for biogenic species (i.e., isoprene and mono-terpene) was carried out in conjunction with the Meteorological fields generated from the PSU/NCAR MM5 (Pennsylvania state University/National

Center for Atmospheric Research Meso-scale Model 5) model for the summer episodes of the year 2006. In this study, isoprene fluxes from the GEIA inventory are highly overestimated, particularly over South China. In addition to an evaluation of the isoprene fluxes, we examined the budget of columnar HCHO generated from several sources (i.e., methane, anthropogenic VOCs, biogenic VOCs, and biomass burning). The methane oxidation shows the largest contribution to the columnar HCHO over East Asia. However, the BVOC oxidations by OH radicals are a major contributor of the HCHO columns in South China. In this study, we further investigated the impact of the uncertainty in biogenic emission inventory on the variations or levels of oxidants (OH, HO_2, O_3, and H_2O_2) in East Asia.

P-1-038 Atmospheric peroxyacetyl nitrate (PAN) observed at Nam Co, Tibetan Plateau, China: the roles of dynamic and photochemical processes

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Key words
PAN Tibetan Plateau photochemical processes

With a very high average elevation (4200m asl) and an extremely low population density (2 residents/km^2), the Tibet Plateau (TP) is the most clean part of the Eurasian continent, but may be influenced by increasing anthropogenic emissions in the region and its surrounding. Recently, potential impacts of human activities on air compositions over the TP region and their consequences have caused concern. However, our knowledge about atmospheric trace gases and their related physicochemical processes over the TP is inadequate due to very limited studies focusing on atmospheric chemistry in the remote region. In summer 2011, we participated in a field campaign at the Nam Co Station (N30°46.44′, E90°59.31′, 4730 m asl), which was one of the activities of the Tibet Ozone, Aerosol, and Radiation (TOAR) project. During this campaign, we measured the concentrations of peroxacetyl nitrate (PAN) and some other reactive gases (O_3, CO, NOx, SO_2, etc.) in the surface layer. PAN was observed using a gas chromatograph equipped with an electron capture detector. In this paper, we present the measurements of PAN during the period August 16-25, 2011. The hourly concentration of PAN varied in the range of 0.10-1.49 ppb and averaged 0.37 ppb. The time series of PAN showed a few abrupt fluctuations, which caused much higher PAN than its normal level. The diurnal variation of PAN displayed later night or earlier morning peaks, inconsistent with local photochemical production. The PAN concentration followed the general trend of O_3 under normal conditions, leading to a significant

correlation between the concentrations of both oxidants. However, nearly no correlation was observed during August 22-24, when strong variations of the gases occurred. To understand this phenomenon, we analyzed the 500-hPa geopotential height field and omega field and simulated the back trajectories of air masses arriving at the site. The results suggest that the observed large changes in the PAN and O_3 levels were caused by the transformation of atmospheric circulations over the TP region. The upward airflow under southerly wind corresponded to low O_3 levels and the downward airflow under northerly wind corresponded to high O_3 levels. High PAN levels occurred during the period of transition from the one transport condition to the other. Therefore, dynamic process plays an important role in variation of both PAN and O_3. The actual origin of higher PAN in the transitional airstream is unknown at this time. More studies are needed for better understanding the variation of PAN and its impact on climate and environment in the TP region.

P-1-039 MAX-DOAS measurements of NO_2, HCHO and CHOCHO at a rural site in southern China

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Key words
MAX-DOAS HCHO CHOCHO

We performed MAX-DOAS measurements during the PRIDE-PRD2006 campaign in the Pearl River Delta region (PRD), China, for 4 weeks in July 2006 at a site located 60 km north of Guangzhou. The vertical distributions of NO_2, HCHO, and CHOCHO were independently retrieved by an automated iteration method. The MAX-DOAS measured NO_2 mixing ratios showed reasonable agreement with the simultaneous, ground based in-situ data. While the tropospheric NO_2 vertical column densties (VCDs) observed by OMI on board EOS-Aura satellite agreed with those by MAX-DOAS, the 3-D chemical transport model CMAQ overestimated the NO_2 VCDs as well as the surface concentrations by about 40%. From this observation, a reduction of NOx emission strength in CMAQ seems to be necessary in order to well reproduce the NO_2 observations. The average mixing ratios of HCHO and CHOCHO were 12 ppb and 1.6 ppb, respectively substatially higher than in other rural or semirural environments. The high ratio of 0.135

between CHOCHO and HCHO corresponds to the high VOCs reactivity and high HOX turnover rate consistent with other observations during the campaign. A 0-D box model using the MCM3.2 mechanism was employed to simulate the formation of HCHO and CHOCHO during the campaign. While the CHOCHO concentrations can be well reproduced by the model, the simulated HCHO concentrations are significantly higher than the observations. We propose that the difference between the measurements and the model results is caused by the irreversible uptake of HCHO on acidic aerosols, a process normally thought to occur in the upper troposphere only.

P-1-040 Long-term analysis of satellite and model simulated tropospheric NO_2 columns over Asian Megacities

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Key words

Tropospheric NO_2 Satellite observation Model simulation Emission inventory Asia

Nitrogen dioxide (NO_2) is one of the major air pollutants playing an important role in atmospheric chemistry and of great concern due to its adverse health effects. The emissions of NO_2 has built up at high concentration in Asia due to rapid economic growth for the past decade. In this study, we analyze the long-term trend of tropospheric NO_2 columns based on satellite observations, GOME, SCIAMACHY, OMI and GOME-2, over the megacities of the countries in Southeast Asia region and including China and Japan during the period of 1996-2011. The results show that most of the cities have unclear increasing trends except for China. Shanghai presents a significant increase of tropospheric NO_2 columns with the increasing trend of 21.5% per year. In additional, Beijing shows the highest concentration of monthly averaged tropospheric NO_2 columns of 25.11×1015 molecules/cm^2 along the study period. We also investigate the seasonal variation of tropospheric NO_2 columns observed by the satellites. The results reveal that with the difference latitude zones, the seasonal variability performs differently in a particular way. The cities that are located in low-latitude and mid-latitude zones have the highest concentration during winter months (November – February) and lowest during summer months (June – August). On the contrary, the cities located around Equator zone have opposite results due to the difference in meteorology. The study of the relationships between four satellites is performed and gave relatively good agreements with the correlation coefficient larger than 0.75. The discrepancy between the satellites is mainly stem from the differences in satellite crossing time and spatial resolutions. Time series of tropospheric NO_2 columns are plotted together with NOx emissions from various regional and global emission inventories and activity statistics in term of monthly and yearly averages during 1996 - 2009. Most of the cities located in mainland illustrate good correlations of

NOx emissions and tropospheric NO_2 columns. Reversely, the cities near coastal area show weak correlation owing to the effects of meteorology that subsequently cause the transport of NO_2 and bring to the discrepancy between NOx emission and satellite data. Furthermore, the validation of satellite tropospheric NO_2 observations is performed by using NO_2 data from surface monitoring in the period of 2001 - 2008. In case of OMI and GOME-2 observations, they present better agreements with surface measurements than GOME and SCIAMACHY. The reasons are due to the finer horizontal coverage of OMI and GOME-2 instruments. The simulation of tropospheric NO_2 columns over the study area have been conducted with chemical transport model and then comparably analyzed with those retrieved from satellite observations. Most of the results from the model simulations are generally in agreement with satellite measurements. However, the difference between the satellite and simulated tropospheric NO_2 columns can be explained by the inaccuracy in emission inventories fed into the model and the uncertainties in the satellite retrievals.

P-1-041 Understanding source origin and emission area of carbonaceous aerosol transported from East Asia and its contribution to the composition of aerosol collected at Cape Hedo, Okinawa, Japan.

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Key words
OC/EC CHAAMS Source origin Emission area Contribution

Black Carbon (BC) or Elemental Carbon (EC) gained more attention recently because not only it is considered as the second largest contributor to global warming but also its radiative forcing depends on source origin. There are large uncertainties in the estimates of global and regional climate effects of combustion emissions due to the lack of long-term global and regional monitoring of aerosol composition. Recently, several observatories are established to investigate the long-term effects of aerosols on climate in East and South Asia under the Atmospheric Brown Cloud (ABC) project. We conducted observations at the Cape Hedo Atmosphere and Aerosol Monitoring Station (CHAAMS) in Okinawa, Japan, which is assigned as a major site of ABC-Asia. Long-range transported carbonaceous aerosols from East Asia to East China Sea were investigated. Source origin and area of emission as well as their contribution to regional environment were evaluated.

Mass concentrations of heavy metals, ionic components, and OC/EC were measured at CHAAMS in 2010. Concentrations of these chemical substances were simultaneously high in spring and winter but low in summer. It was interpreted that pollutants were transported to CHAAMS by migratory high pressures and cold fronts from the Asian continent during spring and winter, whereas monsoon transported oceanic air mass from the Pacific during summer.

The relative influence of aerosol-source categories is analyzed by evaluating the potential source contribution function (PSCF) based on positive matrix factorization (PMF) for source regions. Five source categories were identified: I. sea salt and nitrate, II. dust, III. coal combustion, IV. oil combustion, and V. secondary species. By combining PMF predicted factor contributions with identification of source location through PSCF in III and IV factors it was shown that the source origin of carbonaceous aerosols in North China was coal combustion different from oil combustion for South China, Japan and Korea. The factor V showed excess of sulfate to ammonium. NH_4/SO_4 ratio tended to be low for long transport distance and/or long residence time over the sea. One of the reasons is that ammonium was lost by deposition while sulfate increased by SO_2-to-sulfate conversion. Dust results indicated the presence of local dust around CHAMMS. The relative contributions of these five factors to ambient EC concentrations were 7% from dust (II), 13% from oil combustion (IV), 19% from sea salt and nitrate (I), 29% from secondary aerosol (V), and 33% from coal combustion (III). Contribution of the secondary aerosol factor (V) is higher in spring than in winter. These results can be understood by considering that the production of secondary aerosol depends on light intensity and temperature resulting in enhanced photochemical aging in spring compared to winter. Contribution of coal combustion factor (III) revealed a significant contribution from residential areas in north China in winter. It was found that coal combustion had higher contribution to EC in winter (41%) while oil combustion had higher contribution in spring (20%). It is notable that both coal and oil combustion contributed significantly to EC in CHAAMS.

P-1-042 Variation of molecular hydrogen tropospheric concentrations over Southern Poland - results of the continuous chromatographic measurements.

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Key words
molecular hydrogen atmospheric composition

Although hydrogen is one of the fundamental constituents of the earth's atmosphere its global balance is still poorly clarified. A few developed inventories diverging values for efficiency of sources and sinks of this gas. The European network for the hydrogen concentrations measurement is based on several unevenly spaced measurement points. While in 2009 MPI Jena has delivered accurate scale for hydrogen measurements and the techniques of analyses are well described, still large areas of Central Europe is uncovered by representative stations.

The first measurement point, established under the EUROHYDROS EU program, on the territory of Poland was Kraków city. Different laboratory setups was tested there and compared to each other. The Kraków area has significant car traffic and its geographical location implies frequent temperature inversions in lower troposphere leading to the accumulation of trace gases in atmosphere of the city. Observations launched in 2007 revealed that the concentration of hydrogen fluctuates strongly within diurnal and seasonal timescales. Its average concentration is three times larger than this, observed at the other stations. The European “background” concentrations of hydrogen are not reflected in the Krakow record.

An ideal place to carry out observation of the regional air composition for Central Europe is a research station located in the Meteorological Observatory at Kasprowy Wierch. Measurement point at the top of mountain peak with elevation of 2000m a.s.l. gives an access to the well mixed troposphere. The station delivers also the necessary facilities and logistics. Since year 1996 greenhouse gas measurement program has been operating at this point.

The first measurements of atmospheric concentrations of hydrogen at Kasprowy Wierch were performed in year 2010, based on dedicated gas chromatograph using RGD detector installed at the station. Analysis of hydrogen content in the outside air is performed without any enrichment process with precision better than 1ppb. But a temporary problems with UV lamp power supply module considerably hampers the research and adversely affect the reproducibility of the analysis. The results should be treated as a preliminary diagnosis of the diurnal and seasonal variation of the hydrogen concentration at the mountain site.

The average short term variation of hydrogen concentrations does not exceed 50ppb and reveals the periodic nature except special situations when the concentration abruptly rises up by 150ppb and returns to an average of 480-520ppb. It is possible to observe the variations with larger time constant probably related with mezzo-scale circulation in atmosphere and slight seasonality in the level of hydrogen observed in autumn and in winter (with amplitude of 50ppb).

Measurements carried out at Kasprowy Wierch were also used to balance the hydrogen for the city of Kraków.

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P-1-043 Model Evaluation on Texas Power-Plant Plumes: TexAQS II 2006 Case Study

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Key words

General reactive-dispersion plume model TexAQS II 2006 Model evaluation

The amounts of pollutants emitted from coal-burning power plants have increased, and the pollutants have harmful influences on atmospheric environment. During the second Texas Air Quality Study 2006 (TexAQS II 2006) on 16 September 2006, pollutant concentrations were measured by NOAA WP-3D aircraft with successive transects across power-plant plumes in Texas, USA. To extensively understand and assess atmospheric impacts of the power-plant emissions, a general reactive-dispersive plume model (RDPM) was applied to simultaneously simulate the dynamics and photo-chemistry of the Texas power-plant plumes. First of all, the simulation performances of the RDPM were evaluated by a comparison study, using the observation data obtained from the measurements of a NOAA WP-3D flight during TexAQS II 2006 airborne field campaign. The simulation conditions and parameters, such as the meteorological conditions (e.g., wind speed and direction, stability class, air temperature, mixing height, etc.), emission rates, and background gas and particulate species concentrations, were directly or indirectly determined from the observation data of NOAA WP-3D flight. On 16 September, the WP-3D aircraft observed mainly southerly wind with average wind speed of ~ 7 m/s, traversing the Monticello and Welsh power-plant plumes four times from transects A to D. This power-plant plume cases were selected in this study, because a large number of nitrogen oxides and sulfur dioxide concentrations inside the power-plant plumes were measured without any interruption of other emission sources. For the Monticello and Welsh power-plant plumes, the model-predicted concentrations showed good agreements with the observed concentrations of ambient species (e.g., nitrogen oxides, ozone, sulfur dioxide, etc.) at the four transects. In case of nitrogen oxides and sulfur dioxide, the correlation coefficients (R) between model-simulated and observed concentrations for the Monticello and Welsh plumes range from 0.86 to 0.96 and from 0.57 to 0.84, respectively. Based on these RDPM results, the power-plant plume chemistry and its possible impacts on atmospheric environments were also analyzed.

P-1-044 Aerosol Elemental Carbon Concentration and Isotope Observations over Canada: Constraints on Regional Fossil Fuel Emissions

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Key words
elemental carbon BC fossil fuel emissions carbon isotope Arctic

Black carbon (BC) plays important roles in public health, environment as well as Earth's climate. Based on recent studies, BC can be defined as a solid form of mostly pure carbon that is the most strongly light-absorbing component of particulate matter. It is formed by the incomplete combustions of fossil fuels, biofuels and biomass. Usually BC mass measurements are equally referred as elemental carbon (EC) measurements. Although lots of attention has recently been paid to BC, there are still large gaps in the knowledge of quantifying their emission sources and estimating their impacts on regional and global climate. Because of the short atmospheric life-time of EC, changes in source emission can be inferred from changes in its atmospheric concentration. Because of the inert nature of EC, its carbon isotope compositions (^{13}C) can provide valuable insights to the emission sources/regions for these aerosols.

An observation network of aerosol carbon has been strategically set up and co-located with CO_2 across Canada since 2006. These sites represent different geographic locations with various source influences, including a typical urban site (Toronto), a rural site (Egbert), two boreal forest sites (Fraserdale & ETL, in the middle of the continental land), a high altitude site in Western Canada (Whistler Mt.) and an Arctic baseline site (Alert). An additional urban Beijing site was included here for comparison to study the possible impacts of Asia-Pacific transport on the Arctic.

Weekly integrated quartz filter samples collected at these sites over the period (2006 – 2010) were analyzed for EC concentrations. The weekly values were averaged into two seasonal means of summer-fall (May-Oct.) and winter-spring (Nov.- April) for the entire data set. The results show that: 1). EC concentrations during warm seasons are higher than those in cold seasons at all sites except for Alert (where the opposite patterns are found), which are consistent with source influencing patterns due to anthropogenic emissions; 2). All the EC concentrations during warm seasons at Toronto, Egbert, Fraserdale as well as those during cold seasons at Alert show a decrease trend from 2007 through 2009 and a slight increase in 2010, which are consistent with the changes of the fossil fuel emission inventories (CDIAC: http://cdiac.ornl.gov/trends/emis/meth_reg.html) in North America & Europe; 3). Large variations in EC concentration at the ETL site during the warm seasons in 2006, 2008 and 2010 indicate that the impact of biomass burning emissions could be significant on the atmospheric loading; 4). There are significant differences in carbon isotope compositions of EC between the urban Beijing site and the Alert site during winter seasons, implying that the aerosol EC in the arctic was not likely from the East Asia region.

P-1-045 Volatile organic compounds (VOCs) observed at an alpine station in the Tibet

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Key words
the Tibet Plateau VOCs ozone photochemical precursor

As part of an intensive observation campaign to study the alpine atmospheric chemistry, carried out in the Tibet Plateau, China, 24 air samples were collected and analyzed to characterize volatile organic compounds (VOCs) at the Nam Co station (30°46'N, 90°59'E, 4730 m a.s.l). The station is located in the southeast shore of Nam Co Lake, northern slope of the Nyainqentanglha Mts. During the sampling period, August 2011, the surrounding area of the station was covered with alpine meadow. More than 90 species (C5～C15) including various paraffins, olefins, aromatics, alcohols, aldehydes, ketones, and halogenated hydrocarbons were identified and quantified by a Thermal Desorption/Comprehensive Two-Dimensional Gas Chromatography/Flame Ionization Detection (TD/GC×GC/FID) system.

The total concentration level of the quantified organics averaged 15.9 ppb and varied mostly from 5 to 20 ppb. Though a peak 46.9 ppb was observed in a morning sample, the total VOCs concentration level at the station was much lower than those at other sites in China. Total measured VOCs were dominated by aromatics (34%, including PAHs), oxygenated hydrocarbons (28%), and halogenated hydrocarbons (24%). The leading compounds were naphthalene, carbon tetrachloride, methyl ethyl ketone, xylene, and butanal.

P-1-046 Secondary Organic Aerosol in the Pearl River Delta, South China

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Key words
Secondary organic aerosol Aromatics

Introduction

Secondary organic aerosol (SOA), the dominant fraction of fine particle matter (PM2.5), are produced by homogenous and heterogeneous reactions of volatile organic compounds (VOCs) as well as aging of primary aerosol in the air. To fully establish the role of SOA in climate or air quality models, comprehensive knowledge about SOA formation and sources are still urgently needed, although great improvement has been achieved in the last decade. As for SOA precursors, global emissions of biogenic VOCs (terpenes) are dominant over anthropogenic VOCs (e.g. aromatics). However, recent studies suggested that anthropogenic SOA might be more significant than previously thought, especially in urban regions. In this study, PM2.5 samples were collected in the central Pearl River Delta (PRD), which is one of the most industrialized and densely populated regions in China. Major tracers for SOA were identified to quantify source contributions from different precursors including biogenic precursors (isoprene, monoterpenes, and sesquiterpenes) and anthropogenic precursors (aromatics).

Methods

Filter samples were extracted by sonication with mixed solvent of dichloride methane and methanol. Prior to solvent extraction, structure-like compounds were spiked into the samples as internal standards. The extracts of each sample were filtered, combined and concentrated. Methylation and silylation were undertaken for these samples, respectively, and then followed by the GC-MSD analysis. The typical SOA tracers from significant biogenic (isoprene, monoterpenes, sesquiterpenes) and anthropogenic (aromatics) precursors were determined and applied to do source apportionment of secondary organic carbon (SOC).

Results and Discussion

The apportionment results revealed that over 75% of the estimated SOC were from aromatics in both summer and fall-winter, indicating that anthropogenic hydrocarbons are dominant SOC precursors in the highly populated, industrialized, and urbanized PRD. The composition of SOC in the PRD is quite different from that in the southeastern US where isoprene-derived SOC are dominated. Moreover, the observation in the PRD found that the aerosol acidity had significant influence on SOA tracers' formation. However, the slopes of tracers vs. aerosol acidity showed huge gap between our campaign and chamber simulation, suggesting that cautions should be taken when extrapolating chamber results to ambient samples and further chamber studies are needed especially under the situations as occurring in the PRD region.

P-1-047 Investigating the radiative effect of aerosol on vertical distribution of boundary layer by Lidar: A case study

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Key words
lidar aerosol radiative effect mixing layer

This study is aimed to understand the possible radiative effect of aerosols basing on ground based aerosol measurements and lidar installed in National Taiwan University in Taipei. The optical and radiative properties of aerosols are dominated by aerosol composition, particle size, hygroscopicity property, and shape. In this study, aerosol instruments including integrating nephelometer, open air nephelometer, aethalometer are applied to investigate the relationship between aerosol hygroscopicity properties and aerosol types.

The possible radiative effect of aerosols are approached by vertical atmospheric extinction profiles measured by lidar. Calculated atmospheric and aerosol heating effects was compared with vertical meteorological parameters measured by radiosonde. The result shows light-absorbing aerosol has the potential to affect the stability of planetary boundary layer.

P-1-048 Harmonization and evaluation of surface emissions within the MACC-II project and distribution through the ECCAD database

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Key words
Surface emissions Forecasting database

The MACC-II (Monitoring Atmospheric Composition and Climate) project aims to provide data records on atmospheric composition for recent years, data for monitoring present conditions and forecasts of the distribution of key constituents for a few days ahead. As part of this project, surface emissions at the global and regional scales are being developed for gaseous and particulate species. We will review the results obtained as part of he project, with a focus on anthropogenic and natural emissions. We will provide the first conclusions of a workshop on the

issues related with emissions from shipping.

During the past few years, the ECCAD (Emissions of atmospheric Compounds & Compilation of Ancillary Data) database was developed, in order to distribute the emissions developed as part of MACC-II and to provide a user-friendly access to a large set of publicly available surface emissions and ancillary data, i.e. data on land use, active fires, burned areas and population. All the data are provided at a 0.5x0.5 or 1x1 degree resolution. We will describe ECCAD, a database on emissions developed as part of the Global Emissions InitiAtive (GEIA). ECCAD provides detailed metadata on each of the datasets, including information on complete references and methodology, and links to the original inventories. Several tools are provided for the visualization of the data, for computing global and regional totals and for an interactive spatial and temporal analysis. The data can be downloaded in a NetCDF CF-compliant format.

P-1-049 ANALYSIS OF SURFACE OZONE TRENDS AT THE MT. CIMONE GAW GLOBAL STATION (ITALY)

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Key words
Ozone Trend Europe

Mt. Cimone (44.18N, 10.70E, 2165 m a.s.l.) is the highest peak of the Italian northern Apennines and is characterised by a 360° free horizon. As being located South of the Alps and of the polluted Po basin, the observations of atmospheric composition conducted at this Global GAW-WMO Station, provide useful information for evaluating the role of regional and long-range transport processes in modifying the tropospheric background conditions in the Mediterranean basin/Southern Europe. Moreover, this long-term monitoring activity helps in quantifying the existing trend of climate-altering substances and atmospheric pollutants.

Surface ozone (O_3) measurements are continuously carried out at Mt. Cimone (CMN) since January 1996 at the Italian Climate Observatory “O. Vittori” (ICO-OV), the research infrastructure managed by the National Research Council – Institute for Atmospheric Sciences and Climate (ISAC-CNR). During the period 1996-2009, CMN has been characterised by an average O_3 monthly mean of 54.4 ± 8.9 ppb. The monthly O_3 values (calculated using 1-hour mean values) showed a typical seasonal cycle characterised by winter minima and spring and summer maxima, often resulting in a broad spring-summer peaks.

With the aim of analysing the long-term trends of surface O_3, we calculated slope of the linear regression line of monthly mean anomalies over two reference periods: 1996 – 2009 and 2000 – 2009. Because at CMN, atmospheric compounds variability can be influenced (particularly during the warm months) by “thermal” wind circulations, we performed trend analyses as a function of different “time-window”: all data, nigh-time data (from 23:00 to 4:00 UTC+1) and day-time data (from 11:00 to 16:00 UTC+1).

Basing on the analysis of monthly mean O_3 anomalies, no significant positive or negative linear trends have been observed at the Mt. Cimone GAW Global Station (CMN) over the period 1996 – 2009. Except than for the summer seasons, the period 2000 – 2009 has been characterised by larger annual trends in comparison with the period 1996 – 2009. Only considering the observations recorded for “day-time” data, a statistically significant trend (0.28 ± 0.26 ppb/yr) was calculated over the period 2000 – 2009. However, this latter value appears to be partially determined by specific O_3 episodes. By disaggregating the analysis on a seasonal basis, the highest trends have been detected for the spring season with linear slopes ranging from 0.17 ± 0.46 ppb/yr for “day-time” data selection 1996 – 2009 to 0.59 ± 0.88 ppb/yr for “day-time” selection over the period 2000 – 2009.

P-1-050 Long-term timeseries of CO and C_2H_6 columns at southern-hemisphere mid- and high latitudes and implications for global isoprene emissions

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Key words
carbon monoxide and ethane trends isoprene emission

We present carbon monoxide (CO) and ethane (C_2H_6) columns derived from measurements by ground-based solar Fourier Transform Spectroscopy at Lauder, New Zealand (45^{O}S, 170^{O}E) and at Arrival Heights, Antarctica (78^{O}S, 167^{O}E) from 1997 to 2009. Significant negative trends are calculated for both species at both locations. We use a tropospheric chemistry-climate model UM-CAM to simulate CO and C_2H_6 columns for the period of 1997-2009, assuming only biomass burning emissions to vary interannually. The model-simulated tropospheric columns of CO and C_2H_6 compare well with the measured columns of both species. The model sensitivity calculations indicate that long-range transport of

biomass burning emissions from southern Africa and South America dominate the seasonal cycles of CO and C_2H_6 at both Lauder and Arrival Heights. Variations in biomass burning emissions associated with large-scale El Nino Southern Oscillation and prolonged biomass burning events, e.g. the Australian bush fires, result in large interannual variations in mid- and high SH.

Using in addition thechemical-transport model GEOS-Chem, we investigate how isoprene emissions influence CO columns measured from Lauder and Arrival heights and two locations in Australia (Wollongong: 34^{O}S, 151^{O}E, and Darwin: 12^{O}S, 131^{O}E). Both sets of model simulations imply that with commonly used global total isoprene emissions of 500-600 Tg/year modelled CO columns significantly overestimate the observations at these four sites. When the emissions are reduced to 390 Tg/year, a general improvement of the fits ensues at all four sites.

P-1-051 Changes and trends of ozone in the southern mid-latitude upper troposphere and lower stratosphere derived from long-term ozonesonde and ozone lidar measurements at Lauder, New Zealand

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Key words
Ozonesonde and ozone lidar measurements trends

Ozone in the upper troposphere / lower stratosphere (UTLS) is of particular significance in the global radiation budget because of the low temperatures in this region. It is therefore important to monitor the UTLS region for trends and variability of ozone. Satellite observations of ozone are typically associated with large uncertainties here. We present long-term timeseries of ozone profiles from ozonesonde and lidar measurements made at Lauder, New Zealand (45^{o}S, 169^{o}E), a clean, data-sparse region representative of the southern mid-latitudes. We calculate trends of ozone in the UTLS-region from both timeseries and investigate the impacts of changes in stratospheric ozone, anthropogenic emissions and natural variation on UTLS ozone.

P-1-052 An investigation of atmospheric mercury deposition in Thailand

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Key words

atmospheric mercury deposition MM5/CAMx

Thailand, located in Southeast Asia, has various sources of atmospheric mercury (Hg) emissions that results an increasing concern about adverse affected Hg, mostly after deposition. Recent global assessment on atmospheric Hg transportation and transformation indicated relatively high level of Hg deposition flux in many Asia regions, which Northern of Thailand is a part. To better understand the mechanism of Hg emission to Hg deposition in Thailand, couple model of MM5/CAMx was used. Emissions in the year 2010 were obtained from local development and were processed to input in the CAMx. Simulations were performed for 4 episodes in Jan, April, August and December in year 2010 to investigate seasonal variation of Hg concentration and deposition. Annual deposition as well as emission outflow are investigated and discussed.

P-1-053 Volatile Organic Compounds Compositions of Biomass Burning and Coal Combustion Plumes in China

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Key words

Volatile organic compounds Emission ratios Biomass burning Coal combustion

Volatile organic compounds (VOCs) were measured using proton transfer reaction mass spectrometer (PTR-MS) and other online instruments in several campaigns in Eastern China. Acetonitrile measurements in urban environments and tunnel experiments (Beijing and Shanghai) indicated that emission ratios of acetonitrile to CO in urban emissions are in the range of 0.2-0.3 ppb/ppm, which is significantly lower than emission ratios in biomass burning emissions (1.1-17.3 ppb/ppm). Thus, concentration ratios of acetonitrile to CO were used to identified biomass burning plumes in these campaigns. The enhancement ratios of various VOCs species in biomass burning plumes were determined from linear regressions of concentrations. OVOCs accounted for 54% of the total VOCs in biomass burning emissions, following by alkenes (24%), alkanes (18%) and aromatics (4%). The

dominance of OVOCs in biomass burning plumes is distinct to the VOCs compositions in urban environments. Enhancement ratios of oxygenated VOCs (OVOCs) to CO in China were in the higher range of the values reported in other regions of the world. The main NMHCs species in biomass burning plumes were ethane, ethene, acetylene, propane, propene and benzene, which were consistent with the source profiles developed in the laboratory. The concentrations of methyl chloride and potassium (K^+) in aerosol can not be used as biomass burning tracers in the complicated environments in China, due to the emissions from other sources. Plumes with enhanced concentrations of benzene (up to 16.4 ppb), CO and naphthalene (up to 9.3 ppb) were observed in a campaign conducted at an island of East China Sea. These plumes were identified as coal combustion from nearby factories. The major VOCs species in the coal combustion were C2 hydrocarbons, propane, propene and benzene. No OVOCs emission was found in these coal combustion plumes. The VOCs compositions determined from ambient measurements are valuable for VOCs source characteristics that are limited reported in China, especially for OVOCs species.

P-1-054 Nocturnal Ozone and its Relationship to Air Quality

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Key words
Tropospheric ozone Air quality modeling

At night, ozone can be transported long distances above the surface inversion layer without chemical destruction or deposition. As the boundary layer breaks up in the morning, this nocturnal ozone can be mixed down to the surface and rapidly increase ozone concentrations at a rate that can rival chemical ozone production. Most regional scale models that are used for air quality forecasts and ozone source attribution do not adequately capture nighttime ozone concentrations and transport. We combine ozone profile data from the NASA Earth Observing System (EOS) Tropospheric Emission Spectrometer (TES) and other sensors, ozonesonde data collected during the INTEX Ozonesonde Network Study (IONS), EPA AirNow ground station ozone data, the Community Multi-Scale Air Quality (CMAQ) model, and the National Air Quality Forecast Capability (NAQFC) model to examine air quality events during August 2006. We present both aggregated statistics and case-study analyses that assess the relationship between the models' ability to reproduce surface air quality events and their ability to capture the vertical distribution of ozone at night. We find that the models lack the mid-tropospheric ozone variability seen in TES and the ozonesonde data, and discuss the conditions under which this variability appears to be important for surface air quality.

P-1-055 Submicron particles influenced by continental outflow: results from a high-resolution aerosol mass spectrometer measured at a regional receptor site of China

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Key words
Organic aerosol Positive Matrix Factorization(PMF) regional recepotor site aerosol mass spectrameter(AMS)

Submicron particles have significant impacts on regional air quality, climate change and human health. However, the sources and chemical aging processes of aerosol in the atmosphere are not well understood. A high resolution time-of-flight

aerosol mass spectrometer (HR-TOF-AMS) was deployed at a remote island site (Changdao Island, 37.99°N, 120.70 °E) in Bo Sea to measure aerosol compositions from March 20 to April 24, 2011. The concentrations of submicron particle (PM_1) were 47.1±36.4 μg m^{-3} during the whole campaign (excluded local biomass burning and coal burning influence) and showed distinct differences among plumes with various wind directions and source regions. Nitrate fractions in PM_1 increased as PM_1 loading increasing, especially in the heavy polluted plumes. Comparing to the results in urban environments in Beijing and Shenzhen, the elementary ratio of oxygen to carbon (O:C) at Changdao was significantly high, indicating the heavily processed aerosol compositions during the campaign. The evolution of OA elemental compositions in the Van Krevelen diagram (H:C vs. O:C) showed a slope of -0.63, which was flatter than the value observed in summer of Beijing (2008, -0.88 and 2010, -0.73) and steeper than that in winter of Beijing (-0.55). The results confirmed that the aging of ambient OA followed a slope between -1 and -0.5. Positive Matrix Factorization (PMF) resolved four organic aerosol components from high resolution organic mass spectral dataset, including low-volatile oxygenated organic aerosol (LV-OOA), semi-volatile oxygenated OA (SV-OOA) components, hydrocarbon-like OA (HOA) and coal combustion OA (CCOA). The mass spectrum of CCOA was dominated by fragments (m/z 128, 152, 178 etc.) from polycyclic aromatic hydrocarbons (PAHs). The emission ratios of OA to CO during strong coal combustion and biomass burning plumes were comparable with the results reported in other similar source influenced plumes, which could be used as an refrence value for source emssion ratio in China.

P-1-056 Distribution, variability and sources of tropospheric ozone over South China in spring: Intensive ozonesonde measurements at five locations and modeling analysis

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Key words

ozone vertical profile East Asia stratosphere-troposphere exchange (STE) Asian biomass burning lightning GEOS-Chem 3-D chemical transport model (CTM)

We examine the characteristics of the spatial distribution and variability of tropospheric ozone (O_3) by analysis of 93 ozonesonde profiles obtained at five stations over South China (18-30 degree N) during the first phase of Transport of Air Pollutant and Tropospheric Ozone over China (TAPTO-China) field measurement campaign in spring (April and May) 2004. We use a global 3-D chemical transport model (GEOS-Chem) to interpret these characteristics and to quantify the sources of tropospheric O_3 over South China during this period. The observed tropospheric O_3 mixing ratios show strong spatio-temporal variability, owing to the complex interplay of surface (anthropogenic and biomass burning) emissions, lightning NOx emissions, photochemical production, deep convection, stratosphere-troposphere exchange (STE), as well as intercontinental transport. At lower-latitude stations (Sanya and Hong Kong), the vertical extent of low O_3 mixing ratios (~30 ppbv) in the lower troposphere (LT) increased with time, reflecting clean marine air masses of tropical origin and the onset of the Asian summer monsoon. At higher-latitude stations (Taipei and Lin'an), elevated O_3 mixing ratios were often observed in the LT in the northeasterly continental outflow. In the upper and middle troposphere (UT/MT), a prominent feature was the frequent occurrences of high O_3 mixing ratios shown as tongues extending down from the lower stratosphere (LS) or isolated layers at all stations. Low relative humidity (RH<10-20%) accompanying with most of these tongues or layers implied that they may have originated from the UT/LS in very recent history. The model largely captured the observed pattern of day-to-day variability in tropospheric O_3 mixing ratios at all stations, but often underestimated those tongues or isolated layers of O_3 enhancements observed in the UT/MT, especially at low-latitude stations. Model calculations suggested that underestimated contributions of lightning NOx emissions and/or stratospheric O_3 are likely responsible for this deficiency of the model. We found that tropospheric O_3 along the Southeast China coast was mainly produced within Asia due to anthropogenic, biomass burning and lightning NOx emissions. Stratospheric influence and lightning NOx emissions were responsible for major events of high O_3 observed in the UT/MT at all stations. The stratosphere made major contributions to O_3 in the UT at Tengchong (24%) and Lin'an (33%), two stations located close to the subtropical jet. Lightning NOx emissions in South Asia and equatorial Africa had major influences on tropospheric O_3 over South China, especially in the low-latitude UT/MT. This study emphasized the need for improved understanding of lightning NOx emissions and STE over the Eurasian and African continents and for better representation of these processes in current global models.

P-1-057 Combining GOME-2 and OMI NO_2 products over China with ground-based measurements and global modeling

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Key words
Satellite NO_2 China

Satellite remote sensing provides tropospheric NO_2 column measurements with global coverage and high spatiotemporal resolution. Concurrent measurements from different satellite sensors, GOME-2 aboard MetOp-A, and OMI aboard Aura, at different overpass times, 9:30 local time for GOME-2, and 13:30 for OMI, offer the opportunity to observe the diurnal variation of the NO_2 columns at different regions.

This study will present a tropospheric vertical column density (VCD) retrieval algorithm that improves over the TEMIS NO_2 products(available at the http://www.temis.nl) for the Eastern China region by using a more accurate air mass factor (AMF) calculation. In this retrieval, the slant and the stratospheric columns are obtained from the TEMIS dataset which is itself based on the DOAS technique and a retrieval-assimilation-modeling approach. The AMF is calculated using the a priori vertical profiles from the IMAGESv2 global chemistry-transport model weighted by altitude-dependent scattering weights computed with the LIDORT radiative transfer model, and combined with a high-resolution climatological albedo database based on five years of MODIS measurements. Due to the lack of high quality cloud products for near-cloud-free scenes, no cloud correction will be applied in the retrieval and only cloud free scenes will be considered. The above settings are chosen so as to guarantee a high level of consistency between GOME-2 and OMI retrievals.

The resulting tropospheric NO_2 VCDs will be compared with the TEMIS products and a sensitivity study will be conducted to investigate the impact of different retrieval parameters. A state-of-the-art Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) instrument, which is designed and operated by BIRA and installed at Beijing (40 N, 116 E, July 2008-April 2009) and Xianghe (40 N, 117 E, March 2010-present) will be employed in the validation of the satellite data. NO_2 VCDs simulated with IMAGESv2 will be used to compare with the satellite columns to examine and interpret the diurnal and seasonal variation over China.

P-1-058 Trends and effectiveness of emission control of sulfur dioxide in China: a satellite perspective

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Key words
Satellite SO_2 retrievals Trend analysis SO_2 emission control

The combination of two satellite instruments (OMI and SCIAMACHY) provides high quality space-borne measurements for the trend analysis of sulfur dioxide (SO_2) column density. An improved product of SO_2 retrievals from these two satellites was derived with the consistent local air mass factor (AMF) algorithm which converts the line-of-sight ‘slant’ columns to vertical columns for the period of 2003–2011. The local AMF was calculated using altitude-dependent scattering weights computed from a radiative transfer model (LIDORT) with high-resolution a priori parameters, weighted by relative vertical SO_2 profiles (shape factor) determined locally with a global atmospheric chemical model (GEOS-Chem). The derived vertical columns and modeled vertical SO_2 profiles were compared to measurements from aircraft campaigns in China. The AMFs for retrievals of the two satellites have distinct inter-annual variations, which would have impacts on the trend analysis of SO_2 columns. Trends of the long-term SO_2 columns showed discrepancies between different regions in China: SO_2 columns increased fast during 2003–2007 and then decreased by ~30% in 2011 in North China Plain; SO_2 columns in Southwest China showed no sign of decrease except for during the global economic recession (mainly occurred in 2008¬¬–2009). The trends of SO_2 columns were further compared to a unit-based power plant emission inventory to evaluate the effectiveness of power plant SO_2 emission reductions related to the wide-spread installations of flue-gas desulfurization (FGD) devices since 2005 in China. The combinations of NO_2 and SO_2 measurements were used to examine the operation and efficiency of the FGD devices in power plants.

P-1-059 Ground-based Continuous Monitoring and Analyses of Total Reactive Odd-nitrogen Species (NOy) and Total Nitrates at Three Sampling Sites in remote areas located in the East China Sea and in the Japan Sea

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Key words

Total Reactive Odd-nitrogen Species (NOy) Total Nitrates Continuous Monitoring Trans-boundary Transport

Recent increase of economic activities in the East Asian countries has brought environmental impacts to the Western Pacific Rim region through the transboundary transport of atmospheric pollutants, such as nitrogen oxides. We developed a continuous monitoring system for NOy and total nitrates (= gaseous nitric acid HNO_3(g) + particulate nitrates NO_3(p); abbreviated as $T.NO_3$), and started the monitoring of these quantities since November 2008, November 2009, and March 2008, at Fukue, Suzu, and Hedo, respectively. The location of the sites are as follows; Fukue site: 32.8°N and 128.7E in Fukue Island, Suzu site: 37.45°N and 137.36E at the tip of Noto Peninsula, Hedo site: 26.68°N and 128.15E near the northernmost point of main island of Okinawa. All sites are far from populated areas and rarely affected by the local pollution. Also the sites are in the East China Sea, and in or facing to the Sea of Japan. Therefore, there are no large emission sources of nitrogen oxides between the sites and the Asian continent.

The results show that the annual Average of NOy and $T.NO_3$ concentrations at Fukue are higher than the other two sites. This result can be explained by the shortest distance to the monitoring site from the large NOx emission sources on the continent, such as Beijing, Shanghai, Seoul, etc.. Comparison of the seasonal variation at the three sites has been done. At Fukue, NOy showed maximum and minimum concentrations during winter to spring and during summer, respectively, while $T.NO_3$ showed spring maximum and summer minimum. Hedo site had similar seasonal variation to Fukue. On the other hand, the variation at Suzu was different from these two sites. Seasonal variability of both NOy and $T.NO_3$ was small and seasonal variations were not definite at Suzu, except definite minimum in January. It should be also noteworthy that the concentrations of NOy and $T.NO_3$ in summer at Suzu were higher than those at Fukue. In Japan, north-westerly winds are prevailing in winter and in early spring, indicating the continental air mass dominates over Japan. Polluted air mass from the continent must arrived at Suzu in winter and in early spring as well as at the other two sites.

Investigation on these characteristic features of the seasonal change at the sites was done by combination of the monitoring data the backward trajectory analysis. Air mass origins were predicted by the NOAA HYSPLIT 4 model and classified to several domains for each sampling site. At Fukue and Hedo, high NOy concentration were observed in continental air mass origins. While, at Suzu, the air mass originated from Japan, which is mainly the air mass passing over Japan originated from the Pacific prevailing in summer, had high NOy concentration. This explains why the summer-minimum in the seasonal variation for the NOy concentration found at Fukue and Hedo was absent at Suzu.

Detailed analyses for the concentration change and the process during the transport have been done and will be reported in the presentation.

P-1-060 Particulate and gaseous pollutants over China adjacent seas in spring and early summer of 2011: implication of long range transport

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Key words
particulate pollutants gaseous pollutants China adjacent seas long range transport

To investigate the air pollution over adjacent seas of China, $PM_{2.5}$ and PM_{10} mass concentration, black carbon (BC), and particle number size distributions with the size of 15 to 760 nm, and gas concentrations were measured on board of a research vessel, Dongfanghong 2, during two cruises in Yellow Sea and East Sea of China in spring and early summer of 2011. Atmospheric particles were also collected by a aerosol sampler simultaneously. First of all the contamination of ship emission was excluded based on the following criteria: detection of“burst” (sudden increase) in number concentration of particles smaller than 100 nm, together with gases concentrations such as SO_2 and NO. After that both the characteristics of marine aerosol and long range transport of continental pollution were discussed based on the rest of the data. The average SO_2, NO, O_3, CO and CO_2 concentrations were 3.6 ppb, 0.92 ppb, 54.6 ppb, 0.37 ppm and 381 ppm in spring (the first cruise, in March and April), which were higher than those in early summer (the second cruise, in May and June) with the concentrations of 1.8 ppb, 0.38 ppb, 50.8 ppb, 0.30 ppm and 407 ppm. The total particle number concentrations in spring were in average around 6000 cm^{-3}. However, the concentration was higher (14000 cm^{-3}) in East China Sea in early summer. Pollutants concentrations in Yellow Sea in the first cruise were higher than those in East China Sea. However, pollutants concentrations in Yellow Sea in the second cruise were lower than those in East China Sea. Back trajectories analysis was used to classify all the sampling days into two groups-effected by marine air mass and continental air mass. The

differences of concentrations between these two groups were considered as the influence from the continental pollution. The results showed that over 50% of the pollution in adjacent seas of China was from coastal area.

P-1-061 Evaluation of trace elements contamination in cloud/fog water at an elevated mountain site in northern China

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Key words
cloud/fog water trace element Mt. Tai air mass Principal Component Analysis

The characteristics of trace elements in cloud/fog water at the summit of the highest mountain — Mt. Tai (1534m a.s.l.) in the Northern China Plain were investigated by a combination of techniques including back trajectory model, regional air quality and dust storm models, satellite observations and principle component analysis. Totally 117 cloud/fog water samples within 22 cloud/fog events were collected during three sampling periods in 2007 — 2008. The elemental concentrations of Zn, Al, Mn, Fe, Pb, Cu, Ni, Cr, As, and Cd were determined by Inductively Coupled Plasma Mass Spectrometry, with stringent quality control measures. Higher elemental concentrations were found at Mt. Tai compared with those reported by other overseas studies. The trace element Zn was dominated in the samples, with volume-weighted mean concentration of 249.1 g l-1. The larger proportions and higher concentrations of toxic elements such as Pb and As in cloud/fog water compared with those in rainwater at Mt. Tai suggests higher potential hazards of cloud/fog water as a source of contamination in polluted areas to the ecosystem. Peak concentrations of trace elements were frequently observed during the onset of cloud/fog events when liquid water contents of cloud/fog water were usually low and large amount of pollutants were accumulated in the ambient air. Inverse relationship between elemental concentrations and liquid water contents of the cloud/fog samples were only found in those with high electrical conductivities and liquid water contents lower than 0.3g m^{-3}. Affected mainly by the emissions of steel industries and mining activities, air masses transported from south/southwest of Mt. Tai were frequently associated with higher elemental concentrations. The principle component analysis suggested that the trace elements were mainly originated from anthropogenic sources, although crustal sources contribution could significantly influence the composition of cloud/fog water samples during the dust storm events. The element Mn is attributed to play an important role in the acidity of cloud/fog water. The composition of cloud/fog water influenced by an Asian dust storm event was reported, which was seldom found in the literature.

P-1-062 The Mediterranean summertime ozone maximum: A satellite and model perspective

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Key words

tropospheric ozone satellite air quality

The Mediterranean troposphere exhibits a marked and localised summertime ozone maximum, which has the potential to strongly impact regional air quality and radiative forcing. The Mediterranean region can be perturbed by long-range pollution import from Northern Europe, North America and Asia, in addition to local emissions, which may all contribute to regional ozone enhancements. We exploit ozone profile observations from the Tropospheric Emission Spectrometer (TES) and the Global Ozone Monitoring Experiment-2 (GOME-2) satellite instruments, and an offline chemical transport model (TOMCAT) to investigate the geographical and vertical structure of the summertime tropospheric ozone maximum over the Mediterranean region. We evaluate the model's ability to capture this feature using both surface observations and satellite data, and subsequently quantify contributions to the ozone maximum from anthropogenic and natural volatile organic compound (VOC) emissions, anthropogenic NOx emissions, wildfire emissions and long-range import of ozone and precursors. Our results suggest a dominance of natural VOC emissions on ozone in the Mediterranean basin over anthropogenic VOC emissions. However, local anthropogenic NOx emissions are the overall dominant contribution to near-surface ozone. We also show that in the lower troposphere global VOC emissions account for 40% of the VOC contribution to ozone in the region, whereas, the majority of the ozone from NOx emissions comes from local sources (90%). In the mid and upper troposphere almost all of the ozone comes from long range transport for all emission sources. Our results allow improved understanding of the large-scale processes controlling air quality and climate in the region of the Mediterranean basin.

P-1-063 Estimation of dry deposition fluxes of dust particles over Yellow Sea: total PM_{10} and size distribution

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Key words
Asian dust deposition fluxes Yellow Sea PM10 size distribution

Using PM10 concentrations derived from API data at Qingdao in China and observed PM10 concentrations at Jeju in Korea during 2001-2007, we investigated PM_{10} dry depositions velocities (Vd) and fluxes during dust events in spring over Yellow Sea from 2001 to 2007 using an exponential method. The Vd of PM_{10} during dust events over Yellow Sea had a large variation and ranged from 0.19 cm s^{-1} to 8.17 cm s^{-1}, with an average of 3.38 cm s^{-1}. Differences of dust transport routes and intensities among different dust events may partly contribute to the wide range of the Vd values of PM_{10} over the Yellow Sea during dust episodes. Dry deposition fluxes of PM10 over the Yellow Sea ranged from 68.5 to 2647.1 mg m^{-2} d^{-1}, with an average of 545.4 mg m^{-2} d^{-1}. Compared with previous studies, it was shown that average deposition fluxes during dust events estimated in our study were about 2-10 times higher than those in the whole spring by other studies, suggesting that much more aerosols were deposited into the Yellow Sea during dust events.

Deposition fluxes of different size bins of dust particles over Yellow Sea were also investigated in this study, based on an improved Williams' model and observed size-segregated concentrations of PM_{11} in Beijing and Qingdao in China and Fukuoka in Japan. The dry deposition fluxes increased with the increasing size in the coarse particle size range but no evident variations occurred in the fine mode. The dry deposition fluxes of the coarse particles increase evidently during dust periods, but the relative contributions of the coarse particles to that of PM_{11} do not change significantly regardless of the occurrence of and absence of dust events. Coarse particles are the main contributor to the PM_{11} dry deposition in springtime over the Yellow Sea, and the percentage reaches over 94.2%.

Our results suggested that the dust events have an import effect on atmospheric input into the Yellow Sea. High deposition fluxes during dust events may results in excessive nutrients loading on dust entering the seas, which may further affect marine biochemistry and ecosystem.

P-1-064 Evidence for ENSO influence on Arctic CO inter-annual variability through forest fire emissions

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Key words
Arctic Carbon Monoxide Biomass Burning ENSO

The Arctic's surface temperature has increased more than the global average over the past few decades. Emissions from anthropogenic and natural sources can lead

to the formation of greenhouse gases, such as ozone (O_3), which may affect this climate sensitive region. It is therefore important to understand the major sources of pollution in the Arctic for mitigation purposes and to understand how the Arctic climate might change in the future. Anthropogenic emissions from Europe, Asia and North America have been found to be exported to the Arctic. In addition, boreal forest fires are thought to be a significant source of pollutants in the summer months. The expected increase in anthropogenic emissions and changes in the frequency of boreal fires will have important implications for the Arctic troposphere and future climate.

We have used a 3D chemical transport model and carbon monoxide (CO) as a tracer of anthropogenic and natural emissions in order to understand the major sources of pollutants in the Arctic region. In conjunction with surface observations over the period of 1998-2009, we have studied the processes which control the observed inter-annual variability (IAV) of CO with particular focus on transport pathways and biomass burning emissions. Possible links between the El Niño-Southern Oscillation (ENSO) and its influence on fires through precipitation and temperature have also been investigated.

P-1-065 Secondary Organic Aerosol Formations from the Emissions of Betel Nut and Joss Paper Burning

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Key words

secondary organic aerosol smog chamber biogentic VOCs biomass burning

The present chamber study is part of an integrated project “Taiwan Megacity Environmental Research”, focusing on the potential impacts of secondary organic aerosols (SOA) on air quality, regional climate, and human health in Taiwan megacity. The objective of this study is focused on investigating the formation of SOA from biogenic and biomass burning precursors within the controlled environmental facilities. In the biogenic SOA study, a plant chamber (2.5 m^3, stainless steel and glass) and a photochemical smog chamber (4.5 m^3, Teflon film) were constructed to evaluate the effects of temperature and light intensity on SOA formation from VOCs emitted by betel nut (Areca catechu) and other plants.

Isoprene was found to be the major component of biogentic VOCs from betel nut, and the emission rates depend on temperature and light. Betel nut exhibited a temperature optimum for isoprene emissions of 45 degree Celsius. The VOCs emitted from plants were transferred directly from plant chamber to smog chamber for further photochemical reactions. VOCs photochemical reaction experiments were performed at RH < 30% and at range of 20 to 45 degree Celsius. SOA particle number and size distributions in the smog chamber were measured using an SMPS system. Gaseous product formation and ozone were monitored at different elapsed times during each reaction. Results shows that the formation of SOA generated from photochemical reaction of betel nut emission were affected by temperature, light intensity and OH radical conditions. In the biomass burning SOA study, two types of combustion smoke (smoldering and flaming) from joss paper burning were filtered and transferred to the smog chamber directly for further photochemical reactions. Benzene and toluene were measured to be the major components of aromatic VOCs from joss paper burning. Results shows that the formation of SOA generated from photochemical reaction of filtered joss paper smoke were impacted by combustion, temperature, light intensity and OH radical factors. Formaldehyde was found to be one of the major gaseous reaction products in both biogenic and biomass burning SOA studies. The co-exposure of SOA, ozone and gaseous reaction products (such as formaldehyde and acetaldehyde) can be one of the major health concerns associated with biogenic and biomass burning SOA formations.

P-1-066 An estimation of future change in surface ozone over East Asia under RCPs scenarios

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Key words

surface ozone chemistry transport model future projection

Since the emissions of ozone precursors described in the Representative Concentration Pathways (RCPs) scenarios are projected to be reduced in the future, we examined future development of tropospheric ozone over East Asia based on the RCP scenarios. For this, future projections of tropospheric ozone under RCPs scenarios done by a global chemistry climate model (CCM) were further down scaled by a regional chemistry transport model (CTM) in order to focus on the East Asian region.

The projected future surface ozone in 2050 decreases from the present (2005) state

over the most of East Asian region under RCPs 2.6 and 4.5 scenarios. Both of the scenarios assume an emission reduction of ozone precursors (NOx and NMHCs) in almost the entire East Asia, which is the main reason for the decrease in surface ozone in the region. Furthermore, the ozone concentration in the upwind regions of East Asia is also decreased in these scenarios, which resulted in the reduced ozone influx into East Asia. On the other hand, an increase of surface ozone is calculated under RCPs 8.5 scenario. This scenario also assumes an emission reduction of ozone precursors in the wide areas in East Asia, but the reduction of precursors is not through enough to achieve a reduction in surface ozone in the whole East Asian region as in the case of the other RCPs scenarios. The increase in surface ozone over East Asian region under RCPs 8.5 scenario is largely influenced by the future change in climate and the transport of ozone outside the East Asia. Reduction in cloud cover and the resulted increase in solar radiation in the future climate in East Asia under RCPS 8.5 scenario is one of the candidates for the causes of ozone increase under this scenario. Frequency distribution (FD) of one-hourly surface ozone in mega cities in East Asia greatly changes in each RCPs scenario. Highly elevated ozone concentration is rarely occurred under each RCPs scenario, and the mode of FD does not change so much in RCPs 2.6 and 4.5 but it increases about 10 to 15 ppbv in RCP 8.5 scenario.

The estimated future surface ozone in East Asia show large dependence on future emission scenarios and the dependence is brought by not only the change in local emission of ozone precursors but also the change in climate and long range transport of ozone.

P-1-067 Ozone and CO in the upper troposphere: model/satellite comparison and sensitivity to emissions and meteorology

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Key words
ozone CO satellite upper-troposphere

Multi-annual simulations with the STOCHEM Lagrangian chemistry transport model are used in conjunction with measurements from the Aura Microwave Limb Sounder (MLS) to examine processes controlling the distributions and budgets of ozone and CO in the tropical and sub-tropical upper troposphere (UT). STOCHEM is driven by meteorology from the HadAM3 atmospheric climate model forced by observed sea surface temperatures and nudged towards ECMWF analyses. Advected model parcels coinciding with the time and position of MLS measurements of UT ozone and CO are identified and treated as quasi measurements. This analogous processing of the simulated and measured data facilitates model/measurement comparison and minimises sampling biases. The impact of nudging on the simulated ozone and CO distributions is assessed by comparing fields from both the nudged and free-running model with observations. Variant model runs for the MLS era isolate different contributors (biomass burning, lightning NOx, isoprene, and stratosphere-troposphere exchange) to the ozone and CO budgets of the UT region. The results highlight the key role of lightning NOx in the UT ozone budget despite the lightning source being small relative to other NOx sources. The year to year variation in meteorology is found to be a larger driver of interannual variability in UT ozone and CO than year to year variation in biomass burning emissions. It is shown that a large proportion of ozone variability in the UT arises from stratosphere-troposphere exchange, and that correctly modelling the exchange is a major challenge in enabling useful comparison of ozone simulations and observations in the near-tropopause region.

P-1-068 Secondary Aerosol Formation in Sydney, Australia

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Key words
Secondary Aerosol Photochemistry

Particles are currently one of the most significant pollutants with respect to human mortality and morbidity. Reduction in particle concentrations through source regulation is challenging due to the large number particle sources (both natural and anthropogenic) present in an airshed, and the wide range of particle sizes and chemical species emitted. Secondary particles can make a significant contribution to total particle exposure, particularly in the fine size fraction which is considered to have the largest impact on health. As secondary particles are generated through

photochemical processes, a reduction in the concentration of secondary particles requires that control mechanisms also consider the relevant gas-phase precursors to these particles.

Climate change projections for New South Wales in Australia suggest significant increases in the frequency of drought, increases in the frequency of hot days and increases in the frequency of high fire risk weather. This has important ramifications for air pollution and health, with atmospheric particle smog severity linked to the frequency of hot, sunny days, and with the highest particle pollution concentrations linked to the presence of bushfire plumes in the Sydney airshed. The development of a long term control strategy for particles in Sydney can be informed through the use of comprehensive three-dimensional simulations of the atmosphere, sources and multi-phase chemistry. However the development of such modelling capability requires a good understanding of the contribution made by local and remote particle sources to the total particle exposure within the region. Such understanding requires detailed and high quality experimental studies.

Stage-I of the Sydney Particle Study (SPS) was aimed at increasing our scientific knowledge of the processes leading to particle formation in Sydney through a comprehensive observation program. Atmospheric observations were carried out at the Westmead Air Quality Station between 5 February and 7 March 2011 by groups from CSIRO, NSW Office of Environment and Heritage, Australian Nuclear Science and Technology Organisation, Queensland University of Technology and the Shanghai Institute of Applied Physics. The observation program included the collection of samples for chemical analysis (size-resolved aerosol composition, speciated volatile organic compounds [VOCs] including alkanes, aromatics, carbonyls, isoprene and monoterpenes, acid gases and continuous or semi-continuous measurements of aerosol number size distributions, aerosol mass, aerosol light scattering, aerosol composition, gaseous criteria pollutants oxides of nitrogen, carbon monoxide, sulfur dioxide and ozone [NOx, CO, SO_2, O_3]).

Secondary particles were observed to form on approximately half of the days of the study, despite evidence that photochemical reactions occurred on most days. The presence of secondary particles and SOA was associated with higher concentrations of SO_2, suggesting that acid catalysis promotes the partitioning of oxidised organic compounds to the particle phase. There was indirect observational evidence supported by modelling suggesting that the majority of the SOA mass is generated from biogenic emissions.

Stage-2 of the SPS includes an observational program at the Westmead Air Quality Station between 16 April and 14 May 2011 and will target winter emissions associated with domestic wood heating.

P-1-069 Dicarboxylic acids and their stable carbon isotopic ratio in fine aerosols above E. Mediterranean: implications for photochemical aging and SOA formation.

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Key words
Dicarboxylic acids stable carbon isotopic ratio

Dicarboxylic acids and their stable carbon isotopic ratio in fine aerosols above E. Mediterranean: implications for photochemical aging and SOA formation.

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Low-molecular-weight (LMW) dicarboxylic acids (DCAs) and related polar compounds comprise a significant fraction of organic aerosols and can play an important role in atmospheric chemistry and on the radiative forcing of the Earth's climate. These compounds can be generated from primary sources including incomplete combustion of fossil fuels and biomass burning, they are thought to be formed mainly by secondary processes in the atmosphere and have been used as tracers for secondary organic aerosols (SOA) formation. They are also subjected to photochemical degradation in the atmosphere and production via multiphase chemistry. Compound-specific stable carbon isotope analysis (CSCIA) can in addition provide useful insights related to LMW DCAs sources and atmospheric processing.

Here we report molecular and stable isotopic compositions of diacids, ketoacids and dicarbonyls in fine aerosol samples (PM_1) collected in a remote location in the Eastern Mediterranean on a weekly basis during a complete year period (from October 2009 to October 2010; n = 51). Sampling duration was two days and samples have been analyzed for dicarboxylic acids, ketoacids, and dicarbonyls and their stable carbon isotopic ratio ($d^{13}C$). In addition several compounds including main ions, organic and elemental carbon (OC and EC), water soluble organic carbon (WSOC), total carbon and its d13C were analyzed.

Molecular distributions of LMW DCAs were characterized by the predominance of oxalic acid (C2) followed by malonic (C3) and succinic (C4) acids. Concentrations of total diacids ranged from 25–542 ng m^{-3} with oxalic acid being the most abundant diacid (accounting for up to 68% of total diacids). The majority of LMW DCAs presented a clear seasonal variability with higher values during summer indicating an enhanced photochemical formation. However several exceptions were observed such as phthalic, azelaic and glyoxylic acids indicating influence from combustion sources including biomass burning during winter-time. Delta ^{13}C values of oxalic acid ranged from -13.0 to -24 per mil presenting a clear seasonal variability with lower values during winter. Thus d^{13}C of oxalic acid becomes less negative during photochemical degradation of diacids. Sources of dicarboxylic acids, ketoacids, and dicarbonyls in the Eastern Mediterranean atmosphere as well as their atmospheric processing is presented and thoroughly discussed.

P-1-070 Stable carbon isotopic composition of oxalic acid in marine aerosols from the western North Pacific: Implication for photochemical ageing of organic aerosols

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Key words
oxalic acid stable carbon isotopic composition ageing of organic aerosols remote marine atmosphere

Oxalic acid is the most abundant water-soluble organic species in aerosols, which can act as cloud condensation nuclei and thus have an influence on climate change. To better understand photochemical ageing of organic aerosols in the remote marine atmosphere, we collected aerosol samples in the western North Pacific in 2001-2010 on weekly basis at a remote island, Chichijima (27°04'E; 142°13'N), located in the boundary of westerly and easterly wind regimes. Here we present seasonal distributions of dicarboxylic acids (C2-C11), ketoacids (C2-C9) and α-dicarbonyls (C2-C3), which were determined using gas chromatography (GC) and GC/mass spectrometry (MS) techniques. Stable carbon isotopic compositions of diacids were determined for 2006 using GC/combustion/isotope ratio monitoring MS (Kawamura and Watanabe, Anal. Chem., 2004). Oxalic acid was found as the dominant diacid species in all the marine aerosols, followed by malonic (C3) and succinic (C4) acids. Concentrations of dicarboxylic acids, ketoacids and α-dicarbonyls showed higher values in winter/spring and lower values in summer. The winter/spring maximum was explained by enhanced atmospheric transport of polluted aerosols and their precursors from Asian continent to the North Pacific due to the intensified westerly winds in winter/spring. The results of stable carbon isotopic compositions of diacids for 2006 showed relatively high δ^{13}C values of oxalic acid ranging from -18‰ to -2‰. The isotopic ratios were found to increase from winter to summer and to decrease toward autumn. This seasonal change in δ^{13}C values of oxalic acid with a peak in late June should be involved with photochemical ageing of organic aerosols in the marine atmosphere. The ^{13}C enrichment is likely caused by kinetic isotopic fractionation during the photochemical degradation of oxalic acid in the presence of Fe (III), which has been confirmed in laboratory experiment (Pavuluri and Kawamura, Geophys. Res. Lett., 2012). The enrichment of ^{13}C may also be associated with the isotopic fractionation during gas-particle partitioning reaction of glyoxal and glyoxylic acid, which are potential sources of oxalic acid.

P-1-071 Chemical speciation of gas-phase organic carbon emissions between gasoline and diesel vehicles

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Key words

Volatile Organic Compounds Diesel Gasoline

Emissions from vehicles using gasoline and diesel fuel dominate global burdens of anthropogenic gas-phase organic carbon and are critical for determining air quality in most urban areas. We present comprehensive chemical speciation for both gasoline and diesel fuel determined from 52 fuels samples taken in California during the summer of 2010, coincident with the California at the Nexus of Air Quality and Climate Change (CalNex) campaigns. Diesel fuel ranges in size from 7 to 25 carbon atoms with the vast majority of the mass in the 10 to 20 carbon atom range, while gasoline is tightly constrained in the 5 to 10 carbon atom range. Both have large fractions of branched and cyclic alkanes, but gasoline contains a much larger fraction of aromatics and polycyclic aromatic hydrocarbons. We demonstrate compositional consistency between both gasoline and diesel fuel with fleet average measurements in the Caldecott tunnel (Oakland, CA) and ambient measurements at the CalNex-Bakersfield supersite, which includes the first in situ measurements of numerous compounds in the intermediate volatility range. Using detailed fuel-based source profiles for diesel exhaust, gasoline exhaust, and non-tailpipe gasoline emissions, we estimate fuel-derived contributions from each source to gas-phase organic carbon in the volatile and intermediate-volatility organic compound ranges, and assess distribution across chemical classes. We compare our results to current inventories to assess the impact of including all of the VOC mass we estimate from gasoline and diesel emissions.

P-1-072 Influence of Humidity on Secondary Organic Aerosol Formation in a Smog Chamber

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Key words

Secondary Organic Aerosol Monoterpenes

Literature show large uncertainties in model predictions for climate change and regional air quality. Previous studies show that it is caused by secondary organic aerosol (SOA) formation incompletely integrated in atmospheric chemical models. Recent findings show that SOA formation is largely affected by humidity and aerosol water chemistry. Particles can be in either crystal or deliquescent form as a function of humidity. The purpose of this work is to investigate humidity effects on SOA formation. It was examined for biogenic organics including alpha-pinene and d-limonene. For this study, a smog chamber facility was constructed, evaluated, and used for SOA formation. An indoor smog chamber was constructed with a FEP bag in a box of aluminum profiles and polycarbonate plates. Highly reflective aluminum sheets were put on inside wall of the box. 60 UVC and 12 UVB lamps on the wall and ceiling were used as light sources. Clean air for the chamber was supplied using a compressor after purification with a cooler, an adsorption dryer, activated carbon beds, and a HEPA filter. The wall loss rate constant of toluene and alpha-pinene was 1.0E^{-3}/min and 0.6E^{-4}/min, indicating that wall loss of organic precursors might be negligible. The wall loss rate constant of particles was measured and used to correct wall loss of particles in SOA formation. It tended to follow theoretical dependence on particle size. Ammonium sulfate particles showed the minimum rate constant of ~2.0E^{-3}/min at 100 nm. Hydrogen peroxide was used as oxidants to examine OH radical initiated SOA formation. SOA formation was conducted at dry (RH < 5%) and humid condition (RH > 85%) with neutral and acidic seed particles. More SOA tended to form at humid condition, although it was complicate to account for humidity effects on SOA yields due to its significant dependence on SOA concentrations. This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MEST) (No. 2011-0000221).

P-1-073 A History of a Stressed Urban Atmosphere: Particle Concentrations, Chemistry, and Perceptions in Newcastle, NSW, Australia

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Key words
particle history Newcastle

Introduction

Newcastle, a small city by global standards, is located on the southeast coast of Australia. It has provided the heavy industrial and coal export base for the state. Throughout most of its history, black coal has been the fuel base. As a result of uncontrolled emissions, air quality over the city has been poor. This paper highlights aspects of particle (PM) pollution and particle chemistry during the history of Newcastle's development, incorporating public and local government perceptions of the problem.

Methods

Targeted historical periods were used to illustrate PM problems in the atmosphere. These included 1850-1860 (introduction of rail, beginning of industry); 1880-1890 (boom development years, coal and rail expansion); 1915-1922 (start of iron and steel manufacturing); 1935-1947 (pre-war and war growth and problems); 1955-1970 (beginning of measurement, clean air legislation); 1985-1995 (modern measurements, emissions inventories); post-1999 (the demise of iron and steel). Particle emissions from various sources were estimated using methods from the Australian National Pollution Inventory and the NSW Greater Sydney Pollution Inventory. Perceptions were determined from a range of sources, including newspapers; and government, industry and chamber of commerce reports.

Results and Discussion

Compared to current atmospheric conditions, emissions and concentrations of PM during the periods since the 1880s through the 1990s showed a stressed atmosphere, especially after the introduction of iron and steel manufacture (1915), but before any real attempt to legislate pollution control (1962). For example, major sources of PM emission during the late 1930s included heavy industry, power stations, steam locomotives, steam ships, light industry and residential heating in winter. Emissions levels reached more than ten times those of today. For most of the first half of the 20th century, the perception that air pollution indicated economic prosperity prevailed. But from the 1940s onward, came increasing realization that high PM concentrations created health and aesthetic problems, and Newcastle residents began to demand pollution control. After the NSW Clean Air Act became law, PM concentrations fell slowly but steadily. A major reduction in PM emissions, and the change on PM chemistry, occurred until 1999 when the iron and steel industry closed. Overall, variations in PM emissions depended largely on the economy, and the rise and fall of industry and coal mining.

Conclusions

Using PM as an example pollutant, this study demonstrates the role of uncontrolled emissions in creating a stressed atmospheric over Newcastle, NSW, during much of its development history. The change in public perception from the concept that air pollution indicated a strong economy and workforce, to demands for pollution controls for a better living environment, began in the 1940s. The development of Clean Air legislation, and the increasing use of measurements to quantify PM problems, created a steady decrease in PM emissions and concentrations between the 1960 and the 1990s. The close of the iron and steel industry in Newcastle in 1999 created an immediate improvement in air quality.

P-1-074 Effects of climate interannual variability to summertime surface ozone concentrations in the southeastern U.S.

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Key words
surface ozone isoprene nitrate chemistry climate interaction

We used the GEOS-Chem chemical transport model to simulate summertime surface ozone over the southeastern U.S. during 1987 to 2010 and compared the results to CASTNET measurements, with the goal of examining the model sensitivity of surface ozone to climate interannual variability. The model was driven by assimilated meteorology to eliminate the uncertainty associated with climate simulation. We found that the standard model overestimated afternoon surface ozone concentration in southeastern U.S. in summer by 10 to 30 ppb. At the same time, the model significantly underestimates the sensitivity of surface ozone to surface temperature. Further sensitivity tests were conducted to test the effects of isoprene nitrate recycling, additional carbonyl chemistry, nighttime chemistry, model resolution, as well as the interannual variability of biogenic and biomass burning emissions.

P-1-075 Monitoring the progress of the Montreal Protocol in East Asia

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Key words
Montreal Protocol East Asia

The Montreal Protocol is perhaps the most successful international environmental treaty, responsible for global phase-out of the consumption and production of ozone-depleting substances (ODSs), e.g., chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs) especially in developed countries (non-Article 5). The phase-out of CFCs and HCFCs might increase the use of HFCs as substitutes, which do not deplete the ozone layer but, along with CFCs and HCFCs, are greenhouse gases that contribute to the radiative forcing of climate. However, in developing countries (Article 5) due to less demanding phase-out

schedule, the emission changes of ODSs from developing countries are of special interest, but especially in China, in the fast growing economy. Recent reports show that China's efforts to decrease emissions of primary ODSs have been successful, leading to increased emissions of replacement HCFCs and HFCs.

In this study, we estimated the emission rates of anthropogenic halocarbons, which include CFC-11, CFC-12, HCFC-22, HCFC-141b, HCFC-142b, HFC-23, HFC-134a, HFC-32, HFC-125, HFC-152a, and HFC-143a, for each country (China, Korea and Japan) in East Asia between 2008 and 2011 based on "top-down" interpretation of atmospheric high-precision in-situ at Gosan site, a remote island site on Jeju Island in the yellow sea south of the Korean peninsula. Ambient concentrations of halocarbons have been analyzed every two hours using the "Medusa" cryogenic pre-concentration system with gas chromatograph and mass selective detector (GC-MSD) as part of the Advanced Global Atmospheric Gases Experiment (AGAGE) network. Both air-mass back-trajectory analysis and a tracer method based on the HFC-23/HCFC-22 ratio were used to identify periods of air transport from each country.

Between 2008 and 2011, the emission strengths (slope with HCFC-22 for China and Korea, but HFC-134a for Japan) of halocarbons were analyzed for each country. While the emissions strengths for CFCs are found to have decreased for Korea and Japan about 50%, the strengths of CFCs have increased about 20% between 2008 and 2010 -2011. The HCFCs are more interesting, because emission strength of HCFC-141b have increased about 48% and decreased about 22% for HCFC-142b in China, suggesting more effort is needed to further reduce HCFC-141b emissions in China. On the other hand, the emission strengths of HCFCs remained at consistent levels between 2008 and 2011 for Korea and Japan. The emission strengths of HFCs are have increased for each country, combined with significant contributions of HFC-32, HFC-125, and HFC-134a. However, emission strength of HFC-23, which is a powerful greenhouse gas coproduced during HCFC-22 production, have decreased about 22% in China. The emissions rates of target halocarbons will decrease/increase in accordance with emission changes of HCFC-22 in China and Korea and HFC-134a in Japan between 2008 and 2011. We need to long-term continue monitoring for CFCs, HCFCs and HFCs as examining the regulation under Montreal Protocol in East Asia.

P-1-076 Aircraft measurements of nitrate radical and dinitrogen pentoxide using cavity enhanced absorption spectroscopy

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Key words

Aircraft instrument Nitrate radical Dinitrogen Pentoxide Heterogeneous uptake Ambient Aerosol

We report our recent development of an aircraft instrument measuring two important night-time species, nitrate radical and dinitrogen pentoxide as well as nitrogen dioxide, water vapour and aerosol extinction using cavity enhanced absorption spectroscopy. Results from the deployment of the instrument on the UK FAAM BAe-146 atmospheric research aircraft both in July, 2010 and Jan, 2011 are also presented. Night-time processes involving the gas-phase reactions of nitrate radical and the heterogeneous uptake of dinitrogen pentoxide by ambient aerosol particles under typical European context are analyzed using the field data.

P-1-077 Stable carbon and nitrogen isotopic compositions of tropical aerosols from Tanzania, East Africa: Sources and contribution of C3 and C4 Plants

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Key words
Carbon, Nitrogen, Isotopes, Aerosols particles, Sources, C3 and C4 plants

Stable isotopic ratios of carbon and nitrogen can provide complementary information about sources of aerosols and have long been applied in various environments around the world. However, little is known regarding levels and source implication of isotopic composition of aerosol total carbon and nitrogen in Africa. Here, we report for the first time the delta ^{13}C and delta ^{15}N data in Tanzanian aerosols as well as concentrations of total carbon and nitrogen in $PM_{2.5}$ and PM_{10} aerosols. Their seasonal variations and contributions of burning C3 and C4 plants to the aerosols are also discussed. Aerosol samples were collected on 24 h basis at a rural site (Morogoro) in Tanzania using $PM_{2.5}$ and PM_{10} low volume samplers and quartz filters in parallel. Filter samples were analysed using elemental analyzer (EA) and EA/isotope ratio mass spectrometer (IRMS). Mean TC concentrations in $PM_{2.5}$ were 5.4±1.6 μgC/m^3 in wet season and 7.7±2.0 μgC/m^3 in dry season whereas those of PM_{10} were 7.7±2.0 μgC/m^3 and 11.0±4.0 μgC/m^3 in wet and dry seasons, respectively. TN showed a mean content of 0.5±0.1 μgN/m^3 in $PM_{2.5}$ and 0.7±0.1 μgN/m^3 in PM_{10} in wet season and 0.9±0.3 μgN/m^3 in both size fractions in dry season. No seasonal differences were observed in TC and TN contents. On average TC account for 19.4% and 21.2% of $PM_{2.5}$ mass in wet and dry seasons, respectively while they account for 16.1% in wet season and 18.1% in dry season of PM_{10} mass. TN comprised on average 2.2% of $PM_{2.5}$ mass and 1.6% of PM_{10} mass.

Delta ^{13}C of TC ranged from –26.1 to –20.6‰ in $PM_{2.5}$ and –24.4 to –22.4‰ in PM_{10} whereas delta ^{15}N ranged from 13.4–22.1‰ and 10.4–18.7‰ in $PM_{2.5}$ and PM_{10}, respectively. Strong positive correlations were found between delta 13C and

TC in dry season, and levoglucosan (and nss-K^+) and TC in both seasons, suggesting that possible sources of organic aerosols were condensation of volatile and semi-volatile organics derived from biomass burning including residential wood combustion on pre-existing particles. A very strong positive correlation was obtained between TN and NH_4^+ and also NO_3^-, indicating that organic nitrogen is probably less important in the Morogoro aerosols. The contributions of C3 and C4 plants to TC were 59% and 41% in $PM_{2.5}$ and 59% and 41% in PM_{10}, respectively. In Tanzania, where large areas of its land coverage are covered with Savannah vegetations, contributions of biomass burning of C3 and C4 plants to ambient aerosols may not be easily distinguished. However, the present results clearly indicate the mixture sources of biomass burning of C3 and C4 plants.

P-1-078 A high-resolution emission inventory for coal-fired power plants in China, 1990-2010

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Key words
power plants emission China

A new emission inventory of China's coal-fired power plants with high spatial and temporal resolution is developed for the period of 1990-2010, based on detailed information of unit-level, including capacity, technology, fuel consumption, location, and the time it came into operation and shut down. The high-resolution emission inventory can replace the data currently used in chemical transport models, and allows a close examination of temporal and spatial variations in power plant emissions in China and their driving forces during last two decades. Emissions from China's coal-fired power plants in 2010 were estimated as follows: 8.02 Tg SO_2, 9.05 Tg NOx, 3091 Tg CO_2, and 0.89 Tg $PM_{2.5}$, representing a growth of 112%, 307% and 484% and a decline of 26% from 1990, respectively, compared to 558% growth of power generation during the same period. SO2 emissions were peaked in 2006 at 16.19 Tg, and then decreased by 50% between 2006 and 2010, as the subsequence of installation of flue-gas desulfurization (FGD) equipment. Although low-NOx burners have been widely installed in power plants after 2006, but it failed to curb the increase trend of NOx emissions. $PM_{2.5}$ emissions fluctuated during the past 20 years, as reduction gained from emission control equipment was always offset by increased coal usage. An anomaly of monthly variations in emissions was found during 2008-2009 compared to other

years, reflecting the impact of financial crisis on industrial activities.

P-1-079 Injection heights of springtime biomass burning plumes in Peninsular Southeast Asia and the impacts on pollutant transport

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Key words
biomass burning injection height Peninsular Southeast Asia MISR GEOS-Chem

We analyzed observations from the Multi-angle Imaging SpectroRadiometer (MISR) to determine the injection heights of biomass burning plumes over the Peninsular Southeast Asia (PSEA) in spring and evaluated the impacts of injection heights on pollutant transport. Twenty-five thousand MISR pixels from six hundred smoke plumes during February to April for the years 2001-2010 were analyzed. Forty percent of the analyzed smoke pixels were injected above the local mean boundary layer height (1 km) at MISR overpass time (10:30 local time). We used the GEOS-Chem model to simulate the transport of springtime biomass burning pollutants from PSEA using the injection heights based on MISR. With 40% of biomass burning emissions injected directly between 1-5 km, CO and BC concentrations at 3 km over PSEA increased by 7% and 90%, respectively, compared to a control simulation where all biomass burning pollutants were released in the boundary layer. NOx concentrations in the free troposphere (2~4km) increased by 70~120%, resulting in significant enhancements of PAN (20~40%) and O_3 (7~10%) in the free troposphere. Over the downwind area of southern China and western Pacific, CO and BC concentrations in the free troposphere increased by 3% and 50%, respectively. NOx, PAN and O_3 in the free troposphere increased by 25%, 28% and 8%, respectively, due to more efficient transport of PAN from PSEA. We compared model results to Trace-P aircraft observations over western Pacific during March 2001. In addition, the simulated O_3 concentrations were better correlated with observations in the free troposphere when there were biomass burning events. Our results show that the injection height of biomass burning plumes can significantly impact O_3 formation in downwind areas due to efficient long-range transport of PAN.

P-1-080 Aerial observation and Analyses of Nitrogen Compounds over the East China Sea

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Key words
Nitrogen oxides Nitric acid Aerial observation Aerosol

Recently, anthropogenic emission of nitrogen oxides in East Asia has been increasing due to the rapid economic growth. Nitrogen oxides have high reactivity and are oxidized in the atmosphere during transport to generate gaseous nitric acid (HNO_3(g)) or particulate nitrate (NO_3^-(p)) , which causes acidification of earth's environment. In order to clarify the process of long-range transport involving nitrogen oxides, aerial observations were carried out between Japan and the Asian continent as well as ground observations.

The aerial observations were conducted on October 14, 15 and 17, 2009 (autumn) and December 11, 12 and 14, 2010 (winter) and March 10, 11, 13 and 14, 2012 (spring). The tracks of the level flight were set between Fukue Island located at the west end of Japan and the point above the East China Sea located about 220 km south of Jeju Island, Korea. Circular flights were also performed above Fukue Island. Flight altitudes were 500, 1000, 2000 and 3000 m in both flight types. We measured gaseous total odd nitrogen species (NOy(g)) , HNO_3(g) , O_3, SO_2, CO, black carbon (except in 2009) , aerosol number density, aerosol mass concentrations, bioaerosol and ionic and metal components in particles. In this presentation, analyses of total odd nitrogen species (NOy) , HNO_3(g) and NO_3^-(p) are mainly reported.

NOy(g) concentrations decrease with altitude for all the flights. O_3 concentrations decrease and increase with altitude in 2009 and 2010, respectively. NOy(g) concentration levels have no obvious differences between 2009 and 2010, although HNO_3(g)/NOy(g) ratios in 2010 were smaller than those in 2009. Such differences would be caused by the difference of solar radiation intensity. Solar radiation intensity in winter (2010) was weaker than in autumn (2009) and then the photochemical activity must be reduced in 2010.

We investigated relationship between O_3 and NOy—T.NO_3 (T.NO_3 = HNO_3 + NO_3^-(p)). A positive correlation (R = 0.69 ~ 0.80) was shown in 2009. This indicates that secondary photochemical pollutants, such as PANs, would be main component of NOy in 2009. Meanwhile, NOx (= NO + NO_2) fractions in NOy would be large in 2010, because a negative correlation of O_3 with NOy—T.NO_3 was observed (R = -0.84 ~ -0.44).
The uptake of HNO_3 by particles was investigated. Correlations of Na^+, nss-Ca^{2+}, NH_4^+, Fe and Al with NO_3-(p)/T.NO_3 were examined. All the flights in 2009 and 2010, NO_3-(p)/T.NO_3 with Na^+ showed a good correlation (R2 = 0.41~0.91) and it means the uptake of HNO_3 on the surface of sea salt aerosol is the main formation process of NO_3^-(p). When the dust event was observed (10/17 and 12/11), the NO_3^-(p)/T.NO_3 ratio became higher. This implies that the uptake of HNO_3 on the surface of dust particles also contributes to NO_3^-(p) formation process. This presentation will also describe results of the observation in 2012.

P-1-081 Numerical simulation of organic aerosol at a rural site near the Taipei City in Taiwan

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Key words
organic aerosol numerical simulation

Comparing with the inorganic aerosols, organic aerosol is still poorly understood due to their complicated formation and growth processes as well as interactions with other type of aerosols. In this study, the Community Multiscale Air Quality (CMAQ) Modeling System is used to simulate the fate of volatile organic compounds (VOCs) and other pollutants in a rural station (Hwalin) and a nearby urban station (Taipei/NTU) in Northern Taiwan. Although the Hwalin station is located in a subtropical forest and has strong sources of biogenic VOC, it may also receive anthropogenic emission from the nearby Taipei City and vice versa. The contrast between these two sites allows us to better understand the relative contribution between anthropogenic and biogenic emissions to the formation of organic aerosols. Here we focus on the formation of secondary organic aerosols (SOA) during a heat wave event in the summer of 2011. Concentrations of VOCs and other pollutants measured during a field campaign and nearby monitoring sites are used to improve the inventories of both anthropogenic and biogenic emissions.

The model simulation is then used to help understanding the photochemical processes and microphysical processes of SOA and inorganic aerosol formation. Special emphasis is put on the diurnal variation of SOA and relevant chemical and meteorological factors.

P-1-082 Analysis of surface ozone variability at the dome-c high altitude antarctic station

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Key words
Surface ozone Antarctica

We present 6-year of surface ozone (O_3) observations at the Italian-French station of Dome Concordia (Dome C) in Antarctica (3233 m a.s.l.). Dome C is located about 1200 km inland from the coast of Terra Nova as well as about 1100 km south of the coast of Adelie Land, on a large dome in the western polar plateau.

Surface O3 measurements are continuously performed by using UV absorption analysers (Thermo TEI 49c and TEI49i). These measurements are carried out at the Clean Air Research Observatory (CARO), located 700 m SW with respect to the main base and upwind to the prevalent wind direction. Air is collected by a 2 m steel inlet placed on the roof of the shelter at approximately 5 m above snow surface. Span and zero checks are daily performed, while intercomparison against transfer standards or calibrated instrument are yearly performed during maintenance campaigns.

During the period January 2006 – December 2011, the monthly O_3 behaviour was characterized by a broad maximum during the austral winter (June – August) of about 34 ppbv, minima in March and October (23 and 27 ppbv, respectively) and a secondary maximum for November – January with average concentrations approaching the winter levels. While the winter O_3 maxima appears to be related with large-scale accumulation processes during the Antarctic night, the spring-summer maximum is related to the occurrence of sudden O_3 increases episodes, possibly related with photochemistry occurring within the Antarctic PBL. In this work, we will analyse the interannual O_3 variability at Dome-C as a function of local atmospheric conditions and large-scale air-mass transport. To this aim, HYSPLIT 3D back-trajectories will be used to categorize the observed O_3 concentrations as a function of synoptic-scale air-mass circulation clusters. Moreover, concurrent flask measurements of halogenated gases will be considered

to further elucidate surface O_3 behaviours at this remote measurement site.

P-1-083 Airborne bacteria in Kumamoto, Japan from autumn 2011 to spring 2012

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Key words
bioaerosol BacLight

As one of major atmospheric aerosol groups, airborne bacteria can link isolated microbial communities and enhance the formation of ice cloud. However, little information about the number concentration of bacteria in the atmosphere is available for quantitative and qualitative investigations of their roles in ecosystem evolution and climate change. In this study, fluorescent staining coupled with enumeration was tested in the urban atmosphere of Kumamoto, Japan to measure the number concentration of airborne bacteria. We used LIVE/DEAD BacLight Bacterial Viability Kit and confirmed that it was able to study the number concentration and the viable status of airborne bacteria.

Observations were carried out from October 2011 to April 2012. The concentration of airborne bacteria varied in the range of 7.6×10^4 ~ 1.4×10^6 cells/m^3 during the whole period and sometimes showed a rapid change from day to day and even in different time on the same day. The concentration of viable bacteria was 6.4×10^4~7.8×10^5 cells/m^3 and that of non-viable bacteria was 1.2×10^4~6.7×10^5 cells/m^3. The viability was 50~90%, suggesting that more than half of airborne bacteria were in a viable state. Diurnal variation of airborne bacterial abundance was found in a few days. There was a close relation between the bacterial concentration and the air pollution, in particular the particulate matters of diameter >2 μm. Correlation between bacteria and other factors such as weather conditions were not confirmed.

P-1-084 Process analysis of $PM_{2.5}$ pollution over the PRD in the fall season using the CMAQ model

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Key words

process analysis $PM_{2.5}$ Pearl River Delta CMAQ

The Pearl River Delta (PRD), one of the most urbanized and industrialized regions of China, frequently suffers from the high levels of fine particulate matter and haze in these years. In this study, the Community Multiscale Air Quality (CMAQ, version 4.7) modeling system was used to simulate the $PM_{2.5}$ episodes over the PRD in November 2009. Integrated process rates (IPR) analysis was used to investigate the influences of various atmospheric processes on the evolution of ambient $PM_{2.5}$ and its components during the episodes, and to diagnose the uncertainties of the simulation as well. The results showed that emissions, transport and aerosol processes dominated the variations of $PM_{2.5}$ levels. The central PRD was characterized by intensive emissions and strong $PM_{2.5}$ exports to other sub-regions of the PRD. Transports and emissions were the main contributors to the changes of levels of element carbon (EC), organic carbon (OC) and sulfate, while transport and aerosol processes dominated the variations of nitrate and ammonium in $PM_{2.5}$. Based on the results at representative sites, the aerosol processes decreased the nitrate concentrations in near-ground layers while increased nitrate levels in higher layers. Compared with the source apportionment results of observation-based methods, the lower contributions of aerosol processes to organic matter suggested an underprediction of secondary organic aerosol, which explains the significant negative bias of the predicted OC against the observations.

P-1-085 Observational determination of removal rates of NO_3 and N_2O_5 using data from the RONOCO (ROle of Nighttime chemistry in controlling the Oxidising Capacity of the atmOsphere) campaign

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Key words
NO_3 N_2O_5 Nighttime Chemistry

At night NO_3, usually rapidly photolysed, can build up to significant concentrations, in thermal equilibrium with N_2O_5, and act as the dominant oxidative species driving nighttime chemistry. Physical or chemical removal of these species can therefore act as an important sink of NOx as well as potentially affecting ozone budgets. The aims of the RONOCO (ROle of Nighttime chemistry in controlling the Oxidising Capacity of the atmOsphere) campaign were to quantify the processes that influence the nighttime chemistry using UK-aircraft measurements combined with modelling on several different scales.

The campaign was a consortium project involving the University of Cambridge, University of East Anglia, University of York, University of Leicester, University

of Leeds and the University of Manchester. Measurements, including NO_3, N_2O_5, NOx, O_3, CO, aerosol, HNO_3, PAN and VOCs, were taken on board the FAAM (Facility for Airborne Atmospheric Measurements) BAe-146 research aircraft during flights around the UK in July-August 2010 and January 2011, with a total of ninety hours of flight measurements.

In this paper we explore different ways in which the rate constants for direct chemical loss of NO_3 and the aerosol uptake coefficient of N_2O_5 can be derived from the measurements. Output from a box model based on the MCMv3.2 (Master Chemical Mechanism) is used to test the efficacy of the methods. We investigate how to appropriately partition the total loss of the sum of NO_3 and N_2O_5 between the individual losses under the different chemical environments experienced.

P-1-086 NORS: Demonstration Network Of ground-based Remote Sensing Observations in support of the GMES Atmospheric Service

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Key words
ground-based remote sensing GMES Atmospheric Service atmospheric composition

The principal objective of the NORS project is to improve the quality and validation of the products delivered by the GMES Atmospheric Service (GAS), using independent ground-based remote sensing data from the international Network for the Detection of Atmospheric Composition Change (NDACC). NDACC is a cross-border research network with a strong European contribution, providing high-quality reference observational data for understanding the physical / chemical state of the stratosphere and troposphere, and for assessing the impact of atmospheric composition changes on climate. NORS focuses on a selection of NDACC data that have high priority in the different domains of GAS, namely 'ozone and UV', 'air quality' and 'climate'. The research planned in NORS aims at tailoring these NDACC products to the needs of GAS. It includes a full characterisation of the products and an evaluation of the consistency between the ground-based data and the satellite data assimilated in the GAS production chain. As ground-based remote sensing data form the ideal link between in situ surface concentration and satellite column data, NORS will investigate how integrated tropospheric products and integrated ozone products can be developed.

The project will demonstrate operational rapid delivery of NDACC data to GAS, including a comprehensive set of metadata and a user guide. It will also develop and implement a web-based server for providing consistent validation reports of the GAS products using the NORS data products, on an operational basis. In support of the re-analyses planned in GAS, NORS will deliver time-series of ground-based data back to 2003.

The achievements of NORS will be made available for NDACC as a whole and especially to candidate NDACC stations filling gaps outside Western Europe.

In the present situation, NORS works in support of the GAS prototype project MACC-II. There is a strong collaboration between both projects.

In this presentation, we will show the progress that was made in the NORS project after its first 10 months of operation.

P-1-087 Dust and Black Carbon in Seasonal Snow on Qinghai-Tibet Plateau

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Key words
Black carbon Qinghai-Tibet Plateau HYSPLIT

Snow is the most reflective natural surface on Earth and its albedo (the fraction of sunlight reflected) can be significantly reduced by small amounts of absorptive impurities such as dust and black carbon (BC) particles. Even it is impossible to measure the albedo change directly, it is significant for climate change and hydrological cycle. A field campaign was conducted on Qinghai-Tibet Plateau in January 2012. Snow samples were collected at 6 sites. The average elevation of these sites is more than 4100m and most sites are far from human activity influence. The absorptive impurities are principally dust and BC particles on Qinghai-Tibet Plateau. The BC content of snow ranges from 60 to 560 ppb with an average of 190 ppb while the dust content of snow needs further investigation which is not included in this paper. The concentration of BC is less than that found in northern China in 2010. The source of BC on Qinghai-Tibet Plateau is simulated by using HYSPLIT.

P-1-088 Single particle characterization of black carbon aerosols in the NE Tibetan Plateau, China

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Key words
black carbon size distribution mixing state cross section carbon monoxide

Continuous mass concentration, size and mixing state of refractory black carbon (rBC) aerosols were measured with a ground-based single particle soot photometer (SP2) at Qinghai Lake (QHL) in the northeastern (NE) Tibetan Plateau, China during October 2011. Average rBC mass concentration (0.36±0.27 μg m^{-3}-STP) was significantly higher than the concentrations measured in background and remote regions of the globe (0.001-0.32 μg m^{-3}), indicating that QHL was contaminated by rBC particles. The mass size distribution of rBC in volume equivalent diameter showed a primary mode peak at 175 nm, with a secondary mode peak at 495 nm. About 36.8% of the observed rBC particles within the detectable size range were mixed with large amounts of non-refractory materials present as a thick coating. Relative high percentage of internally mixed rBC with positive correlation between rBC concentration and RH indicated that rBC might be as one of the main CCN to influence precipitation intensity in this region. Internally mixed rBC fraction showed positive correlation with rBC concentration, which indicated that high rBC pollution (> 0.40 μg m^{-3}-STP) was mainly caused more by regional transport than by local emissions. Backward air trajectories indicated that enhanced rBC concentrations were influenced by central Gansu and eastern Qinghai province, with good accordance with locations of urban areas and industrial centers. Moreover, the results also showed that Pakistan and northern India may be the significant rBC source regions for QHL during the sampling period. The light absorption cross-section of 39.1 m^2 g^{-1} at 880 was 2.4 times higher than Aethalometer default value of 16.6 m^2 g^{-1}, suggesting that other chemical species as rBC coating enhancing the light absorption. High correlation was found between rBC and CO, with a slope of 1.5 ng m^{-3} $ppbv^{-1}$ that was representative of the anthropogenic emission sources in QHL region.

P-1-089 WMO GAW Programme addressing air pollution and climate change

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Key words
atmospheric observations greenhouse gases air quality

The Global Atmosphere Watch (GAW) Programme of the World Meteorological Organization (WMO) is the only programme involved in long-term world-wide observations of the chemical composition and selected physical characteristics of the atmosphere.　GAW is built up as a partnership of contributions from about 80 countries. GAW observations are used to reduce environmental risks to society and meet the requirements of environmental conventions, to strengthen capabilities to predict climate, weather and air quality, to contribute to scientific assessments in support of environmental policy and deliver integrated products and services of relevance to users.

The GAW focal areas are ozone and UV radiation, greenhouse gases, selected reactive gases, aerosols, precipitation chemistry and urban air pollution. Quality assurance/quality control plays a very important role, for example for the quality of greenhouse gas observations especially in relation to climate services. In this respect WMO/IAEA organizes regularly meetings to discuss laboratory and field measurement techniques, calibration and quality control.

Tropospheric ozone changes regarding observations, state of understanding, and model performances have been addressed in a joint workshop. This workshop initiated regional work on the assessment of past and current trends in troposphreic ozone and capacity of the models to reproduce those changes, with the report available from http://www.wmo.int/pages/prog/arep/gaw/gaw-reports.html.

Well known products are the WMO Greenhouse Gas Bulletins published each year in time for the UNFCCC COP meetings and the issuance of Antarctic Ozone Bulletins during the Southern Hemisphere spring. Other areas for application of data are volcanic ash events and sand and dust storms. Recently a survey of ceilometer and lidar stations has been undertaken, with the possibility for use during volcanic events. Important collaboration has taken place with GESAMP on atmospheric input of chemicals to the ocean. A global precipitation assessment is being finalized.

Recently the UNEP/WMO Integrated Assessment of Black Carbon and Tropospheric Ozone has been published. The report shows that reducing the emissions of these two short-lived climate forcers and common air pollutants could slow the rate of climate change markedly over the next half-century. These substances also have harmful effects on human and ecosystem health, thus reducing them will also improve air quality. The GAW SAG Aerosols at the last meeting in October 2011 addressed the issue of black carbon and the challenges for the atmospheric chemistry community to perform the observations required by policy makers.

The GAW Urban Research Meteorology and Environment, GURME, addresses urban problems, including in megacities. These issues are presented in another abstract.

P-1-090 Sources of springtime tropospheric ozone over North China: A modeling analysis of ozonesonde and satellite observations

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Key words
ozonesonde ozone profiles intercontinental transport

Tropospheric O_3 concentrations and emissions of NOx have both increased significantly over China as a result of rapid industrialization during the past decade. These trends degrade local and regional air quality and have important effects on background tropospheric O_3 and surface O_3 over downwind North Pacific and North America. In-situ observations of tropospheric O_3 over China are therefore essential to testing and improving our understanding of the impact of Asian anthropogenic (versus natural) emissions and various chemical, physical, and dynamical processes on both regional and global tropospheric O_3. Despite their critical importance, in-situ observations of tropospheric O_3 profiles over China have been few and far between in most of the country. To investigate the ensemble of processes that control the distribution, variability, and sources of springtime tropospheric O_3 over China and its surrounding regions, an intensive ozonesonde sounding campaign, called Transport of Air Pollutants and Tropospheric Ozone (O_3) over China (TAPTO-China), was conducted at nine locations across China in the springs of 2004 (South China) and 2005 (North China). In this paper, we use a global 3-D model of tropospheric chemistry (GEOS-Chem) to examine the characteristics of distribution and variability and quantify various sources of tropospheric O_3 over North China by analysis of intensive ozonesonde data

obtained at four stations in North / Northwest China during the second phase of TAPTO-China (April-May 2005). These four stations include Xining (36.43N, 101.45E), Beijing (39.80N, 116.18E), Longfengshan (44.44N, 127.36E), and Aletai (47.73N, 88.08E). We drive GEOS-Chem with two sets of assimilated meteorological observations (GEOS-4 and GEOS-5) from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office (GAMO), allowing us to examine the impacts of variability in meteorology. We show that the observed tropospheric O_3 mixing ratios exhibit strong spatio-temporal variability. The model generally simulates well the ozonesonde observations but tends to underestimate O_3 in the upper troposphere over Beijing and Longfengshan. We find that Asian fossil fuel emissions, stratospheric O_3, African lightning NOx emissions, as well as intercontinental transport are the main contributors to tropospheric O_3 over North China in spring. While the lower-tropospheric O_3 is largely influenced by Asian fossil fuel emissions (except over Aletai, Northwest China), lightning NOx emissions have a larger impact on the upper-tropospheric O_3 than Asian fossil fuel emissions (except over Longfengshan, Northeast China). Model simulations suggest that the European fossil fuel emissions contribute more to the lower-tropospheric O_3 over Aletai than the Asian fossil fuel emissions. We will also show that tropospheric O_3 measurements by Tropospheric Emission Spectrometer (TES) aboard the NASA EOS Aura satellite can be used to study tropospheric O_3 variability at Xining.

P-1-091 Soil biogenic NO emission as a strong source for tropospheric NO_2 — A case study for arid regions (Taklamakan desert, Xinjiang, P.R. China)

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Key words
soil biogenic NO emission satellite observation arid area DOAS

The estimate of global soil biogenic NO emissions ranges from 4 to 21 Tg y^{-1}; the huge range is particularly due to the uncertain contribution from semi-arid, arid, and hyper-arid regions. These regions cover approx. 40% of the planet Earth’s total land surface. However, we have only poor knowledge about soil NO emissions from these areas, mainly due to a very small number of measurements in remote desert areas.

DOAS (Differential Optical Absorption Spectroscopy) technique has been applied to measure atmospheric trace gases since decades. It uses distinct wavelength depended absorption of UV-VIS light by molecules for their quantitative measurement. Corresponding satellite-borne observation has global coverage and provides long term data sets, which are of particular interest for remote regions.

We attempted to study the impact of soil biogenic NO emissions from arid soils by analyzing long term (2005-2011) tropospheric NO_2 vertical column densities (VCDs by OMI DOMINO v2.0). Our target area is the Ruoqiang county in the Taklamakan desert (NW China, 38.5-39.5°N, 87.2-89.2°E) which includes desert/Gobi desert soils, three agriculturally managed oases (cotton, Jujube), areas of natural (tree/shrub) vegetation, and man-made constructions (LANDSAT).

The spatio-temporal distribution of tropospheric NO_2 shows that: (1) high VCDs are observed over oases, natural vegetation, and national roads, (2) there are significant seasonal variations (highest during summer, lowest during winter),(3) sudden increases in spring, and (4) springtime VCDs are higher over agriculturally managed oasis-fields as compared to areas of natural vegetation. Furthermore, results of ground based MAX-DOAS measurements (performed in a particular oasis, Milan) compare with corresponding satellite MAX-DOAS results astonishingly well in many cases.

Since regional anthropogenic (industrial) sources of tropospheric NO_2 are very small and do not own a significant seasonal variation (despite they are expected to show the larger (heating) source strength in winter), we deduce that: (1) the main source of tropospheric NO_2 in Ruoqiang county is soil biogenic NO emission; (2) differences of tropospheric NO_2 VCDs between winter and other seasons represent the source strength of soil NO emission, (3) application of high fertilizer rates (up to 600 kg ha^{-1}) in combination with controlled and effective irrigation in Xinjang's agricultural areas has a strong impact on the high springtime biogenic NO emission from soil by influencing soil nutrient accumulation and soil moisture.

Our more top-down oriented study indicates, that in non-industrial arid areas biogenic NO emissions from soils may play an important role in local/regional atmospheric chemistry. Ongoing research will consider (synoptic) meteorological information (trajectories) and the results of chemical transport models (e.g. MESSy) to assess relationships between observed VCDs and biogenic soil NO sources as well as for up-scaling purposes in the Takalamakan and other arid (cold desert) areas.

P-1-092 Nitrogen dioxide content in the atmospheric boundary layer: measuring technique and results of observations in Moscow Region

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Key words
Nitrogen dioxide atmospheric boundary layer low troposphere

Nitrogen dioxide (NO_2) is one of key components of the chemistry of Earth's atmosphere. Anthropogenic sources of NO_2 produce about 2/3 of the total emissions of this impurity. NO_2 content in the atmospheric boundary layer (ABL) over major cities may exceed the natural background by orders of magnitude. To organize monitoring of the impurity in urban environment, a method of estimating NO_2 integral content in ABL is proposed.

A method is based on measurements of the spectrum of the scattered solar radiation coming from the zenith during daytime and twilight period near 450 nm. The method may be applied for clear sky and overcast. Developed method uses DOAS technique to determine NO_2 slant columns. Usage of twilight measurement allows retrieving accurately stratospheric NO_2 content. Calculation of air mass factors is performed by radiative transfer model MCC++ for interpretation of ABL NO_2 column. Effects of the major factors, affecting the accuracy of the estimation, such as aerosol, clouds and albedo of the underlying surface, are analyzed. It is shown that in cloudless condition and with the cloud lower boundary located above the near-surface layer of NO_2, the determination of the integral ABL content is possible with random errors of 10–35%.

Developed method is used at two observational points located in Moscow as well as at Zvenigorod Scientific Station located at 60 km to the west from Moscow beginning from 2008. Performed observations allow investigate NO_2 emissions in Moscow in 2008-2011.

Integral NO_2 contents in ABL have significant time variation from background value ($5*10^{14}$ mol/cm^2) to high values of $4*10^{17}$ mol/cm^2. We observed significant daily and weekly variations correlated with traffic jam.

Combining the measurements of integral NO_2 in ABL with the EPA ISCST3 air quality model we developed a method for estimation of NO_2 emissions. We estimate that automobiles gave about 90 Mg/year or 65% of total emission.

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P-1-093 Global Daily Ammonia Measurements from AIRS

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Key words
AIRS Ammonia

Global ammonia (NH_3) emissions have been increasing due to the dramatically increased agricultural livestock numbers together with the increasing use in nitrogen fertilization (Sutton et al., 1993). Atmospheric ammonia has impacts on both local scales, acidification and eutrophication of the ecosystems, and international (transboundary) scales through formation of fine ammonium containing aerosols. Measurements with daily and large global coverage are challenging and have been lacking partly because the lifetime of NH_3 is relatively short and partly because it requires high sensitivity for the retrievals that can be only obtained from areas with high thermal contrasts near the surface (Clarisse et al., 2010). We have developed new daily and global tropospheric products of NH3 from AIRS hyperspectral measurements. These products add value to AIRS's existing products that have made significant contributions to weather forecasts, climate studies, and air quality monitoring. This line of products should cover a period from 2002 through the lifetime of AIRS and could continue with future instruments such as CRIS on NPP and NPOESS to obtain consistent climate records on this minor gas. The algorithm development is based on our experience and existing utilities developing AIRS's alternative algorithm using optimal estimation for tropospheric carbon monoxide profiles (Warner et al., 2010). The new products from this study are being evaluated through comparisons with in situ and other remotely sensed measurements, model simulations, and science analyses.

P-1-094 Elemental profiles and fingerprints for fugitive dust in Chinese deserts

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Key words
Elemental profiles Chinese deserts Asian dust Fingerprints Anthropogenic pollution

Source profiles for fugitive dust from source regions in northern China are quite necessary for apportioning ambient concentrations to sources in downwind regions of Asia dust (e.g., Beijing, Shanghai, Taiwan, Korea, Japan, and North America) using the chemical mass balance (CMB) receptor model. Seventeen surface soil samples from six sites of Chinese deserts (Taklimakan desert, Gobi in Xinjiang, Anxinan, Ulan Buh, Central Inner Mongolia and Erenhot), the most important source of Asian dust, were collected, resuspended, and sampled through TSP, PM_{10}, and $PM_{2.5}$ inlets onto Teflon filters. 26 major and trace elements were analyzed by EDXRF (Epsilon 5, PANalytical B. V., the Netherlands) and 18 elemental profiles were calculated.

Based on the signatures of crustal and contaminative elements, Chinese deserts are distinguished into two major sources of Asia dust: northwestern deserts (Taklimakan, Gobi in Xinjiang, and Anxinan) and northern deserts (Ulan Buh deserts, Central Inner Mongolia, and Erenhot). Northern deserts comprise relatively lower mineral elements (Si, Al, Fe, Ca, K, Mg, Na, and Ti) and higher pollutant elements (S, Zn, Mo, Cu, Cr, Pb, Cd, and As) than northwestern deserts, especially in $PM_{2.5}$. Comparing element concentration of dust episodes in three cities from different region of China (Horqin, Beijing, and Xi'an) with the corresponding source, mass percentage of mineral elements (Al, Ca, Fe, Mn, Si,

and Ti etc.) matched very well between ambient aerosol and source, validating the difference between northern and northwestern source. Additionally, Mg/Al, Na/Al, and S/Fe are determined as fingerprints to identify two desert sources. The elemental ratios of Ca/Al, K/Al, Fe/Al, and Ti/Fe in Chinese desert sources of Asian dust compared well with those found at downwind regions like Korea, Japan, and the North Pacific, thus could be considered as source signatures to distinguish Asian dust from other regions like African dust. Enrichment factors (EFs) of Cu, Cr, Zn, Pb, As, Mo, and Cd are one to two orders of magnitude higher than UCC (upper continental crust) average level, and are much higher for northern deserts than northwestern deserts, implying that northern deserts are significantly influenced by anthropogenic pollution sources.

P-1-095 Wet and dry deposition amounts of carbonaceous components at Japanese monitoring sites

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Key words

Carbonaceous components Black carbon Atmospheric deposition Atmospheric budget Short-Lived Climate Forcer

Black carbon has been paid attention to a potent source of human-induced climate change in the past few years. It has been pointed out the globally averaged radiative forcing of black carbon is comparable to that of CO_2. Compared to CO_2, black carbon has a much shorter lifetime, and thus it is known as a “Short-Lived Climate Forcer” (SLCF). Characterization of atmospheric carbonaceous aerosol including black carbon has been investigated worldwide. On the other hand, atmospheric deposition of carbonaceous components were monitored at a few sites in Europe, North America and Africa, which will obscure removal process and atmospheric concentration distribution of black carbon. In this study, carbonaceous components in precipitation and suspended particulate matters were monitored at remote, rural, urban sites in Japan, and wet and dry deposition amounts of carbonaceous components were evaluated.

Field observations have been implemented at Niigata (rural), Sado (remote), and Tokyo (urban) sites in Japan since April 2011 to June 2012. The amounts of elemental carbon (EC) and organic carbon (OC) in precipitation and aerosol were measured by the IMPROVE protocol using a DRI model 2001 carbon analyzer. Wet deposition amounts were calculated as the products of aqueous concentration and precipitation amounts, and dry deposition amounts were as the products of aerosol concentrations and deposition velocity estimated by the inferential method. Seasonal variations of wet and dry depositions, scavenging efficiencies, and

transportation processes were discussed.

Average concentrations of EC in precipitation were 75.7 μg/l, 27.9 μg/l and 23.7 μg/l at Tokyo, Niigata and Sado, respectively. Concentrations of OC and EC in precipitation and aerosol at Tokyo were higher than those at Niigata and Sado. Both of the OC/EC ratios for precipitation and aerosol samples were highest at Sado and lowest at Tokyo. Wet deposition amounts of carbonaceous components of OC and EC at Sado (May 2011 to Feb. 2012) and Tokyo (May 2011 to Feb. 2012) sites were 1355.93 mg/m^2, 18.30 mg/m^2 and 1668.08 mg/m^2, 51.09 mg/m^2, respectively. The importance of wet deposition of carbonaceous component was comparable to dry deposition. At all sites, the scavenging ratio of OC was higher than that of EC. The wash-out processes of OC and EC were more significant at an urban site because of high aerosol concentrations. The scavenging ratio of OC (0.23) at Tokyo was two times higher than that at Sado (0.10). On the other hand, the scavenging ratio of EC (0.10) at Tokyo was slightly higher than that at Sado (0.07). These observations infer that wash-out process of OC at an urban site is more important than that of EC. By back trajectories analysis, the events of high EC concentrations in precipitation during winter at Sado were significantly affected by transportation from the Asian continent, whereas at Tokyo, it would be mainly derived from local emissions.

P-1-096 Characteristics of regional particle pollution by chemical components of PM2.5 on urban and regional sites in Zhejiang Province

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Key words

particles $PM_{2.5}$ PM_{10} chemical composition transport

To characterize regional particle pollution in the Yangtze River Delta areas, as one of three economic development zones in China, the comprehensive field campaign was conducted from 29th October to 28th November 2011 at two super sites, an urban site at Jinhua city, and a regional site at coast rural area of Wenling, in Zhejiang province. Both 24-hour $PM_{2.5}$ and PM_{10} samples were collected at these two sites. The mass concentrations and major chemical compositions were analyzed. The results showed that the average mass concentrations of $PM_{2.5}$ and PM_{10} were 81.93 $\mu g \cdot m^{-3}$, 98.30 $\mu g \cdot m^{-3}$ in Jinhua, and 52.24$\mu g \cdot m^{-3}$, 60.85$\mu g \cdot m^{-3}$ in Wenling, respectively. The ratios of $PM_{2.5}/PM_{10}$ of the two sites were 0.84±0.07 and 0.85±0.11, respectively, suggesting that fine particles were dominant in PM_{10}, and determined the fluctuation of PM_{10}. Secondary inorganic compounds (sulfate, nitrite and ammonium) accounted for 46.5% in Jinhua and 53.2% in Wenling in fine particles, indicating the secondary pollution was severe in Zhejiang. Moreover, organic matter (OM) concentrations were also high, accounting for 23% and 25% in $PM_{2.5}$ at Jinhua and Wenling, respectively. Elemental carbon (EC), as an indicator of primary emission, accounted for 7% and 4% in $PM_{2.5}$, indicating the primary emissions were also important in Zhejiang, especially in urban area. In addition, back trajectory analysis showed particle chemical compositions and sources were quite different when the air mass was from different origins. The particle compositions and sources from different origins will be discussed in detail to characterize particle regional transport.

P-1-097 Reconstructed histories of ozone depleting substances and related trace gases in the northern hemispheric atmosphere from Greenland firn air

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Key words

Trace gases Halocarbons Firn air Ozone depleting substances

Firn air, collected in Greenland as part of the North Eemian (NEEM) ice drilling campaigns, provides a record of changes in northern hemispheric atmospheric composition since the early 20th century, and captures the start of industrial emissions of the majority of manmade trace gases in the atmosphere today. We present here an overview of the atmospheric histories of a number of trace gases (mostly halogenated species) measured in the NEEM firn profiles. These gases fall in to a number of families as follows. (1) Ozone depleting chlorofluorocarbons (CFCs), halons and hydrochlorofluorocarbons (HCFCs); including some recently detected minor species. The majority of CFCs have been declining in abundance as a result of the cessation of production mandated by the Montreal Protocol, although some minor species have continued to increase, as have the HCFCs. (2) Hydrofluorocarbon (HFC) "replacements" for ozone depleting substances (ODS) have grown, in many cases very rapidly. Some, such as fluoroform (HFC-23) have relatively long histories of emission (1950s), while others (e.g. HFC-227ea) have only become detectable in the last one or two decades. (3) A number of short-lived ODSs and "Very Short-Lived Substances" (VSLS; having atmospheric lifetimes of less than half a year) have also been measured. Many of these appear to have increased from low or zero abundance in the early 20th century, reached peak concentrations in the 1970s to 1980s, and then subsequently declined. These include chlorocarbons such as dichloromethane (CH_2Cl_2), dichloroethane (CH_2ClCH_2Cl), tetrachloroethene (C_2Cl_4), and chloroform ($CHCl_3$). The first three, however, appear to have begun to increase again, while $CHCl_3$ is no longer decreasing. Methyl chloride (CH_3Cl), while exhibiting an apparent largely 'natural' background concentration, also appears to have increased and then decreased over the same period. Some brominated VSLS also exhibit somewhat similar behaviour to chlorinated VSLS, including 1,2-dibromoethane (CH_2BrCH_2Br), bromoethane (C_2H_5Br), and 'halothane' ($CF_3CHBrCl$), although all of these continue to decrease in the atmosphere. Dibromomethane (CH_2Br_2) and dichlorobromomethane ($CHCl_2Br$), while evidently of predominantly natural origin, also show distinct evidence of an anthropogenic contribution. The possible implications for changes in abundance of some of the aforementioned ODSs on halogen loading in the stratosphere are considered.

P-1-098 A multi-model evaluation of processes controlling spring and summer tropospheric ozone in the Arctic: The POLARCAT Model Intercomparison Project (POLMIP)

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Key words
ozone Arctic troposphere model intercomparison POLARCAT

The Arctic has warmed rapidly in the past few decades compared with observed global-mean temperature increases. Recent calculations suggest that changes in short-lived pollutants such as ozone and aerosol may have contributed significantly to this warming. Arctic tropospheric budgets of short-lived pollutants are impacted significantly by long-range transport of gases and aerosols from Europe, Asia and N. America, and in summer by boreal wildfires. Previous assessments based on limited observations at the surface, have demonstrated that chemistry-transport models have highly variable and generally poor skill in reproducing short-lived pollutant concentrations in the Arctic. This suggests a significant limitation in the ability of models to predict Arctic and hemispheric climate response to changes in mid-latitude emissions.

The POLARCAT Model Intercomparison Project (POLMIP) aims to exploit the large number of observations collected in the Arctic troposphere as part of International Polar Year in 2008, to evaluate a series of 10 state-of-the-art global and regional atmospheric chemical transport models. Here, we present an evaluation of model processes controlling the Arctic tropospheric ozone budget across these models. The extensive aircraft, surface and satellite observations allow unprecedented evaluation of controls on spring and summer tropospheric ozone. A large variability between models in Arctic burdens of reactive organic species (particularly oxygenates) and partitioning of reactive nitrogen (NOy) is demonstrated, with potential consequences for the Arctic ozone budget. We identify model chemical and physical processes contributing to these inter-model differences. In addition, we investigate the importance of model stratosphere treatment in controlling modelled Arctic ozone and HOx budgets, both through direct ozone input to the troposphere and indirectly through the effects of stratospheric ozone on photolysis rates. Idealised tracers are used to quantify relative pollutant contributions from mid-latitude source regions, boreal fires and stratospheric air mass input, and their variability between models. We present a novel comparison of modelled and observed Arctic CO, ozone and NOy in coordinates defined by air mass origins, revealing enhancements associated with

specific transport regimes that are consistently poorly represented in models. Key processes are identified that require optimal representation in models to ensure realistic response of Arctic tropospheric ozone to mid-latitude emission changes.

P-1-099 A record of atmospheric carbon monoxide back to 1950 AD reconstructed from Greenland firn air

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Key words

carbon monoxide emissions historical

We present a reconstruction of Northern Hemisphere (NH) high latitude carbon monoxide (CO) concentration from Greenland firn air. Firn air samples were collected at three deep ice core sites in Greenland (NGRIP, Summit and NEEM). CO records from the three sites agree well with each other as well as with recent atmospheric measurements, indicating that CO is well preserved at these firn air sites. CO atmospheric history was reconstructed back to 1950 AD from the firn measurements using a combination of two forward models of gas transport in firn and an inverse model. The reconstructed history suggests that Arctic CO was already higher in 1950 than it is today. CO concentration rose gradually until the 1970s, and peaked in the 1970s or early 1980s, followed by a decline to today's levels. We compare the CO history with the atmospheric histories of methane, light hydrocarbons, molecular hydrogen, CO stable isotopes and hydroxyl radical (OH), as well as with published CO emission histories and results of a long historical run from a Chemistry-Transport model. We find that the reconstructed CO history is impossible to reconcile with available emission histories unless large changes in OH are assumed. We argue that the available CO emission histories strongly underestimate NH emissions, and fail to capture the emission decline starting in the late 1970s, which was most likely due to reduced emissions from road transportation.

P-1-100 Modeling changes in Arctic air pollution due to increased industrial activities

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Key words

Arctic air quality Regional modeling

The Arctic is undergoing very rapid changes, such as decreasing sea-ice extent during summer. As a result, transit shipping via the Northern Sea Route, along the northern coast of Scandinavia and Russia, is already occurring. As shipping through the Arctic increases, emissions of air pollutants (aerosols, ozone, and their precursors) into the lower troposphere are likely to become more significant. In addition to shipping, emissions linked to extraction of Arctic oil/gas deposits and associated infrastructure will also increase. As part of the EU ACCESS project, we investigate the role of current and future anthropogenic activities in the Arctic on regional air pollution and the concentrations of short-lived climate forcing

agents in the Arctic troposphere using a regional chemical transport model. The model simulates emissions, transport, mixing, and chemical transformation of trace gases and aerosols simultaneously with the meteorology. The model is used to examine ozone and aerosol formation in fresh and aged plumes. The sensitivity of results to model resolution is also examined. Results are also used to quantify the impacts of ship and oil/gas emissions on regional scales in the Arctic. Model results will be discussed in the context of a DLR aircraft campaign in July 2012, based in northern Norway, to investigate emissions from Arctic shipping and oil/gas extraction (including measurements of trace gases and aerosols).

P-1-101 TransCom model simulations of CH4 and related species: linking transport, surface flux and chemical loss with CH4 variability in the troposphere and lower stratosphere

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A chemistry-transport model (CTM) intercomparison experiment (TransCom-CH_4) has been designed to investigate the roles of surface emissions, transport and chemical loss in simulating the global methane distribution. Model simulations were conducted using twelve models and four model variants and results were archived for the period of 1990–2007. All but one model transports were driven by reanalysis products from 3 different meteorological agencies. The transport and removal of CH_4 in six different emission scenarios were simulated, with net global emissions of 513±9 and 514±14 $TgCH_4\ yr^{-1}$ for the 1990s and 2000s, respectively. Additionally, sulfur hexafluoride (SF_6) was simulated to check the interhemispheric transport, radon (222Rn) to check the subgrid scale transport, and methyl chloroform (CH_3CCl_3) to check the chemical removal by the tropospheric hydroxyl radical (OH). The results are compared to monthly or annual mean time series of CH_4, SF_6 and CH_3CCl_3 measurements from 8 selected background sites, and to satellite observations of CH_4 in the upper troposphere and stratosphere. An average IH exchange time of 1.39±0.18 yr is derived from SF_6 time series. Using six sets of emission scenarios, we show that the decadal average CH_4 growth rate likely reached equilibrium in the early 2000s due to the flattening of anthropogenic emission growth since the late 1990s. Up to 60% of the IAVs in the observed CH_4 concentrations can be explained by accounting for the IAVs in emissions, from biomass burning and wetlands, as well as meteorology in the forward models. The modeled CH_4 budget is shown to depend strongly on the troposphere-stratosphere exchange rate and thus on the model's vertical grid structure and circulation in the lower stratosphere. The 15-model median CH_4 and CH_3CCl_3 atmospheric lifetimes are estimated to be 9.99±0.08 and 4.61±0.13 yr, respectively, with little IAV due to transport and temperature. First results are published in Patra et al. (2011; www.atmos-chem-phys.net/11/12813/2011/)

P-1-102 Simulated Atmospheric Composition Retrievals for the Panchromatic Fourier Transform Spectrometer (PanFTS)

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Key word:
PanFTS Atmospheric composition multispectral retrieval sensitivity precision and systematic errors

The US National Research Council Earth Science Decadal Survey recommended the Geostationary Coastal and Air Pollution Events (GEO-CAPE) mission to make unprecedented measurements of the spatial and temporal variability of trace gases and aerosols that influence air quality. The current baseline plan for the atmospheric science component of GEOCAPE calls for hourly measurements over North America from 10° N to 60° N latitude. The Panchromatic Fourier Transform Spectrometer (PanFTS) is a candidate instrument for the GEO-CAPE mission. With sensitivity from 0.28 to 11 micron and high spectral resolution, PanFTS combines the functionality of separate UV, visible and IR instruments in a single package designed to measure all of the trace species required to accomplish the GEOCAPE atmospheric science objectives. This presentation summarizes study results that demonstrate the capability of PanFTS to meet or exceed GEO-CAPE atmospheric science requirements. Simulations were performed for a wide variety of locations, including core urban areas such as Los Angeles, downwind urban areas such as Essex (downwind of Baltimore) and remote rural locations such as southeast Oregon. Day- and night-time, as well as weekday and weekend, scenarios are considered. A multispectral retrieval approach was used to obtain estimates of the sensitivity, precision and systematic errors for species of relevance to GEO-CAPE, including O_3, CO, CH_4, NO_2, SO_2 and NH_3, using simulated PanFTS measurements over the continental
US.

P-1-103 Thermal characteristics of carbonaceous aerosol observed at anurban site of Ulaanbaatar, Mongolia in winter and spring of 2008

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Key word:
carbonaceous aerosol thermal evolution patterns

In order to understand the characteristics of atmospheric carbonaceous aerosol at an urban site in Ulaanbaatar, Mongolia, 24-h PM2.5 fine particulate matter samples

were collected in winter (January 9~February 17) and spring (March 11~May 2) of 2008. Filter based organic carbon (OC) and elemental carbon (EC) were analyzed by the thermal–optical transmittance (TOT) method with NIOSH 5040 temperature protocol. Very high OC was measured in winter which was much higher than those measured at other urban sites in Asia. Daily average OC concentration varied in the range of 3.5~51.3 microgram carbon per cubic meter with a mean of 26.2±12.3 microgram carbon per cubic meter while daily average EC concentration ranged from 0.3~3.2 microgram carbon per cubic meter with a mean of 1.5±0.7 microgram carbon per cubic meter from January 9 to February 17, 2008.

During the spring season OC varied in the range of 2.2~23.5 microgram carbon per cubic meter with a mean of 8.6±4.4 microgram carbon per cubic meter while EC ranged from 0.7~3.9 microgram carbon per cubic meter with a mean of 1.6±0.6 microgram carbon per cubic meter. Average contributions of EC to total carbon were 5.6±2.5% and 18.9±7.3% during winter and spring season, respectively, showing that emission of EC increased in spring. It can be explained that wood burning at home stoves is increased significantly in spring season than winter. The OC and EC were moderately correlated during the winter (0.64) and spring (0.54), which suggested that they were emitted from different sources. In order to better understand the characteristics of atmospheric carbonaceous aerosol at an urban atmosphere, thermal evolution patterns of carbonaceous aerosol were characterized based on the multiple temperature steps; OC1, OC2, OC3, OC4, and EC. On average, contributions of OC1, OC2, OC3, OC4, and pyrolyzed carbon (PC) were measured to be 44.5%, 11.3%, 9.1%, 3.3%, and 32.4% for the winter period while they were 42.3%, 10.4%, 13.4%, 2.1%, and 31.8% during the spring period, respectively. The haze level was classified into three categories based on the 5- day air mass back trajectories and 24-h atmospheric pressure changes. Stagnant (ST) condition showed that air mass was originated within 1000 km from the sampling site while Continental (CT) cases showed that it was originated from Russian and Kazakhstan continent. Low pressure (LP) cases indicated that atmospheric pressure was lower than 1040 hPa with >8 hPa variations within 24 hours. EC concentration was found to be 1.6±0.7 microgram carbon per cubic meter under ST conditions, which was about 2.5 times higher than that measured under LP conditions in winter time. Total OC concentration during the ST condition was approximately 5 times higher than that observed during the LP condition, which is mainly due to increase of OC1 and PC in winter period. Higher OC1 indicated that lower molecular weight water-soluble organic compounds were dominant under the ST condition.

P-1-104 Seasonal Characteristics of Chinese Outflows Determined from the Water-soluble Compositions of PM1.0 Using PILS at Gosan ABC S uperstation

Xiaona Shang, Jihyun Han, Sungho Woo, Muhyun Jung, Meehye Lee (Earth & Environmental Science) uperstation

Keyword:

PILS Chinese outflow $PM_{1.0}$

Water-soluble ions, TOC, and WSOC in PM1.0 were quantified at 1-hour intervals utilizing Particle-Into-Liquid-Sampler (PILS) and off-line IC and TOC Analyzer at Gosan ABC superstation during the late fall in 2010 and March in 2011. The major water-soluble ions and carbonaceous compounds contributed about 80% to the total mass. In the spring, the continental outflows were transported fast mainly from the Beijing region, in which NO_3^- was the most dominant aerosol species with the high SO_2 concentration. In contrast, the fall event was characterized by enhance SO_4^{2-}, K^+, and organic carbon with the high NO_2 concentration. This distinct seasonal variation was controlled by the degree of atmospheric processing and the type of emission sources that were greatly influenced by meteorological settings in the study region. The major sources of $PM_{1.0}$ include coal combustion, vehicle emission, and biomass burning. Additionally, sea salts were sporadically enhanced, which were likely to be influenced by salts deposit in the northeastern China as well as the Yellow Sea. In particular, biomass burning was found to contribute the noticeable haze event observed in November 2010. In the haze event, the concentrations of SO_4^{2-}, NO_3^-, NH_4^+, K^+, and TC were highly elevated and there were excellent correlations between these species and PM_{10} mass, PAN (peroxyacetyl nitrate), and visibility. These results demonstrate that the haze particles were formed through photochemical activities and was likely to be transported a long distance.

P-1-105 Satellite observations of atmospheric composition for air quality and climate science: Moving to geostationary Earth orbit

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Advances in tropospheric remote sensing over the past decade have shown the potential of satellite measurements to quantify the sources and distributions of gases important for air quality and climate. However, current satellite observations of tropospheric composition are made from low Earth orbit and provide at best one or two measurements each day at any given location. Coverage is global but sparse, often with large uncertainties in individual measurements that limit examination of local and regional atmospheric composition over short time periods. This has hindered the operational uptake of these data for monitoring air quality and population exposure, and for initializing and evaluating chemical weather forecasts. This paper discusses how the air quality and chemistry-climate science communities would benefit from an observing framework for atmospheric composition that includes observations from geostationary Earth orbit in addition to the current low Earth orbit, suborbital and surface measurements. These would provide time-resolved high-density observations of continental domains for mapping pollutant sources and variability on diurnal and local scales. We describe progress towards transitioning measurement capability already demonstrated in low Earth orbit to geostationary orbit in the context of the NASA GEOCAPE mission over North America and outline how it complements plans for other geostationary missions from international partners. Together, these present the possibility of achieving a constellation of geostationary platforms to achieve continuous coverage at northern mid-latitudes by the end of this decade.

P-1-106 Observational levels and trends of gaseous SO2 and HNO3 from 1997 to 2009 at Waliguan Station, China

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For the first time, the long-term measurement results of SO_2 and HNO_3 at Waliguan Global Atmosphere Background station are presented and their trends are analyzed. From 1997 to 2009, the annual mean concentrations of SO2 and HNO_3 at Waliguan are 1.28 ± 0.41 $\mu g/m^3$ and 0.22 ± 0.19 $\mu g/m^3$, respectively. The HNO_3 concentrations are much high in warm season than in cold season, but the SO_2 concentrations are lower in warm season than in cold season. The SO_2 concentrations have a very significant decreasing trend ($P<0.0001$) of -0.2 $\mu g/m^3/a$ in 1997-2002, but in 2003-2009, it showed a significant increasing trend ($P<0.05$) of +0.6 $\mu g/m^3/10a$, which is not consistent with the decreasing trends in many other regions over the world. The trends of HNO_3 are not significant in statistics. The trend of HNO_3/SO_2 ratio is not significant in 1997-2002 but it has a significant downward trend ($P<0.05$) in 2003-2009. Weak increasing trends at significant level ($P<0.05$) of SO_2 with +1.3 μg/m3/10a and HNO_3 with +0.07$\mu g/m^3/10a$ during year 2003-2009 at Waliguan are found in the originations of airmass from Tibetan Plateau based on the result of backward trajectories clustering analysis.

Poster Presentations, Session 2

P-2-001 Mixing of Asian mineral dust with anthropogenic pollutants and its impact on regional atmospheric environmental and oceanic biogeochemical cycles over East Asia: a model case study of a super-duststorm in March 2010

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Key words
Mixing processes Dust particles Anthropogenic pollutants Heterogeneous chemistry Dissolved Fe depositions

Mixing of Asian mineral dust with anthropogenic pollutants allows pollutants (e.g. sulfate and nitrate) to be transported over longer distances (e.g. to the northern Pacific, even to North America) along with dust particles. This mixing therefore affects the atmospheric and oceanic environment at local, regional and even continental scales. In this study, we used a three-dimensional regional chemical transport model (NAQPMS) to examine the degree of mixing between Asian mineral dust and anthropogenic pollutants in a super-duststorm event during 19-22 March 2010. Influences of the mixing processes on regional atmospheric environmental and oceanic biogeochemical cycles were also investigated. A comparison with measurements showed that the model reproduced well the trajectory of long-range dust transport, the vertical dust profile, and the chemical evolution of dust particles. We found that along-path mixing processes during the long-range transport of Asian dust led to increasingly polluted particles. As a result, ~60% of the sulfate and 70-95% of the nitrate in the downwind regions was derived from active mixing processes of minerals with pollutants sourced from the North China Plain and enhanced by transport over South China. This mixing had a significant impact on the regional-scale atmospheric composition and oceanic biogeochemical cycle. Surface HNO_3, SO_2 and O_3 were decreased by up to 90%, 40% and 30%, respectively, due to the heterogeneous reactions on dust particles. Fe solubility rose from ~0.5% in the Gobi region to ~3-5% in the northwestern Pacific, resulting from oxidization of SO_2 on dust particles. Total Fe (II) deposition in the ocean region of East Asia reached 327 tons during the 4-day dust event, and created a calculated primary productivity of ~520 $mgC \cdot m^{-2} \cdot d^{-1}$ in the Kuril Islands, which can support almost 100% of the observed mean marine primary productivity

in spring in this region (526 mgC·m^{-2}·d^{-1}).

P-2-002 The seasonal characterization of components and size distributions for submicron aerosols in Beijing

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Key words
chemical components size distribution AMS seasonal variation

From January to October in 2008, the characterization of submicron aerosol (PM1) in Beijing was studied using an aerodyne aerosol mass spectrometer (AMS). This paper presents the seasonal features of different chemical components (sulfate, nitrate, ammonium, and organics) and size distribution in PM1.
The measurements show that the bulk mass concentration of PM1 was the highest in summer, and the lowest in fall. Organics was the largest fraction among the five components of aerosols in all seasons, accounting for 36%-58% of PM1, the concentration of organics was highest in winter. While those of inorganic components, sulfate, nitrate, ammonium were the highest in summer. Principal component analysis technique deconvolved and quantified two types of organic aerosols, hydrocarbon-like and oxygenated (HOA and OOA, respectively). The HOA showed the highest level in winter, and occupying 70% of organics. However, OOA displayed the highest level in summer, and lower in fall and winter.
The size distribution of all the species showed apparent peaks in the size of 500-600 nm. The size distributions of organics were much broader than other species, particularly in fall and winter. The size distribution of sulfate, nitrate and ammonium demonstrated similar patterns, and extending broader in winter. The contributions of different species were size-dependent, the finer the particle the more contribution of organics. The organics occupied more than 60% in particles smaller than 200 nm, and in winter, the organics contributed 50% to PM1. In spring and summer, the HOA was the dominating organic fraction in particles smaller than 200 nm. While the OOA contributed more to particles larger than 300nm. In winter, the HOA contributed more than OOA to all particles of PM$_1$.

P-2-003 Satellite remote sensing of particulate matter in the atmosphere of Dakar, Senegal

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Key words
aerosol remote sensing

Air pollution is a major issue for global environment as well as human health and well-being. Atmospheric particulate matter (aerosols) has an impact on human health as it can easily penetrate into the respiratory system. It has also an impact on global climate because it interacts with the Earth radiative budget and the cloud system. Western Africa is characterized by a large amount of both natural and anthropogenic aerosols. The question of emissions by African mega-cities (such as Dakar, Bamako or Lagos) is particularly crucial because the west African urban population has the world largest growth rate. Our study aims at using high resolution satellite imagery to detect aerosols at the scale of the city of Dakar, Senegal. In Dakar, the “Centre de Gestion de la Qualité de l’Air” (CGQA) is managing a network of 5 air quality monitoring stations providing PM10 and PM2.5 measurements. We have benefited from the Planet Action program for 7 FORMOSAT-2 images over the area of Dakar at a spatial resolution of 8 min April and May 2011. The aerosol optical depth (AOD) has been evaluated from calibrated radiances and a specific algorithm developed by (Hagolle et al., 2008). Mineral dust is shown to be a major component of the atmospheric load. FORMOSAT-2 derived AOD shows are correlated with ground-level measurements acquired at the same time in the city of Dakar (university Cheik Anta Diop) but show a significant underestimation. The vertical extent of the aerosol layers is estimated by using LIDAR observation in M’Bour (Léon et al., 2009). Our estimation of the ground-level PM10 from the AOD by using the LIDAR derived AOD scale height correlates well with the observations of the air quality stations. Current developments focus on the use of an aerosol model accounting for non-sphericity of dust particles. Further applications of this study are expected in the frame of forthcoming satellite missions such as Venµs, LDCM or Sentinelle-2.

P-2-004 NO$_x$ in Chinese Megacities

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Key words
Nitrogen oxides (NO$_x$) Megacity NO$_x$ concentrations NO$_x$ sources

In the past 30 years, China has undergone great economic improvements, the urbanization rate is increasing, megacities and city clusters are expanding, leading to the development of China’s economy and society. However, the city expansion has resulted in a series of environmental problems, such as increasing energy consumption and consequently emission of pollutants, as well as deterioration in

urban air quality. In spite of many measures taken by the Chinese government, air pollution from nitrogen oxides (NO_x=NO+NO_2) is still serious and the NO_x concentration is increasing. This article analyses NO_x pollution and its trends in Chinese megacities from three aspects, authoritative routine reports, campaign based research observations, and satellite observations. The article also discusses the main sources of NO_x in Chinese Megacities. The discussion is based on a review of research papers on the China NO_x emission inventory.

P-2-005 Molecular Distribution and Stable Carbon Isotopic Composition of Dicarboxylic Acids, Ketocarboxylic Acids and a-Dicarbonyls in Size-Resolved Atmospheric Particles: Implications for Secondary Organic Aerosol Formation Mechanisms

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Key words
SOA Isotopic Composition Source Formation Mechanism

Size-resolved airborne particles (9-stages) in urban Xi’an, China during summer and winter were measured for molecular distributions and stable carbon isotopic compositions of dicarboxylic acids, ketocarboxylic acids and a-dicarbonyls. To our best knowledge, we report for the first time the size-resolved differences in stable carbon isotopic compositions of diacids and related compounds in continental organic aerosols. High ambient concentrations of terephthalic (tPh, 379 ± 200 ng m^{-3}) and glyoxylic acids (wC2, 235 ± 134 ng m^{-3}) in Xi'an aerosols during winter compared to those in other Chinese cities suggest significant emissions from plastic waste burning and coal combustions. Most of the target compounds are enriched in the fine mode (<2.1 mm) in both seasons, peaking at 0.7-2.1 mm. However, summertime concentrations of malonic (C3), succinic (C4), azelaic (C9), phthalic (Ph), pyruvic (Pyr), 4-oxobutanoic (wC4) and 9-oxononanoic (wC9) acids, and glyoxal (Gly) in the coarse mode (> 2.1 mm) are comparable to and even higher than those in the fine mode (< 2.1 mm). Stable carbon isotopic compositions of the major organics are higher in winter than in summer, except oxalic acid (C2), wC4, and Ph. d13C of C2 showed a clear difference in sizes during summer, with higher values in fine mode (ranging from -22.8‰ to -21.9‰) and lower values in coarse mode (-27.1‰ to -23.6‰). The lower d13C of C2 in

coarse particles indicate that coarse mode of the compound originates from evaporation from fine mode and subsequent condensation/adsorption onto pre-existing coarse particles. Positive linear correlations of C2, sulfate and wC2 and their d13C values suggest that wC2 is a key intermediate, which is formed in aqueous-phase via photooxidation of precursors (e.g., Gly and Pyr), followed by a further oxidation to produce C2.

P-2-006 Cloud condensation nuclei from fresh and aged air pollution in the megacity region of Beijing: size-resolved measurements and parameterization of aerosol chemical composition, hygroscopicity and CCN activity

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Key words
Aerosols CCN in Beijing

Atmospheric aerosol particles serving as cloud condensation nuclei (CCN) are key elements of the hydrological cycle and climate. CCN properties were measured and characterized during the CAREBeijing-2006 campaign at a regional site south of the megacity of Beijing, China. Size-resolved CCN efficiency spectra recorded for a supersaturation range of S = 0.07 % to 0.86 % yielded average activation diameters in the range of 190 nm to 45 nm. The corresponding effective hygroscopicity parameters (κ) exhibited a strong size dependence ranging from

~0.25 in the Aitken size range to ~0.45 in the accumulation size range. The campaign average value ($\kappa = 0.3 \pm 0.1$) was similar to the values observed and modeled for other populated continental regions.

The hygroscopicity parameters derived from the CCN measurements were consistent with chemical composition data recorded by an aerosol mass spectrometer (AMS) and thermo-optical measurements of apparent elemental and organic carbon (ECa and OC). The CCN hygroscopicity and its size dependence could be parameterized as a function of only AMS based organic and inorganic mass fractions using the simple mixing rule $\kappa p \approx 0.1 \cdot forg + 0.7 \cdot finorg$

When the measured air masses originated from the north and passed rapidly over the center of Beijing (fresh city pollution), the average particle hygroscopicity was reduced ($\kappa = 0.2 \pm 0.1$), which is consistent with enhanced mass fractions of organic compounds (~50 %) and ECa (~30%) in the fine particulate matter (PM1). Moreover, substantial fractions of externally mixed weakly CCN-active particles were observed at low supersaturation (S = 0.07%), which can be explained by the presence of freshly emitted soot particles with very low hygroscopicity ($\kappa < 0.1$). Particles in stagnant air from the industrialized region south of Beijing (aged regional pollution) were on average larger and more hygroscopic, which is consistent with enhanced mass fractions (~60%) of soluble inorganic ions (mostly sulfate, ammonium, and nitrate). Accordingly, the number concentration of CCN in aged air from the megacity region was higher than in fresh city outflow ((2.5-9.9)×103 cm^{-3} vs. (0.4-8.3)×103 cm^{-3}) although the total aerosol particle number concentration was lower (1.2×104 cm^{-3} vs. 2.3×104 cm^{-3}). A comparison with related studies suggests that the fresh outflow from Chinese urban centers generally may contain more, but smaller and less hygroscopic, aerosol particles and thus fewer CCN than the aged outflow from megacity regions.

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P-2-007 Gravity-current driven transport of Haze from North China Plain to Northeast China in winter 2010-Part1: observations

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Key words
Haze Transport North China Plain Northeast China Plain

Haze exerts a large effect on visibility reduction and has serious impacts on air quality and human health. Understanding the sources and transport of haze is of importance to improve the regional air quality and evaluate its health effects. In this study, we investigated a typical haze episode that occurred in northeast (NE) China during 4-6 November, 2010 by analyzing the ground PM10 measurements from 11 monitoring sites, aerosol Lidar observations, synoptic charts, MODIS satellite imageries, and back trajectories. Our analyses suggest that the regional haze formed in North China Plain (NCP) under stagnant conditions can be transported to NE China in ~1-3 days across Bohai Bay and Liaodong Bay - a typical transport pathway associated with the topography of northern China. The haze episode appeared to evolve progressively from southwest to northeast in the region of NE China, in agreement with the appearance of PM10 peak values, wind patterns, MODIS images and the back trajectories of air masses. Due to the haze impact, the NE China showed significantly elevated particulate matter pollution by a factor of ~4-6 with the peak concentrations reaching ~410 μg m^{-3}. The results together indicate that the regional transport from the NCP has a significant contribution to the PM pollution in NE China, thus efforts to control the source emissions over NCP would be effective to improve the air quality in NE China.

P-2-008 Investigation of atmospheric nitrate and ammonium and their impact on air quality and climate

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Key words
nitrate and ammonium chemistry response

The capability to simulate nitrate and ammonium aerosols has been developed in NASA GMI model by implementing a thermodynamic equilibrium model that

treats gas and aerosol multiphase chemical equilibrium reactions in a SO_4-NO_3-NH_4-H_2O system. We evaluated the simulations of the new aerosol components by comparing the model results with the ground station observations from US CASTNET and Europe EMEP networks. We also compared their mass, AOD at 550 nm, and direct radiative forcing with other models by participating AeroCom II aerosol direct radiative forcing activity. Our major task is to investigate how nitrate and ammonium impact on atmospheric chemistry. Previously, our model treated HNO_3 solely as a gas phase tracer. This semi-volatile species now partitions between gas and aerosol phases, and the tracer in each phase is subject to different chemical and physical processes. Our preliminary analysis indicated that gas phase HNO_3 decreased globally in all seasons due to loss partially into nitrate aerosol. Decreased gas phase HNO_3 reduced global NO_x and OH through reducing HNO_3 photodissociation. Global O_3 response to these changes is more complicated, but the average global O_3 change was less than 2%

P-2-009 Comparisons of NO_x Emissions over South Korea

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Key words
NO_x emission REAS CAPSS

1. Introduction

Northeast Asia including China, Korea and Japan is characterized by high emissions of anthropogenic air pollutants. Among them, China emits an overwhelming fraction of NO_x, and the emission amount of NO_x in Northeast Asia is drastically increasing. NO_x is emitted from anthropogenic sources such as fossil fuel combustion and biomass burning, as well as natural sources such as lightning and microbiological processes in soil and plays important roles in oxidation processes in the atmosphere. In this study, we focused on the NO_x emission inventories of South Korea.

2. Methods

In this study, two bottom-up emission inventories, Regional Emission inventory for ASia (REAS) and Clean Air Policy Support System (CAPSS), which are the latest emission inventories about the air pollutant emissions about South Korea were compared to find out the trend of NO_x emission from the past to the present. Also, these two emission inventories compared to the top down NO_x estimation by inverse modeling to estimate the accurate amount of NO_x emitted from South Korea.

3. Results

The total NO_x emission trends, sectoral and regional comparisons were carried out. In total NO_x emissions, REAS was bigger than CAPSS about 3.0E+05 ton per year. The NO_x emission of REAS was bigger than that of CAPSS in Seoul and Busan. On the other hand, the NO_x emission of REAS was smaller than that of CAPSS in North and South area in Korea.

4. Discussion and conclusion

The difference of absolute value of NO_x emission between REAS and CAPSS was mainly caused by the difference in Transport sector, especially emissions from diesel buses (REAS was bigger than CAPSS about 2.5E+05 ton per year) and diesel cars (REAS was bigger than CAPSS about 5E+05 ton per year).

The regional differences of NO_x emission between REAS and CAPSS could be explained by the difference in Vehicle Kilometers Travelled (VKT) and emission factors (EF) used in REAS and CAPSS. REAS used longer VKT in Busan, Seoul, and Incheon than CAPSS. Also, REAS used bigger EF than CAPSS for diesel buses and diesel cars (5.01 times bigger for Diesel Buses, and 3.85 times bigger for diesel cars).

P-2-010 Improvement of ozone forecast over Beijing based on ensemble Kalman filter with simultaneous adjustment of initial conditions and emissions

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Key words
Ozone data assimilation Ensemble Kalman Filter

In order to improve the surface ozone forecast over Beijing and surrounding regions, data assimilation method integrated into a high-resolution regional air quality model and a regional air quality monitoring network are employed. Several advanced data assimilation strategies based on ensemble Kalman filter are designed to adjust O_3 initial conditions, NO_x initial conditions and emissions, VOCs initial conditions and emissions separately or jointly through assimilating ozone observations, exploring possible solutions to reduce the uncertainty in precursor emission and initial conditions when precursor observations are scarce. As a result, adjusting precursor initial conditions demonstrates potential improvement of the 1-hour ozone forecast almost as great as shown by adjusting precursor emissions. Nevertheless, either adjusting precursor initial conditions or emissions show deficiency in improving the short-term ozone forecast at suburban areas. Optimizing ozone initial values brings significant improvement to the 1-hour ozone forecast, and its limitations lie in the difficulty in improving the 1-hour forecast at some urban site. A simultaneous adjustment of the above five variables is found to be able to reduce these limitations and display an overall better performance in improving both the 1-hour and 24-hour ozone forecast over these areas. The root mean square errors of 1-hour ozone forecast at urban sites and suburban sites decrease by 51% and 58% respectively compared with those in free

run. Through these experiments, we found that assimilating local ozone observations is determinant for ozone forecast over the observational area, while assimilating remote ozone observations could reduce the uncertainty in regional transport ozone.

P-2-011 Comparison of ozone concentration and its trends in the lower troposphere at European ozonosonde stations and at Beijing.

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Key words

ozone concentration trends in the lower troposphere comparison Asian and European stations

This poster deals with the comparison of ozone concentration and trends in the lowermost troposphere at European ozonosonde stations and at Beijing and at Sapporo in Japan up to 4 km, because the greatest differences are expected in the lowermost troposphere due to strong industrial development in Beijing. We use the ozonosonde data from these stations: Payerne, Hoheipeissenberg, Uccle, Lindenberg and Legionowo.in the period 2001-2008. We also use measurements of ozone vertical profile from Japanese station Sapporo which is situated in the rural areas. All these stations use the ECC type of ozonosonde. We grouped months into the seasons and at each season we search for tropospheric ozone concentration and trend. The results shown that there are some obvious different variation characteristics in the ozone vertical profiles at European and Asian stations, and the greatest differences are observed in the lowermost troposphere in Beijing due to strong human activities.

P-2-012 Low cost sensor networks for measuring urban air quality

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Key words

Air Quality Sensor Networks NO_x CO High Density Low Cost

Measurements at the appropriate spatial and temporal scales are vital for monitoring and understanding spatially heterogeneous systems with complex and largely uncertain sources, such as the urban environment. This work shows that miniature, low-cost sensor nodes incorporating electrochemical gas sensors originally developed for industrial applications sensing at parts-per-million (ppm) mixing ratios can, when suitably configured, be used for urban air quality studies at parts-per-billion (ppb) levels. Networks based on these systems can be low cost and highly portable, thus allowing their use in air quality sensor networks with fine spatial and temporal measurement scales, in both static and mobile configurations. Routine air quality monitoring networks currently rely on either relatively sparse networks of expensive instrumentation (e.g. chemiluminescence) or higher density networks with a lower time resolution (e.g. NO_2 diffusion tubes). As a result, in many cases important aspects of urban air quality, e.g. spatial and temporal granularity and 'hot spots' cannot be reliably captured. Urban air quality may therefore not be adequately constrained and any derived measurement metrics e.g. personal exposure will be unreliable.

In this paper we demonstrate that electrochemical gas sensors can be used to measure key air quality gases such as CO, NO and NO_2 at the low ppb mixing ratios present in the urban environment, with laboratory instrumental detection limits estimated to be < 4 ppb for CO and NO and < 1 ppb for NO_2.

We then demonstrate the fundamental capabilities of these sensors in the field, including comparisons with reference instruments, and illustrative measurements from a number of mobile and static multi-node deployments involving high-density networks of in some cases in excess of 40 nodes and over periods of hours to months. The overall structure of time series collected using these sensors is clearly detailed, with both long term statistical information and individual pollution events being evident in the data, illustrating the variability over a wide range of time scales that can be detected using these sensors.

The widely varying mixing ratios (temporally and spatially) reported by these studies demonstrate that in many cases the urban environment cannot be characterized using sparse (static) networks, and that observations from networks with higher resolution add substantially to those from available fixed monitoring sites.

We conclude that this low-cost and versatile technology has the potential to provide a more complete assessment of the high-granularity air quality structure generally observed in the urban environment. When appropriately deployed, the methodology can also contribute to scientific and public policy issues, crossing climate, health/exposure, as well as air quality on local and regional scales, and has the potential to offer a new paradigm in air quality quantification and monitoring.

P-2-013 Source Apportionment of EC and OC in Beijing: Comparison between 14C Measurement and Chemical Transport Model

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Key words
carboneceous aerosol radiocarbon chemical transport model

For the source apportionment of carbonaceous aerosols, combined analysis using radiocarbon (14C) measurement and chemical transport model (CTM) is effective (Morino et al., EST, 2010). Concentrations of fine-mode aerosols, including elemental carbon (EC), organic carbon (OC) and 14C, were observed with a time resolution of 6 hours near the urban center of Beijing in June 2010. We evaluated the model performance of CTM (CMAQ v4.6, Byun and Schere, AMR, 2006) on simulation of EC and OC. The CTM well reproduced the diurnal variation of ozone and EC, while it largely underestimated OC concentration. From sector-based tag simulation, we directly compared observed and simulated TC (=EC+OC) from fossil and nonfossil sources. Fossil-TC was well reproduced by the CTM during nighttime and largely underestimated during daytime. By contrast, nonfossil-TC concentration was underestimated both during daytime and nighttime. This result suggests that the CTM largely underestimated fossil-secondary-OC (SOC) during daytime assuming EC to primary-OC (POC) ratio from fossil sources to be correct. Underestimation of nonfossil-TC suggests that the CTM underestimated POC from biomass burning or cooking and/or biogenic SOC. Further studies on their validation and model improvement are necessary. The CTM indicated that contributions from nonfossil sources were 20% and 60% to EC and POC, suggesting that biomass burning and domestic burning had important contributions to POC in Beijing. Emissions outside Beijing contributed by 25% and 50% to EC and POC.

P-2-014 Ambient ammonia and ammonium aerosol in Beijing urban area during summer 2009

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Key words

Ambient ammonia ammonium aerosol PM2.5 Beijing urban area

We present the measurement of ambient ammonia (NH_3), ammonium (NH^{4+}), sulfur dioxide (SO_2), sulfate (SO_4^{2-}), nitrogen oxide (NO_x), nitrate (NO^{3-}), and chloride (Cl^-) concentrations on July and August 2009 in Beijing urban area. The mean concentrations of NH_3, SO_2 and NO_x were 27.66, 11.31 and 98.79 ug/m3, respectively, and 13.47, 21.13, 12.79 and 1.69 ug m^{-3} for NH_4^+, SO_4^{2-}, NO^{3-} and Cl^- in PM2.5 during summer 2009. The diurnal variation of NH_3 showed the similar pattern to that of NH^{4+} aerosol with higher concentration during daytime and lower concentration at night. NH_3 and NH^{4+} concentrations were weakly correlated, implying that gas-particle reactions are influenced by many factors such as sources, meteorology and removal. NH^{4+} aerosol was primarily associated with SO_4^{2-} at the site. To gain an insight into the impact of transport on NH_3 and NH^{4+} levels at Beijing urban site, air mass backward trajectories were calculated and analyzed in combination with corresponding pollutants concentrations. The results indicate that transport of air masses from the North China Plain region and from the major coal mining regions west of Beijing is responsible for the high concentrations of NH_3 and NH^{4+}.

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P-2-015 A study of ozone control strategy using the OZIPR in the Seoul Metropolitan Area

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Key words

Seoul metropolitan area Ozone NOx VOCs Control strategy

To establish area specific control strategy for ambient ozone in the Seoul Metropolitan Area (SMA), the maximum ozone concentration in each local government in the SMA were estimated by using the OZone Isopleth Plotting Package for Research (OZIPR) model. The modeling domain was 26 local government districts in the SMA. The modeling period was June 2000 and the emission inventory data used were from National Institute of Environmental Research (NIER). OZIPR modeling results for five local government districts in the Seoul Metropolitan Area (SMA) in June 2000 were used for the sensitivity

analysis. Among the five local governments, Suwon was chosen to identify other variance through the period from April to September in 2000.
Except the islands of Incheon, whole Seoul and Incheon areas were volatile organic compounds (VOCs)-limited, i.e., decreasing the oxides of nitrogen (NO_X) emission alone may increase the maximum ozone concentration upto certain point. In Gyeonggi, 12 areas were VOC-limited while 12 areas were in between VOC-limited and NO_X-limited, i.e., decreasing both NO_X and VOCs emission may decrease the maximum ozone concentration. The sensitivity analysis result showed that the modeling result of the SMA being VOC-limited region be still valid for a wide range of input parameters' variation. The monthly modeling result showed different ozone values, but still showing the characteristics of VOC-limited region. The same calculation was carried out for June 2004 and it was found that Seoul was still in the VOC-limited condition. Thus, the ozone control strategies in Seoul Metropolitan Area should be made differently according to unique characteristics of each area.

P-2-016 Ground-based MAX-DOAS measurements of tropospheric NO_2 vertical column densities over Beijing: Seasonal, weekly and diurnal variations and satellite data evaluation

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Key words
tropospheric NO_2 MAX-DOAS Beijing megacity

Air quality in Beijing megacity has been of great concern in atmospheric and environmental science community as well as public media. Satellite measurements of tropospheric NO_2 were applied to evaluate the seasonal variation of urban air quality in Beijing and the effects of emission control measures during the November 2006 Sino-African Summit and the August-September 2008 Olympic Games. Considering the sensitivity of satellite observations to pollution located near the ground, as well as the uncertainties contained in satellite retrieval process, ground-based measurements of tropospheric NO_2 columns and its vertical distributions are needed to validate the satellite observations.
We have performed ground-based MAX-DOAS measurements in Beijing since 6 August 2008. Our measurement site (39.95°N, 116.32°E, 96 m a.s.l.) is located in the courtyard of China Meteorological Administration (CMA), Beijing. The compact and relatively small Mini MAX-DOAS instrument from Hoffmann Messtechnik GmbH of Germany has been operating quasi-continuously.

Measurements were made at 3°, 6°, 10°, 20°, 30°, 45°, and 90° elevation angles consecutively, with an integration of about one minute for each elevation angle. Owing to both the diurnal and seasonal variations in PBL, the diurnal variation patterns of tropospheric NO_2 are found to be rather different from one season to another. In contrary to previous studies, we found an apparent weekly cycle of the tropospheric NO_2 VCD over Beijing by investigating its diurnal variation patterns. The NO_2 VCD in the late afternoon was the largest on Saturday and the lowest on Sunday. Our data have shown clearly that the August tropospheric NO_2 VCDs were much smaller in 2008 than in the post years of 2009, 2010 and 2011, especially in the morning and early afternoon. The decreases in the tropospheric NO_2 VCD over Beijing during the 2008 Olympic Games are estimated to be 39-54%, with comparisons to the other years.

The tropospheric NO_2 VCDs derived by our ground MAX-DOAS measurements are shown to have to a good correlation with the SCIAMACHY and OMI satellite data. However, the satellite observations underestimate the tropospheric NO_2 VCDs over Beijing systematically, by 43% for SCIMACHY and by 38% for OMI, compared with the MAX-DOAS measurements. It is suggested that the aerosol pollution can be large enough to result in the underestimation of the tropospheric NO_2 VCDs over Beijing by SCIMACHY and OMI with respect to the ground-based MAX-DOAS measurements, while the smoothing effect could play an additional role.

P-2-017 Seasonal variation of source-receptor relationships of photochemical ozone to its precursor emissions over East Asia based on CMAQ-HDDM

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Key words
HDDM photochemical ozone

O_3 concentration over East Asia shows a clear seasonal variation due to the change of summer/winter meteorological condition (such as monsoon change) and emission activities. It is well reported that the seasonal change of O_3 concentrations over Japan is characterized by two peaks in spring and autumn, and minimum in summer. In East Asia, anthropogenic emissions are rapidly changing resulted from economic shifts and control measure advances, therefore, accurate examination of seasonal change of source-receptor (S-R) relationship of O_3 concentration over East Asia is an important environmental issue. HDDM (higher-order decoupled direct method) is an efficient and accurate sensitivity analysis technique to determine the S/R relationships of NOx and NMVOC emissions to O_3 formation. We applied the CMAQ-HDDM method to clarify the seasonal changes of S/R relationship. The base-case simulation of 4 months for January, April, July, and October in 2007 are performed prior to HDDM sensitivity simulation, and well

validated compared to the surface observation data. In sensitivity simulation by HDDM, based on the first- and second-order sensitivity coefficients representing the responsiveness of O_3 concentrations to anthropogenic emissions, we investigated the ozone sensitive regime in each season. We also focused on the seasonal dependency of S-R relationships from East Asian regions being transported to EANET (Acid Deposition Monitoring Network in East Asia) observation sites in Japan based on the zero-out source contribution (ZOC), where the ZOC is defined as the difference between the concentration of the base case simulation and the concentration that would occur if the source did not exist. We examined the source contributions of anthropogenic NOx and VOC emissions from China, Korea, and Japan based on ZOC during episodic pollution events occurred on Apr, 2007. The source contributions from anthropogenic emissions at EANET observation sites were dominated by China (6-16 %), followed by Japan (up to 5 %), Korea (up to 3 %).

P-2-018 Factor Analysis of Combined Organic and Inorganic Aerosol Mass Spectra from High Resolution Aerosol Mass Spectrometer Measurements

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Key words
Positive Matrix Factorization Organic Aerosol High Resolution Aerosol Mass Spectrometer Sources and Processes

Introduction: Organic aerosols (OA), a complex mixture from various sources, have highly uncertain impacts on radiative forcing and human health. A better understanding of sources and evolution processes of OA is of importance for assessing aerosol impacts and reducing the uncertainties in models. For the first time, positive matrix factorization (PMF) was performed to the combined organic and inorganic aerosol spectral matrices. The sources, mixing characteristics, acidity, and photochemical processing of OA and inorganic aerosols (IOA) are investigated.
Methods: An Aerodyne High-resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS) was deployed on the campus of Queens College in New York City in summer 2009 for in-situ measurements of the mass concentrations, chemical composition and size distributions of submicron aerosol species. The high-resolution (HR) mass spectral and error matrices of OA and IOA are prepared and combined into one unified dataset for PMF analysis.
Results and Discussion: PMF analysis resolved 8 factors in this study. The hydrocarbon-like OA (HOA) and cooking OA (COA) contain very minor

inorganic species, indicating the different sources and mixing characteristics between primary OA and secondary species. The two factors that are primarily ammonium sulfate (SO_4-OA) and ammonium nitrate (NO_3-OA), respectively, are overall neutralized, of which the OA in SO_4-OA shows the highest oxidation state (O/C = 0.69) among OA factors. The semi-volatile oxygenated OA comprises two components, i.e., a less oxidized (LO-OOA) and a more oxidized (MO-OOA). The MO-OOA represents a local photochemical product with the diurnal profile exhibiting a pronounced noon peak, consistent with those of formaldehyde (HCHO) and Ox. The much higher NO+/NO2+ fragment ion ratio in MO-OOA than that from ammonium nitrate alone provides evidence for the formation of organic nitrates. The amine-related nitrogen-enriched OA (NOA) contains ~25% of acidic inorganic salts, elucidating the formation of secondary OA from amines in acidic environments. The evolution processes of OA are further explored with the triangle plot and Van Kerevelen diagram.
Conclusion: Our results demonstrate that PMF analysis by incorporating inorganic aerosols is able to study the mixing of organic aerosols (OA) and inorganic species, the acidity of OA factors, and the fragment ion patterns related to photochemical processing. Thus, it is of great interest to perform the same analysis to other HR-ToF-AMS datasets.

P-2-019 Ozonesonde observation above Mexico City

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Key words
ozone Mexico

Air quality in Mexico City has improved significantly over the past decade due to various countermeasures such as relocation of factories, introduction of low-sulfur fuel and three-way catalytic converters, and registration of pollutant emitting plants. However, the overall level of air pollution is still higher than in developed countries such as Japan, and further efforts needs to be made. Particularly, ambient ozone, whose formation depends intricately on solar irradiation, air-mass transport, and distributions of precursor and destroying substances, and countermeasures against which have not been established, has to be investigated on aspects of meteorology, atmospheric chemistry, and information science. To understand the life cycle of ozone in the three-dimensional space, we conducted balloon-borne sounding in Mexico City and Japan.

Mexico City is situated on a highland basin at 19 degree latitude. The approximate elevation is 2300 m, and mountains up to 5636 m surround the city on the west, east and southwestern sides. The low latitude and high elevation causes high ultraviolet radiation. The rapid growth of the convection boundary layer in the basin together with the induced cold air flow through the northern and southeastern gaps constitute peculiar characteristics of mixing and ventilation of air pollutants in Mexico City. Major pollutant sources in Mexico City are the northern industrial area and the vehicular traffic all over the city.

Sounding balloons were released from Servicio Meteorologico Nacional (SMN) on 17, 22 and 23 November 2011, and 7, 8, 9, 12, 13 and 14 March 2012. Radiosondes were attached at 8:30 and 17:30, and ozonesondes at 11:30 and 14:30. The temperature on the observation days was not significantly different from the mean of the past 17 years. The convective boundary layer grew to about 3 km from the ground. The wind near the ground was mostly weak northerly, and the wind direction above the mixing layer was either westerly or easterly in November and always westerly in March. The wind speed was lower than in Japan, resulting in 30 - 40 km horizontal distance from the release point to the balloon-burst point at about 30 km ASL compared to 150 - 200km in Japan. The ground monitoring network showed higher ozone concentration in the southern part of the Mexico City basin, reflecting southward transport of the photochemically produced ozone. The vertical ozone profiles revealed production and mixing of ozone from pre- to post-noon. Except near the ground where ozone reacts with nitrogen monooxide, the ozone concentration was almost uniform in the convective boundary layer. Other notable features were lower ozone concentration in the upper air of Mexico City than in Japan, which is consistent with previous satellite observations, and sharp drops around 5km ASL observed in November.

P-2-020 Missing OH source in a suburban environment near Beijing : observed and modelled OH and HO_2 concentrations in summer 2006

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Key words
OH Beijing HO_2 RACM-MIM-GK LIF

Measurements of ambient OH and HO_2 radicals were performed by laser induced fluorescence (LIF) during CAREBeijing2006 (Campaigns of Air Quality Research in Beijing and Surrounding Region 2006) at the suburban site Yufa in the south of Beijing in summer 2006. On most days, local air chemistry was influenced by aged air pollution that was advected by a slow, almost stagnant wind from southern regions. Observed daily maxima of OH and HO_2 were in the range of $(4 - 17)\times10^6$ cm^{-3} and $(2 - 24)\times10^8$ cm^{-3}, respectively. During daytime, OH reactivities were generally high ($10 - 30$ s^{-1}) and mainly contributed by VOCs and their oxidation products. The comparison of modeled and measured HO_x concentrations reveals a systematic underprediction of OH as a function of NO. A large discrepancy of a factor 2.6 is found at the lowest NO concentration encountered (0.1 ppb), whereas the discrepancy becomes insignificant above 1 ppb NO. This study extends similar observations from the Pearl-River Delta (PRD) in South China to a more urban environment. The OH discrepancy at Yufa can be resolved, if NO-independent additional OH recycling is assumed in the model. The postulated Leuven Isoprene Mechanism (LIM) has the potential to explain the gap between modelled and measured OH at Beijing taking into account conservative error estimates, but still lacks experimental confirmation. This and the hereby unresolved discrepancy at PRD suggest that other VOCs besides isoprene might be involved in the required, additional OH recycling. Fast primary production of RO_x radicals up to 7 ppb h^{-1}

was determined at Beijing which was dominated by the photolysis of O_3, HONO, HCHO, and dicarbonyls. For a special case, Aug 20, when the plume of Beijing city was encountered, a missing primary HO_x source ($\approx$3 ppb h^{-1}) was determined under high NOx conditions similar to other urban areas like Mexico city. CAREBeijing2006 emphasizes the important role of OVOCs as a radical source and sink, and the need for further investigation of the chemical degradation of VOCs in order to better understand radical chemistry in VOC-rich air. Measurements of ambient OH and HO_2 radicals were performed by laser induced fluorescence (LIF) during CAREBeijing2006 (Campaigns of Air Quality Research in Beijing and Surrounding Region 2006) at the suburban site Yufa in the south of Beijing in summer 2006. On most days, local air chemistry was influenced by aged air pollution that was advected by a slow, almost stagnant wind from southern regions. Observed daily maxima of OH and HO_2 were in the range of (4–17)×106 cm−3 and (2–24)×108 cm−3, respectively. During daytime, OH reactivities were generally high (10–30 s−1) and mainly contributed by VOCs and their oxidation products. The comparison of modeled and measured HO_x concentrations reveals a systematic underprediction of OH as a function of NO. A large discrepancy of a factor 2.6 is found at the lowest NO concentration encountered (0.1 ppb), whereas the discrepancy becomes insignificant above 1 ppb NO. This study extends similar observations from the Pearl-River Delta (PRD) in South China to a more urban environment. The OH discrepancy at Yufa can be resolved, if NO-independent additional OH recycling is assumed in the model. The postulated Leuven Isoprene Mechanism (LIM) has the potential to explain the gap between modeled and measured OH at Beijing taking into account conservative error estimates, but still lacks experimental confirmation. This and the hereby unresolved discrepancy at PRD suggest that other VOCs besides isoprene might be involved in the required, additional OH recycling. Fast primary production of RO_x radicals up to 7 ppb h^{-1} was determined at Beijing which was dominated by the photolysis of O_3, HONO, HCHO, and dicarbonyls. For a special case, Aug 20, when the plume of Beijing city was encountered, a missing primary HO_x source ($\approx$3 ppb h^{-1}) was determined under high NO_x conditions similar to other urban areas like Mexico city. CAREBeijing2006 emphasizes the important role of OVOCs as a radical source and sink, and the need for further investigation of the chemical degradation of VOCs in order to better understand radical chemistry in VOC-rich air.

P-2-021 Recent Observations of High OH Concentrations and Rapid Ozone Formation in Biomass Burning Plumes Simulated with the ASP Model

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Key words
Biomass burning Ozone Hydroxyl radical HONO

Biomass burning is a major source of atmospheric trace gases and particles that impact air quality and climate at urban, regional, and global scales. Within minutes after emission, rapid, complex photochemistry within a smoke plume can cause large changes in smoke composition. In some plumes, this can lead to significant increases in the concentrations of ozone and aerosols after less than an hour of aging, while in other, generally boreal, plumes only small changes are observed. Being able to understand and simulate this rapid chemical evolution under a wide variety of conditions is thus a critical part of forecasting the impact of these fires on urban and regional air quality.

The Aerosol Simulation Program (ASP) has been previously used within a Lagrangian parcel model to simulate the formation of ozone and secondary organic aerosol (SOA) within several African and North American plumes. This work has suggested heterogeneous NO_x chemistry as a potential explanation for the rapid formation of ozone observed within several smoke plumes. However, these previous studies were limited by a lack of direct measurements of HONO, OH, or photolysis rates within the smoke plumes.

In this work, we will present ASP simulations of the chemical evolution of two recently sampled biomass burning smoke plumes. The first was sampled over the Yucatan during the 2006 Megacity Initiative Local and Global Research Observations (MILAGRO) campaign while the second was sampled over California during the 2009 San Luis Obispo Biomass Burning campaign. Both airborne experiments provide a rich data set of trace gas and aerosol observations for model testing and evaluation. This includes observations of several species, such as OH, and other important parameters, such as photolysis rates, which were measured for the first time in biomass burning plumes. We will discuss the sensitivity of the model simulations to uncertainties in the emissions, dilution rate, and gas- and particle-phase chemistry. We will then use these simulations to test several explanations for the high levels of OH and ozone observed within these smoke plumes, such as (1) the photolysis of HONO emitted directly by the fires, (2) the secondary formation of HONO from NO_x via heterogeneous chemistry within the smoke plume, and (3) the recycling of OH during the oxidation of some organic gases within the smoke plumes, as has been observed for isoprene.

P-2-022 CO profile information from SCIAMACHY and compare with model simulation

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Key words
Carbon monoxide SCIAMACHY

From SCIAMACHY near IR observations the total atmospheric column density of CO can be retrieved including also the surface-near layers. Due to the high radiance contrast difference between the clear and cloudy part of the satellite pixel in the near IR; even for rather small cloud fraction (about >10%), the averaged CO partial column/profiles can be derived.
In this study we apply the so called cloud slicing technique to SCIAMACHY CO observations, and present the partial columns of seasonal and global variation from SCIAMACHY observations for different, mainly polluted regions of the earth. We compare them to corresponding CO results obtained with the same procedure from atmospheric models. In general, good agreement is found; however, there are also systematic differences of vertical and horizontal transportation between both dataset.

P-2-023 Studies of the uncertainties in determining the urban area-wide fluxes of greenhouse gases

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Key words
greenhouse gases urban scale fluxes

We have been measuring the urban area-wide fluxes of CO_2 and CH_4, from the city of Indianapolis, Indiana, as part of the INFLUX campaign. INFLUX involves measurements of fluxes using a combination of approaches, including aircraft-based mass balance, use of $14CO_2$ and CO/CO_2 relationships, and inverse modeling. INFLUX involves accurate and precise measurements of CO, CO_2, and CH_4 from aircraft, and a network of tower platforms around the city. Here we focus on the impact of uncertainties in background concentrations, boundary layer height measurement, and sampling statistics on the overall measurement uncertainties. We will conclude with a discussion of how we might best conduct such measurements from "mega-cities".

P-2-024 Hg isotopic fractionation in utility boiler of large scale coal fired power plant

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Key words
Hg isotopes Coal-fired power plant

Coal-fired power plant (CFPP) emissions of mercury (Hg) represent the largest anthropogenic Hg source to the atmosphere. High precision Hg stable isotope analyses of coals suggest that coals from different geographic locations (e.g.

different countries, coal basins and coalfields) and different coal-forming periods display unique isotopic fingerprints. Variations up to 3‰ for delta 202Hg (representative of mass dependent fractionation, MDF) and 1‰ for capital delta 199Hg (representative of mass independent fractionation, MIF) have been observed across N-American and Eurasian coals. There is therefore a substantial interest in tracing CFPP Hg emissions at local, regional and global scales. However, CFPP operating conditions, Hg capture technologies and post-emission Hg transformations may potentially alter the original feed coal Hg isotope signatures. Here we investigate Hg isotope fractionation between feed coal and coal combustion products in six utility boilers of two large CFPPs in Huainan City, Anhui Province, China. We observed identical trends in all six boilers: relative to feed coal with delta 202Hg ranging from -0.67 to -0.18‰, 1) bottom ash is enriched in the lighter isotopes with delta 202Hg of -1.96 to -1.64‰, 2) fly ash recovered from electrostatic precipitators is enriched in the lighter isotopes with delta 202Hg of -1.91 to -0.82‰, 3) Hg rich gypsum recovered from a wet flue gas desulphurization system shows comparable values with delta 202Hg of -0.99 to -0.47‰. No MIF was observed during the transport and transformation of Hg inside the boiler. An isotope mass balance suggests that stack Hg emissions are slightly enriched in the heavy Hg isotopes relative to feed coal and that the enrichment depends largely on the Hg capture technology and emission factor. The implications of this study will be discussed in the context of tracing CFPP Hg emissions to the global environment.

P-2-025 Trace Gas Retrievals for GEO-CAPE Using DISCOVER-AQ Measurements

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Key words
GEO-CAPE DISCOVER-AQ air quality forecasting megacity ozone and its photochemical drivers

The Geostationary Coastal and Air Pollution Events (GEO-CAPE) mission is a US National Research Council-recommended mission identified in their Earth Science Decadal Survey. The atmospheric science objectives of GEO-CAPE are to identify human and natural sources of aerosol and ozone precursors and track air pollution transport. Observations of daily evolution of features from GEO-CAPE's geostationary platform will enable better assessment of population exposure as well as air pollution processes, thereby facilitating remediation of chronic pollutant exposure and also better forecasting.

In this presentation, we demonstrate the ability of multispectral measurements such as those expected from GEO-CAPE to retrieve O_3, NO_2 and HCHO with sensitivity to the boundary layer. For O_3, there is useful vertical definition, so that important regional transport effects may be tracked. For this purpose, we have developed an integrated retrieval tool that includes several modules: (1) solar and viewing geometry; (2) trace gas absorption cross section; (3) aerosol/cloud single scattering properties; (4) surface albedo/emissivity; (5) radiative transfer; (6) solar spectrum; (7) instrument model; (8) retrieval constraints; (9) inversion.

We also show that the retrieval capabilities of GEO-CAPE will provide useful information about air quality processes in a range of real-world conditions. For our evaluation, we use several profile measurements obtained from the DISCOVER-AQ (Deriving Information on Surface conditions from Column and Vertically Resolved Observations Relevant to Air Quality) mission; its first intensive focused on the region within and influencing the Boston-New York-Washington DC megacity agglomeration. DISCOVER-AQ measured concentrations of aerosols and trace gases from the surface to 4 km altitude. O_3 and its photochemical drivers (sampled as NO_2 and HCHO) were of particular interest to DISCOVER-AQ and our study. The DISCOVER-AQ measurements were combined with ozonesonde and Weather Research and Forecasting – Chemistry (WRF-Chem)-modeled profiles in the clean, less variable regions above 3.5 km.

P-2-026 Inverse Estimate of Long-Term CO Emission in China between 2005-2010 with Green’s Function Method

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Key words
Inverse estimate Carbon monoxide Chinese emission

Carbon monoxide (CO) emission amounts in China are inversely optimized with Green’s functions method, CO vertical profile measurements from MOPITT satellite instrument, and the GEOS-Chem chemical transport model (CTM) for the recent 6 years (2005 - 2010). Observations from surface sites (JMA and NOAA/GMD) are used for independent validation of a posteriori emissions. Model simulations with a posteriori emissions successfully reproduce the CO outflows from China to East China Sea and the Japanese archipelago in winter and spring, and compensate the under-estimates over the central eastern China region, considerably. A posteriori emissions in China exhibit significant seasonal variation in which the seasonal peak and bottom are found in winter-spring and summer, respectively. CO emission in March is on average 54 % higher than in August. This seasonal cycle is consistent with other recent studies. Chinese CO sources

obtained by the inversion are 164.5, 171.5, 180.8, 160.3, 152.5, and 156.1 Tg/year for 2005-2010, respectively, presenting inter-annual variations due to socioeconomic conditions (e.g., controls on pollutant emissions by the 2008 Beijing Olympic game and the global depressions in 2009).

P-2-027 Impacts of additional HONO sources on major aerosol components over the North China Plain

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Key words
HONO NO_y aerosol component heterogeneous reaction WRF-Chem model

Nitrous acid (HONO) is one of the major sources of the hydroxyl radical (OH) which is the key oxidant in the atmosphere. Direct emissions, gas-phase reactions, and heterogeneous reactions are generally considered as HONO sources, but the detailed formation mechanism of HONO is still under discussion. High HONO concentrations in urban or rural areas are frequently observed but most current air quality models, e.g., the Weather Research and Forecasting/Chemistry model (WRF-Chem), cannot reproduce HONO observations very well, particularly in the daytime, due to the only inclusion of gas-phase production of HONO. We have incorporated photo-excited NO_2 molecules, heterogeneous reaction on aerosol surfaces, and direct emissions of HONO into WRF-Chem. Five simulations were conducted to assess effects of each and the three additional HONO sources on concentrations of major chemical components. Percentage change calculations of major aerosol components and concentration ratios of gas NO_y (NO_{yg}) to NO_y and particulate nitrates (NO^{3-}) to NO_y were taken due to the three additional HONO sources in the North China Plain (NCP) in August of 2007. Results indicate that inclusion of the three additional HONO sources in WRF-Chem can confidently reproduce HONO simulations compared with observations. Heterogeneous reaction on aerosol surfaces is a key contributor to concentrations of HONO, NO^{3-}, NH^{4+}, and $PM_{2.5}$ over the NCP. The three additional HONO sources produce a ~5-20% increase in monthly mean daytime concentration ratios of NO^{3-}/NO_y, a ~15-52% increase in largest hourly mean concentration ratios of NO^{3-}/NO_y, and a

~10-50% increase in monthly mean concentrations of NO^{3-} and NH^{4+} in large areas of the NCP. For the Bohai Bay the largest hourly increases of NO^{3-}, NH^{4+}, and $PM_{2.5}$ can respectively exceed 90%, 80%, and 40% due to the three additional HONO sources. This implies that the three additional HONO sources can lead to aggravation of regional air pollution, further impairment of visibility, and enhancements to the incidence of haze in some industrialized regions with high emissions of NO_x and particulate matter under favorable meteorological conditions.

P-2-028 An investigation of AOD over Northeast Asia, combining chemistry-transport modeling results with geostationary COMS/GOCI-retrieved products through a data assimilation technique

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Key words
Long-range transport (LRT) Geostationary Ocean Color Imager (GOCI) Aerosol optical depth (AOD) Data assimilation Northeast Asia

In order to study the main characteristics of AOD over Northeast Asia, the AOD products retrieved from MODIS sensors onboard Terra and Aqua have been used with 3-D chemistry-transport modeling results. However, the MODIS sensor has a limitation in continuously monitoring aerosol optical properties, because its local overpass times are only once per day at 10:30 and 13:30 LST for Terra and Aqua, respectively. In order to overcome this temporal limitation of the Low Earth Orbit (LEO) satellites that pass over the region of interest once a day (or several days), a Korean geostationary satellite, Communication, Ocean, and Meteorological Satellite/Geostationary Ocean Color Imager (COMS/GOCI)-retrieved data were used in this study. Meanwhile, CMAQ modeling was also carried out for a period of 1 April to 31 May, 2011, in order to assess the impacts of Long-Range Transport (LRT) events on the particulate matter (PM) concentrations. The GOCI-retrieved AOD products were obtained through Yonsei aerosol retrieval algorithm and CMAQ model simulations considered dust and biomass burning emissions and their transports. The CMAQ-calculated AOD was then improved by integrating hourly GOCI-retrieved AOD via a data assimilation technique for the purpose of producing more accurate AOD products over Northeast Asia. It clearly showed the several long-range transport events of the small- and large-scale AOD plumes from Central East China (CEC) to the Korean peninsula. In addition,

according to statistical analysis of the assimilated AOD for the LRT and non-LRT events at five AERONET sites, the average AOD increased by LRT events was found to be 0.63 above the background AOD value (0.31) (approximately 200% increase of average AOD). Ultimately, this research aims to a development of chemical weather forecast system over Northeast Asia and the Korean Peninsula. It is also expected that this system will also be useful for the applications of the data from the GOCI-2 and Geostationary Environmental Monitoring Spectrometer (GEMS) sensors, both of which will be launched in the year of 2018 to monitor the air quality over the Asian regions including entire China, Korea, Japan, Southeast Asia, and parts of India and Central Asia.

P-2-029 Preliminary result of near-surface urban temperature inversion observed at Taipei 101 Skyscraper and its relationship to particulate matter mass concentration in Taipei City

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Key words
skyscraper balloon radiosonde particulate matter mass concentration

This study is part of the 3-year intensive observation project of secondary organic aerosols (SOA) in the mega cities of Taiwan. We observed the near-surface boundary layer status for studying SOA vertical transportation. Taipei 101 Skyscraper (508 m), the secondary tallest commercial building around the world, is the metropolis landmark of Taipei, Taiwan. The minute-resolution of wind field, air temperature and humidity sampled at the northeastern corner of 34th (151 m) and 82th (353 m) balconies are collected from January of 2011 until now for studying urban meteorological boundary layer and its relationship to PM mass concentration in city. 31 sets of balloon-borne radiosondes near Taipei 101 Skyscraper were launched for comparing skyscraper balconies' measurements. Grimm aerosol spectrometer was temporarily used to measure PM mass concentration during monthly instrument maintain at Taipei 101.

P-2-030 Analysis of criteria gas monitoring data between Mexican and Japanese cities

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Key words
monitoring criteria gas Mexico Japan

The relevance of air monitoring lies in its utility to determine quantitatively the atmospheric pollutant concentrations in specific areas. These concentrations could be compared with the maximum-permissible-levels that can be exposed the population according to the legislation that applies or can be used to establish and update standards for the improvement of air quality.

Mexico and Tokyo is representative of megacities in the world due to the concentration of population, whereas Guadalajara and Monterrey cities in Mexico and Ehime Prefecture in Japan are considered suburban areas.

In Mexican Cities, environmental problems are increasing in recent years. In 2000, Mexico City Metropolitan Area reported the highest number of days (304), on which the Mexican standard for environmental exposure to ozone was exceeded, whereas in Guadalajara and Monterrey city, ozone concentrations have incremented by 11% and 22%, respectively, in the last 5 years (SEMARNAT-INE, 2011).

Mexico City data were taken from the web page of the Secretary of the Environment of Federal District Government. In the case of Guadalajara and Monterrey data were downloaded directly from data base of the National Institute of Ecology.

Japanese data were obtained from the hourly database collected by the National Institute for Environmental Studies. The time period considered for the analysis was according to the Japanese fiscal year 2009 (April 2009 to March 2010). However, in Monterrey case the period was until December 2009, due to the data for 2010 are not available. For diurnal variation analysis the period chosen was from April to December 2009, in order to do an affair comparison.

We compare concentrations of air pollutants: those labeled as criteria gases in both countries (sulfur dioxide, carbon monoxide, particulate matter, nitrogen dioxide and ozone) Mexico and Japan. As representative areas, we chose Guadalajara, Monterrey, and Mexico City from Mexico, and from Japan, Tokyo metropolitan area and Ehime prefecture. For pollutants like NO_2, CO and $PM_{2.5}$, Mexican cities show higher concentration levels than Japanese cities, in the case of SO_2 the highest concentration depends of the season, while for O_3 the levels in the two countries are not significantly different.

As a result of our research, we can observe that Mexico City exhibits an increment of SO_2 concentration during fall and winter seasons, there is an increased emission by sources such as biomass burning and fossil fuel consumption, which are, according to SIMAT, 2010 principal sources of SO_2 in Mexico City. From April to September, SO_2 concentration in Ehime is higher than in Mexican areas. This can be due to the amount of industrial establishment in Ehime area; it has industries related to shipbuilding, chemicals, and oil refining, paper and textile products. Additionally to this analysis of gas monitoring data we count with a meteorological analysis in the chosen period. On the other hand, it is necessary to propose actions for mitigation of these pollutants and others not included in this work and to do research the principal sources in each area.

P-2-031 Partitioning of atmospheric carbon dioxide over central Europe: insights from combined measurements of CO_2 mixing ratios and their carbon isotope composition.

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Key words
Carbon Dioxide Flux partitioning

1. Introduction.

The observed changes in the accumulation rate of carbon dioxide in the atmosphere reflect the net balance between ocean and land CO_2 fluxes. However, observations of atmospheric CO_2 mixing ratios alone cannot provide information on partitioning between biogenic and fossil-fuel related fluxes. Such apportionment can be accomplished through quantification of fossil-fuel related CO_2 loads derived from ground-level measurements of atmospheric $^{14}C/^{12}C$ isotope ratios. A number of studies have demonstrated the strength of using the radiocarbon content in atmospheric carbon dioxide as a tool for quantifying fossil-fuel related components of atmospheric CO_2 budget on both small and large scales. Attempts were also made to use the $^{13}C/^{12}C$ and $^{18}O/^{16}O$ isotope ratios of atmospheric carbon dioxide for partitioning of local CO = sources in urban and rural environment.

2. Sites.

Through combined measurements of the carbon isotope composition of atmospheric CO_2 and its mixing ratios performed on regular basis at two sites of contrasting characteristics (city of Krakow and Kasprowy Wierch remote mountain station) we quantify on a monthly basis the fossil-fuel related and biogenic components of the atmospheric CO_2 load at both locations and their seasonal variability.

3. Methods.

Automated gas chromatographs (Agilent 6890) were employed at both sites to measure atmospheric CO_2 mixing ratios. For measurements of isotope ratios sampling of atmospheric CO_2 was performed at both sites using a method based on sorption of CO_2 on molecular sieve. The $^{13-}$C content of the collected CO_2 was determined using dual inlet isotope ratio mass spectrometry (Delta S, Thermo Finnigan). The radiocarbon content in the collected CO_2 was measured using benzene synthesis followed by liquid scintillation spectrometry.

4. Results.

Assuming that the measured atmospheric CO_2 mixing ratio at a given site consists of three components: (i) regional background component, which provides the bulk of the atmospheric CO_2 load, (ii) biogenic component, and (iii) fossil-fuel derived component , when the carbon isotope composition of atmospheric CO_2 is measured in addition to atmospheric mixing ratio, the mass and isotope balance equations can be formulated and resolved. The concentration and carbon isotope ratios of atmospheric carbon dioxide measured regularly at Krakow and Kasprowy Wierch sampling stations supplying the data necessary for calculation of CO_2 partitioning. Additional data were taken from NOAA GLOBALVIEW database and measurements performed by IUP University of Heidelberg at Jungfraujoch station.

5. Discussion.

The most characteristic feature of the partitioning of the CO_2 load in the local atmosphere of Krakow is the distinct seasonality of its fossil-fuel related and biogenic components. The fossil-fuel component varies between ca. 20 ppm in winter and approximately 7 ppm in summer. There is also a significant inter-annual variability observed, both with respect to the maximum and minimum values of this component. In addition, the fossil-fuel component reveals a slight decreasing trend of 0.82 (0.38) ppm per year. The biogenic CO_2 component derived for the Krakow atmosphere also reveals a distinct seasonality, although shifted in phase by ca. 6 months with respect to the fossil-fuel component. The maximum contribution of biogenic CO_2, up to ca. 30 ppm, was observed during summer and the early autumn months while distinct minimum (around ca. 10 ppm) occurred during winter and early spring.

6. Conclusion.

It turned out that in a typical urban setting in central Europe such as the city of Krakow, with its numerous distributed sources of CO_2 emissions associated with burning of fossil fuels, the biogenic CO_2 component is of the same magnitude as the fossil-fuel related component.

P-2-032 Estimating secondary organic aerosol concentration and its radiative forcing in East Asia

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Key words

Secondary Organic Aerosol Volatility Basis Set Radiative forcing

Secondary organic aerosols (SOA) comprise a large proportion (50~90%) of organic aerosols that typically constitute significant mass fractions of the total dry fine aerosols in the atmosphere. East Asia is one of crucial regions for organic aerosols whose primary anthropogenic sources account for about one fourth of the global emissions. However, the contributions of SOA to organic aerosols as well as total aerosol concentrations have not yet been assessed in a regional scale in East Asia. We use here a 3-D global chemical transport model, GEOS-Chem, to examine the production and distribution of SOA in East Asia and further to quantify its contributions to aerosol mass concentrations as well as its climate effects. We first improve the SOA simulation of the two-product approach in GEOS-Chem by applying the volatility basis set (VBS) approach and evaluate the model by comparing against observations – global AMS data and water soluble organic carbon (WSOC) data at the Gosan site in Jeju island, Korea for 2009. A global comparison of the model against the AMS data show improved agreements using the VBS method relative to the two-product approach. We find also that the improved model simulates higher annual mean SOA concentrations (0.33 versus 1.20 µg m^{-3} with two-product versus VBS methods, respectively) and are in much better agreements with the observed WSOC concentration (1.50 µg m^{-3}) at the Gosan site in Jeju island. Resulting annual mean radiative forcings of SOA are -0.69 W m^{-2} and -0.93 W m^{-2} at the TOA and at the surface, respectively. Our analysis reveals a significant contribution of SOA to aerosol mass concentrations in the free-troposphere and in surface air on an annual and regional mean basis in the model, indicating important implications of SOA for regional air quality and climate over East Asia.

P-2-033 The effects of particle pollution control implementation during the Asian Games in Guangzhou, China

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Key words

$PM_{2.5}$ chemical composition secondary formation Pearl River Delta region 2010

Asian Games

In order to investigate the particle pollution improvement during the 2010 Asian Games in Guangzhou, the comprehensive field campaign PRIDE-PRD 2010 was conducted in Pearl River Delta (PRD) region during Nov.11th to 30th, in which 24-h $PM_{2.5}$ and PM_{10} samples were collected in urban site, Guangzhou, and its downwind rural site, Heshan. The major chemical components in $PM_{2.5}$ of organic carbon (OC), elemental carbon (EC) and inorganic water-soluble ions were measured by using Sunset EC/OC analyzer and ion chromatography. The average $PM_{2.5}$ concentrations in Guangzhou and Heshan were 73.3 μg m^{-3} and 98.2 μg m^{-3}, respectively. PM_{10} was also measured in Heshan with the concentration of 131.6 μg m^{-3} and the ratio of $PM_{2.5/}$ PM_{10} was 75%. Compared with the previous study during Oct.4th to Nov 5th 2004 (PRIDE-PRD 2004), $PM_{2.5}$ decreased obviously by 29.6 μg m^{-3}, and it indicated that the air pollution control measures for the Asian Games were effective in Guangzhou urban area. Carbonaceous aerosol (OM+EC) was an important component in $PM_{2.5}$ in both Guangzhou and Heshan, which Organic matters (OM), accounting for 31% and 26%, and EC accounting for 5% and 6%, respectively. Roughly estimation of POC and SOC based on EC tracer methods were accounting for 15% and 4%, 13% and 3% in $PM_{2.5}$ in Guangzhou and Heshan, respectively. In addition secondary inorganic ions (sum of SO_4^{2-}, NO^{3-} and NH^{4+}, named as SNA) accounted for 39% and 42% of $PM_{2.5}$ in Guangzhou and Heshan, respectively. In total secondary formation (sum of SNA+SOA) accounted for appropriately 50% in $PM_{2.5}$ both in Guangzhou and Heshan, suggesting secondary particle pollution was important in the PRD region. Compared with the results of campaign PRIDE-PRD 2004, EC concentration decreased 3.5μg m^{-3} (by 49%) in $PM_{2.5}$ in Guangzhou, implying the primary emission was controlled in a great degree. The decrease of SO_4^{2-} implied stationary sources (e.g. coal combustion) were reduced in a certain extent in the PRD region. The experience of air pollution control during the Asian Games indicated that enhancement the primary emission control benefited reduction of both primary and secondary pollution, while regional secondary formation control will be focused on sooner in the future.

P-2-034 In-plume secondary pollutants formation from the Greater Paris region and its impact to surrounding regions

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Key words
secondary pollutants formation megacity CHIMERE modeling regional pollution impact

Megacity primary emissions such as NO_x, primary carbonaceous species have important impact on local air quality, while the secondary pollutants such as ozone, secondary inorganic aerosol and secondary organic aerosol produced related to these species can affect regional air quality due to their long life time. In the framework of the FP7 / EU project MEGAPOLI, an intensive campaign was launched in the Greater Paris Region in July, 2009 and in January/February 2010. The major objective was to quantify different sources of organic aerosol (OA) within a megacity and in its plume. The state-of-the-art regional chemical transport model (CTM) CHIMERE has been applied on the simulation of these campaigns. The results were evaluated with the detailed aerosol and gaseous precursor measurements conducted at an urban and two sub-urban sites. And a satisfying agreement was found for NO_x, VOC species, O_3 and inorganic and organic aerosol. In this work, the regional CTM CHIMERE is evaluated with airborne measurements from the French ATR-42 research aircraft (for plume characterization) during the 1-month's summer campaign. Modeled BC plume is evaluated as a primary urban tracer which indicates the location of the urban plume and is relatively similar as observed BC plume. Modeled O_3 is in good agreement with the observations and most affects downwind regional air quality at about 100 km away.

Modeled in-plume and background sulfate is sometimes overestimated. Large values are found not only within the Paris plume, but also outside, due in particular to ship emissions and industrialized harbors. Both modeled and observed nitrate show low background levels due to low levels of ammonia concentrations in the Paris region, as well as relatively high temperature in summer which shifts thermodynamic equilibrium to gas phase. There are weak plumes due to Paris emissions, affecting downwind regions.

Modeled organic aerosol is close to the measurement when the volatility of POA and chemical aging of semi-volatile VOC are taken into account. Slopes of the plume OA levels versus O_x ($=O_3+NO_2$) indicate SOA formation normalized with respect to photochemical activities. Simulated and observed slopes are in good agreement. Biogenic secondary organic aerosol contributes to the background level, while anthropogenic secondary organic aerosol (SOA) formation in the urban plume most contributed to the plume OA. On the average, OA levels increase by at least 10% due to Paris emissions at a distance of up to 200 km downwind of the agglomeration. For specific plumes, the transport distance can be even larger.

P-2-035 Assessing the contribution of atmospheric reactions to the concentrations of oxy-PAHs in the atmosphere

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Key words
PAH Quinone

Polycyclic aromatic hydrocarbons (PAHs) exist ubiquitously in the environment and are formed through high-temperature incomplete combustion and pyrolysis of fossil fuels and other organic materials. PAHs have been extensively studied since the mid 1970s owing to their potential mutagenic and carcinogenic properties. Recently, increasing interest has been paid to oxygenated derivatives of PAHs, particularly quinones, which are reported in being more toxic than their parent PAHs.

Both gaseous and particulate matter (PM) associated quinones, like PAHs, may be released into the atmosphere through incomplete combustion processes in diesel / gasoline engines, incomplete combustion of waste and biomass burning. Gaseous and heterogeneous atmospheric processing of PAHs can yield further quinone products via photochemistry and reactions with atmospheric oxidants (OH, NO_3 and O_3) or through biological transformation. However, quantitative data are limited on the relative importance of primary and secondary emissions of quinones.

In this study we report the simultaneous airborne measurements of 17 PAH and 11 quinone compounds in both gas and particulate phases; including the first observations for 2 methyl-1,4-naphthoquinone, 2-methylanthraquinone, 2,3 dimethylanthraquinone, benzo(a)pyrene-6,12-dione and benzo(a)pyrene-1,6-dione. Denuder, filter and PUF samples were collected simultaneously from urban roadside and suburban locations and independently from a remote site. Samples were analyzed using a recently developed GC-MS analytical method measuring 6

of the quinones in their derivatived forms.

The measured concentrations for PAH and quinone compounds range from 0.02 - 18 ng m^{-3} and 0.02 - 7 ng m^{-3}, respectively. Trafficked source profiles are calculated and presented from the simultaneous roadside and suburban measurements, indicating that vehicle emissions act as a major source for quinone compounds. Photochemical trajectory modeling and quinone to parent-PAH (Q:P) ratios are used to assess the long range chemical transport and relative contribution of potential sources for measurements made at the remote site. Larger Q:P ratios combined with back trajectories indicate that quinones measured from air masses moving from relatively clean environments may be products of atmospheric reactions.

P-2-036 Source attribution of background ozone concentrations in the US Intermountain West

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Key words
Background ozone Source attribution Ozone exceedance

The North American background ozone, defined as surface ozone concentrations in the absence of North American anthropogenic emissions, provides a baseline for risk and exposure assessments in setting the air quality standard. It represents the maximum ozone reduction that would be achieved through North American domestic emission controls. Sources of the North American background ozone include intercontinental ozone transport, as well as natural sources from the stratosphere, lightning, and wildfires. Better quantifying these sources is of importance as the ozone air quality standards become more stringent. We will present three-year statistics (2006-2008) of background ozone concentrations over the US calculated using the GEOS-Chem global 3-D model of atmospheric composition with1/2 degree by 2/3 degree horizontal resolution. We focus on the US Intermountain West where the background ozone is particularly high due to high elevation, arid terrain, and large-scale subsidence. The model is successful in reproducing the measured ozone concentrations and variations in the Intermountain West. It reproduces the ozone exceedances up to 70 ppbv, but it cannot reproduce exceptional high-ozone events associated with stratospheric intrusions or wildfires. We show the model captures the timing of springtime stratospheric intrusions, but underestimates their magnitudes and does not resolve the fine structures of the intrusions. Model simulation of wildfire ozone impacts is highly sensitive to the spatial and temporal resolution of the wildfire emission

inventory. We will also discuss the impacts from lightning emissions, and investigate their uncertainties in the model estimation.

P-2-037 Source contributions to seasonal vegetative exposure to ozone in the US

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Key words
ozone source apportionment

Frequent exposure to high levels of ozone leads to vegetation damage and can result in substantial economic losses. Cumulative ozone exposure metric, W126, has recently been proposed by the US EPA for use as a secondary ozone standard. Information on source regions contributing to the non-attainment of this standard is crucial for developing a successful strategy to mitigate the negative effects of ozone on vegetation. In this work we quantify W126 source contributions for the US regions exceeding the proposed levels of the W126 standard using the adjoint of the global chemical transport model GEOS-Chem. Our calculations are performed for a three-year period (2006–2008) and the adjoint sensitivities are adjusted on an hourly basis using the ozone observations from the Air Quality System. Given that the W126 metric is highly non-linear and that the adjoint model provides only first order results, we explore the robustness of the calculated sensitivities and the range over which they are applicable.

P-2-038 Organic Speciation of Fine Particulate Matter in Hong Kong: Quantification by Thermal Desorption-GC/MS and Implication on Source Identification and Apportionment

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Key words
NPOCs $PM_{2.5}$ organic tracer source apportionment

Sources of ambient organic aerosols (OA) come from both primary emissions and secondary formation in the atmosphere from oxidation of various volatile organic compounds. Identifying sources and quantifying their contributions is an essential initial step of air quality management. Ambient OA consists of numerous individual organic compounds and a number of nonpolar organic compounds (NPOCs) are unique to certain primary OA sources and thereby can serve as tracers in source identification and apportionment.

NPOCs in ambient particulate matter (PM) commonly include n-alkanes, branched

alkanes, hopanes and steranes, and polycyclic aromatic hydrocarbons (PAHs). We have developed an in-injection port thermal desorption-GC/MS (TD-GC/MS) analytical technique for measurements of NPOCs. This method significantly reduces the analysis time and the labor involved in the sample pretreatment step in the traditional method of solvent extraction (SE) followed by sample concentration.

Sampling campaigns were conducted during the period from November 2010 to February 2012 in Hong Kong. Four air quality monitoring stations were selected including one suburban site (HKUST campus), two urban sites (Tsuen Wan and Yuen Long), and one roadside site (Mong Kok). The TD-GC/MS method was deployed to quantify NPOCs in approx. 200 high-volume $PM_{2.5}$ samples collected in the campaigns.

With the large NPOC data set as well as the measurements of elemental carbon and organic carbon, the interspecies relationships between select NPOCs and their concentration ratios to elemental carbon (EC) among the ambient samples were examined.

Select PAH/EC ratio-ratio plots reveal that PAHs had diverse sources and vehicular emissions were unlikely a dominant source for PAHs in Hong Kong. Comparison of the ambient data and source sample data shows that biomass burning and other regional sources likely dominated large PAHs in ambient Hong Kong. The spatial variation pattern confirms hopanes to be primarily from vehicular emissions and the ratio-ratio plot analysis shows that gasoline-powered engines are the dominant source for the hopane compounds. The hopane/EC ratio in ambient samples, however, was found to be on average less than half of the ratio in vehicular emissions-dominated source samples. This suggests that significant atmospheric oxidation of hopanes have occurred after their emissions in Hong Kong's urban atmosphere and highlights the necessity in considering oxidation loss in applying organic tracer data in source apportionment studies. $PM_{2.5}$ and OC are apportioned to sources using the Chemical Mass Balance (CMB) and Positive Matrix Factorization (PMF) receptor models in this study. The bias caused by the oxidation of organic tracers on the source apportionment results will be discussed.

P-2-039 Evaluation of tropospheric NO_2 columns over Beijing retrieved from OMI using multi-seasonal MAX-DOAS measurements

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Key words
OMI MAX-DOAS tropospheric NO_2 columns evaluation temporal and spatial multi-seasonal

Vertical column densities (VCDs) of tropospheric nitrogen dioxide (NO_2) retrieved from space, while useful for understanding the characteristics of nitrogen and other related pollution, likely contain large random and systematic errors. Such errors have rarely been quantified over China due to lack of more accurate measurements. In this study, NO_2 columns retrieved from the Ozone Monitoring Instrument (DOMINO v2) are evaluated with ground-based measurements from July 2008 to April 2009 using a Multi Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) instrument sited at the Institute of Atmospheric Physics (IAP) in urban Beijing. The multi-seasonal measurements allow for a more comprehensive evaluation under various meteorological and chemical conditions. Differences in temporal and spatial representation between satellite and ground based measurements are taken into account.

The two measurements are consistent, with the slope and correlation approaching to unity under the linear temporal fitting, when the spatiotemporal representativeness is strictly accounted for. Loosening the spatiotemporal constraint reduces the slope and correlation to 0.5-0.65 and 0.6-0.8, respectively. Furthermore, it is found that the surface albedo used in OMI retrievals is likely overestimated over parts of Beijing compared to values from The Moderate Resolution Imaging Spectroradiometer (MODIS)/Aqua, serving as a potential cause of differences between OMI and MAX-DOAS measurements. Removing the data affected by the suspicious albedo increases the slope by about 0.2 and consequently improves the comparison between space and ground retrievals. More detailed analyses are under way for effects of surface albedo and other atmospheric parameters (clouds, aerosols, etc.) on space-retrieved NO2 columns.

P-2-040 The effects of boundary layer height and surface reflectance in the relation between AOD and particulate matter

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Key words
AOD MODIS

It has been suggested that satellite aerosol optical depth (AOD) retrievals can provide a surrogate for aerosol concentrations in the boundary layer. We explore whether this is applicable for Santiago of Chile by comparing the Moderate

resolution Imaging Spectroradiometer (MODIS) AOD product with 10 years of in situ PM10 and $PM_{2.5}$ measurements. As a result, an opposite seasonality was found. In order to investigate the reasons for this disagreement, we introduce a simple aerosol model to estimate AOD. The simulated AOD was compared with the MODIS data and with the AOD data collected by sunphotometers. The main sources of error appear to be an inadequate estimate of the surface reflectance and aerosol properties in the MODIS algorithm. Cirrus clouds do not appear to play a relevant role explaining the seasonality. A similar bias has been reported over other regions of the world.

P-2-041 Improving remote sensing of sulphur dioxide over Central Chile

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Key words
Sulphur dioxide Remote sensing

Over Central Chile, there are significant sources of sulfur dioxide (SO_2) such as copper smelters, carbon power plants, volcanoes and oceanic biogenic processes. It has been estimated that the year 2005 the main copper smelters Caletones, Ventanas and Chagres emitted 77GgS. The Ozone Monitoring Instrument (OMI) provides since 2004 daily estimates of the SO_2 burden over Central Chile. These retrievals may provide monitoring of SO_2 sources with a background noise of 0.2 DU, but they require careful evaluation given the complex terrain and circulation. A key parameter is the so called Air Mass Factor (AMF) which converts slant columns in vertical columns. The AMF depends of SO_2 profile and total ozone in the column, among other things. We aim at improve the retrieval by calculating the AMF assimilating vertical SO_2 profiles calculated with a high resolution (10km horizontal) chemical transport model. We simulate the satellite signal and study its sensitivity to the vertical stratification of SO_2 using a radiative transfer model. We focus over Central Chile where anthropogenic sulfur sources are constrained with a 20% -30%. The results show that taking into account the complex circulation, the AMF value change up to 20%   60% along Central Chile, when compared with the operational value.

P-2-042 Regional sources apportionment of carbonyls in Beijing

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Key words
carbonyls emission ratio sources apportionment PMF

Carbonyls play a significant role in atmospheric photochemistry. They are important intermediates in the photo-oxidation progress of hydrocarbons, serve as a major source of free radicals, and precursors of other secondary pollutants. So it is very significant for obtaining information on their sources.

In this work, active sampling with 2,4-dinitrophenylhydrazine (DNPH) absorbent cartridge and high performance liquid chromatography (HPLC) analysis method was developed for measurement of 22 carbonyls, including 19 aldehydes and 3 ketones. Five field campaigns at 38 sites within Beijing and its surrounding areas were conducted on June 24, July 22, August 22, September 14 in 2010, and January 13 in 2011. Two samples were taken on every of these five days with one in 9:00-12:00 and the other in 13:00-16:00. A multivariate positive matrix factorization (PMF) receptor model was attempted to apportion the sources of ambient carbonyls in Beijing. The ratio of PMF identified primary carbonyls to CO was compared with emission inventories to evaluate the result of source apportionment. Temporal and spatial variations of carbonyls sources were discussed and differences between summer and winter were also investigated.

In summer times, seven PMF factors were distinguished, including fuel evaporation, liquefied petroleum gas (LPG) vehicle emission, gasoline vehicle emission, diesel vehicle emission, industry + solvent, biogenic emission + the oxidation products, and secondary products + background. Secondary products + background was the major contributor for total carbonyls (over 50%), followed by diesel vehicle emission (15.4%), industry + solvent (10.7%), and biogenic emission + the oxidation products (7.5%). The contributions from the rest three factors were minor. The contribution percentage of primary emission to C1 - C4 aldehydes agrees well with previous observation based studies in Beijing. Primary fraction of carbonyls showed much better correlations with CO than those obtained from total carbonyls. And the emission ratios calculated from linear regression of primary carbonyls to CO agree well with the TRACE-P inventory for formaldehyde and ketones groups, whereas the calculated emission ratio of C2-C5 aldehydes group is 127% high than inventory. Carbonyls fractions from different factors exhibited temporal and spatial variations. Diesel vehicular emissions contributed more carbonyls in the morning due to larger traffic density, whereas carbonyls from biogenic sources were higher in the afternoon related to higher temperature and more intensive irradiation. From the perspective of spatial variations, carbonyls from diesel vehicles were higher in urban and southern

suburban area, whereas biogenic sources contributed larger mixing ratio in surrounding suburban areas with high forest coverage. The secondary products + background factor have a similar spatial pattern with non-methane hydrocarbons (NMHCs) in the afternoon. This might indicate the relationship between secondary products with their precursors. Contrary to source apportionment results in summer, primary emissions played more important roles in winter. In winter, only three factors were identified. Coal combustion emitted 54.9% of total carbonyls, and the contribution of traffic source and other unidentified sources were 21.4% and 24.0%, respectively.

P-2-043 Volatile organic compounds emission ratios in Beijing and comparison with an emission inventory

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Key words
Volatile organic compounds emission ratios evlauation on emission inventory Beijing

Volatile organic compounds (VOCs) play a significant role in photochemical formation of ground-level ozone and secondary organic aerosols (SOA), and therefore understanding their emission characteristics is essential to reduce ambient levels of secondary pollutants. Regional measurements of hydrocarbons and carbonyls were conducted from July 2009 to January 2011 at 27 locations in Beijing. Additionally, VOCs were continuously measured by an online dual gas chromatographic instrument equipped with flame ionization and mass spectrometer detectors (GC-FID/MSD) and a proton transfer reaction mass spectrometer (PTR-MS) in summer (from 5 August to 20 September, 2011) and winter (from 29 December, 2011to18 January, 2012) at an urban site (PKU).
Emission ratios for anthropogenic VOCs relative to CO were derived from these three data sets, respectively, and compared with each other, in order to investigate seasonal differences of VOCs emission ratios and to evaluate whether VOCs measurements at PKU site could represent VOC emission characteristics in entire Beijing region. During winter, higher emission ratios for ethene and benzene were observed, indicating the influence of coal combustion processes. Most VOCs emission ratios obtained at PKU site during summer generally agree within a factor of two with the values from regional measurements, but obvious underestimations

were found for benzene, toluene, and C5-C6 alkanes. The emission ratios calculated based on regional measurements were used to evaluate the accuracy of VOCs speciation in an anthropogenic emission inventory. For most of measured VOC species, their calculated emission ratios are within a factor of two with the values from TRACE-P emission inventory, suggesting that the chemical speciation of anthropogenic VOCs emission in Beijing appears well understood by this inventory. However, relatively larger discrepancies were found for benzene, C6-C12 alkanes, C3-C6 alkenes, C8-C10 aromatics, and C2-C5 aldehydes. The emission ratios of benzene and C2-C5 aldehydes were underestimated by 45% and 56% in this inventory, respectively. By contrary, calculated emission ratios for C6-C12 alkanes, C3-C6 alkenes, and C8-C10 aromatics are were significantly lower than the values in TRACE-P inventory. Heavy alkanes and aromatics are important precursors to SOA, while C8-C10 aromatics, C2-C6 alkenes and C2-C5 aldehydes play important roles in ozone photochemical formation, and therefore further research on these species emission and measurement is needed in the future.

P-2-044 Ammonia emissions from a representative in-use fleet of vehicles in guangzhou

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Key words
ammonia in-use vehicles emissions rates Guangzhou

Emissions rates of ammonia (NH_3) are reported for a fleet of 130 light-, medium-, and heavy-duty vehicles recruited in the city of Guangzhou, China. NH_3 measurements were performed using Nessler's Reagents spectrophotometry and nationwide standard chassis dynamometer test cycles required by Chinese EPA. Emissions of CO and NO_x were also measured during these test cycles. Emission factors of NH_3 were calculated for each type of vehicle and used to estimate the total emissions of NH_3 from motor vehicles in Guangzhou in 2009. Emission factors of NH_3 show large variations among different categories of vehicles, with a range from 4 to 138 mg km-1. The average emissions of NH_3 in Guangzhou in 2009 were estimated to be 983 t, with a range from 373 t to 2136 t. In addition, it was found that vehicles with the highest NH_3 emission rates possess the following characteristics: medium- and heavy-duty vehicles, certified with out-of-date

emission standards, mid-range odometer readings, and higher CO and NO_x emission rates. The results of this study will be useful for developing NH_3 emissions inventories in Guangzhou and other urban areas in China.

P-2-045 An Observation-Based Model for Secondary Inorganic Aerosols

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Key words
Observation-based model Secondary inorganic aerosol Process analysis

A mechanistically complete and time-dependent model, Observation-Based Model (OBM) for Ambient Particulates, is developed for resolving chemistry processes, thermodynamic equilibriums and size distribution of secondary inorganic aerosols (SIA) in an ambient atmosphere. OBM-SIA uses in-situ atmospheric and meteorological observations to determine the sensitivity of sulfate and nitrate concentrations to changes in their precursors (i.e., volatile organic compounds, nitrogen oxides and sulfur dioxide). OBM-SIA is relatively easy and inexpensive to operate and makes use of data that are increasingly available in China and other developing countries, thereby offering a cost-effective tool for the analysis of SIA-precursor relationships in an urban atmosphere.

In this study, OBM-SIA is applied on a pollution episode in Hong Kong in December 2009. Process analysis in the OBM-SIA reveals that the gas-phase reaction between OH and NO_2 is constantly the major pathway for the production of NO_3^-. While for the SO_4^{2-} production, oxidation of S (IV) by O_3 and O_2 with catalysis of Fe show the most significant contributions. Different from previous knowledge, the pathway of S (IV) oxidation by H_2O_2 only contributes to less than 10% of total sulfate, which is resulted from the suppression of H_2O_2 production by peroxyl radical combination under high NO_x conditions. Formation of NO^{3-} is somewhat more sensitive to the reduction of NO_x and anthropogenic volatile organic compounds (AVOC) concentration. For each percent reduction in NO_x and AVOC, a 0.53 and 0.66% reduction in mass of NO^{3-} formation would result in, respectively. On the other hand, reduction of SO_4^2 formation is most sensitive to reduction of SO_2 and it would become more effective when more SO_2 has been reduced.

P-2-046 Long-range transport of polluted and dust aerosols at Phimai, Thailand by surface measurements and CALIOP data analysis

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Key words
Long-range transport Atmospheric Aerosols Southeast Asia

In Southeast Asia, there was little knowledge of atmospheric aerosols, while biomass burning of crop residue is a dominant source of $PM_{2.5}$ and tropospheric ozone. An intensive field study on chemical and optical properties of atmospheric aerosols was carried out during 2006-2008, at the Observatory of Atmospheric Research (OAR), in Phimai, Thailand. The fine and coarse aerosols were measured, and chemical analysis was made for elemental carbon (EC), organic carbon (OC), water soluble ions, and trace elements. Single scattering albedo was estimated by sky radiometer and from chemical composition. The satellite data of MODIS and CALIOP was used to detect biomass burning and atmospheric aerosols from space. The mass concentration and major chemical components of fine aerosols were much higher in the dry seasons when easterly and/or northeasterly were predominant, than in the wet seasons when southwesterly was predominant. According to the back trajectory analysis, the air masses arrived at Phimai were transported from Indian Ocean in the wet seasons, while they were transported from East Asia and/or South China Sea in the dry seasons. In fine particles, ammonium sulfate, EC and OC were major components. The concentration ratios of EC to non sea-salt sulfate in the early dry season were almost equal to those at Amami-Oshima in southwest Japan, where the anthropogenic aerosols were transported from the urban-industrial area of East Asia. The ratios in the latter dry season when air masses were transported from

South China Sea, however, were much higher than those in the early dry season. It is caused by aerosols emitted from biomass burning of agricultural residues in/around Thailand, which was detected by the MODIS fire maps especially in the latter dry season. The SPRINTARS model also suggests that more than half of non sea-salt sulfate arrived at Phimai through one year could be transported from East Asia, while biomass burning could be the dominant source of EC and OC. Dust aerosols were transported from East China and Indochina, in the early- and the latter dry season, respectively. In contrast, the dust concentration in the wet seasons was usually low, while high concentration of dust was frequently measured. The case study on the episode of high dust aerosols during 17-20 June 2008 was compared with the CALIOP data. It showed that high dust aerosols were located from Saudi Arabia to Somalia, from Pakistan to Arabian Sea, from Bangladesh to the Bay of Bengal, and the maximum vertical height of the dust layer reached up to 7 km. The backward trajectory analysis and the SPRINTARS model also indicate that the high dust aerosols at Phimai in the wet season could be caused by the long range transport of dust aerosols generated in the desert areas of west Asia, in addition to the local dust. Hence, air pollutants in East Asia and desert dusts in west Asia are major sources of atmospheric aerosols in Thailand, in addition to biomass burning in Indochina and the Megacity of Bangkok.

P-2-047 Aerosol composition in a sub-urban site at Cabauw tower, the Netherlands

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Key words
organic aerosol aerosol chemistry

We present aerosol mass spectrometric measurements with an Aerodyne High-Resolution
Time of Flight Aerosol Mass Spectrometer (HR-ToF AMS) from three intensive observation periods at Cabauw Tower, The Netherlands. Aerosol composition was measured in May 2008, March 2009 and November 2011, respectively. Higher average total mass loadings were observed for May (9.72 μg m^{-3}) than for November (4.96 μg m^{-3}) and March (5.62 μg m^{-3}). The mean aerosol composition was observed to be dominated by organics in May and November (average contribution of 40%), whereas it was dominated by nitrate in March (42%). Sources of the observed organics will be discussed based on results from positive matrix factorization analysis together with the impact of NO_x chemistry on observed aerosol mass loadings.

P-2-048 The investigation of nano- and micro-particulate concentrations in an Asian

street market

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Key words
nano- and micro-particulate concentration urban street market

Most people in Asian metropolitan areas live in communities with a mixture of homes, shops, restaurants, street markets, temples, and small factories, where they are closely linked with residents' daily lives. Since this kind of mixed community (as opposed to truly residential communities in western countries) has various sources emitting pollutants in the immediate living environments, high spatial variations in pollutant levels could be found in the same neighborhood, even within short distances. Among the various microenvironments, street market is an environment with clustered stores and street vendors, as well as long-term and regular business activities, which in term could deteriorate local air quality. The objectives of the study were to investigate the variability of nano- and micro-sized particulate concentrations and the factors affecting particulate air quality in an urban street market in Taichung metropolitan area.

In the current study, levels of particles were monitored in the street market area and a tourist's breathing zone. Continuous multi-sized particulate matters (PM), ultrafine particle (UFP) and polycyclic aromatic hydrocarbons levels were measured using a light-scattering monitor/sequential mobility particle sizer (series 1.108 and SMPS+C, Grimm), a condensation particle counter (model 3007, TSI), and a photoelectric aerosol sensor (PAS 2000CE, EcoChem Analytics), respectively. Results showed that mean daytime PM_1, $PM_{2.5}$ and PM_{10} concentrations during the weekdays were 29.58, 53.91 and 69.23 microgram/m^3 in respective, while weekday nighttime levels were the means of 24.08, 44.37 and 57.87 microgram/m^3 in the street market. During the weekend, daytime PM_1, $PM_{2.5}$ and PM_{10} mean concentrations were 43.57, 76.72 and 92.83 microgram m^{-3}, while nighttime concentrations were 52.81, 93.56 and 113.27 microgram m^{-3} for PM_1, $PM_{2.5}$ and PM_{10}, respectively. In general, particulate mass concentrations varied by weekday and weekend, where maximum concentrations during the weekend were higher than those in weekday. Besides, maximum concentrations were about the same magnitude during daytime and nighttime periods, with most of the high levels lying in the particle size of 200-400 nm. For particles with diameter size less than 1 micrometer, particulate number concentrations also differed by weekday and weekend periods. During the weekdays, daytime concentrations were higher than the nighttime levels, whereas the pattern was reversed during the weekend. The main particle size for number concentrations was in the range of 10-70 nm in the weekdays, compared to the size of 10-200 nm during the weekend. Every night after 8pm, peak number concentrations lay in the particle size of 20-100 nm, with weekend levels two times higher than the

weekday levels, which could be attributed to the impact of human activities during the weekend. Finally, using multi-way analysis of variance, wind speed and wind direction were found to affect $PM_{1\text{-}2.5}$ and $PM_{2.5\text{-}10}$ concentrations significantly; $PM_{1\text{-}2.5}$ concentrations were also affected by ambient temperature. There was no interaction between these factors. As regards to submicron particle concentrations, wind speed was also an influential factor, while the interaction between the factors varied with different particle size ranges.

P-2-049 Pollution characteristic of black carbon aerosol at a rural site of South China's coastal area

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Key words
black carbon WRF-HYSPLIT rural site

Because of its remarkable climate and health effect, black carbon aerosols (BC) become the focus of research for the present. Knowledge of the distribution and sources of BC in background areas is essential to understand its impact on radiative forcing and the environment in regional scale. Observation was launched in fall of 2009 for about 40 days at a rural site of South China's coastal area. The average concentration of BC is 2.34 ± 1.33 ug•m^{-3}, at a high level when compared with other background sites. High resolution meteorological data from the Weather Research and Forecast model (WRF) was used to run Hybrid Single Particle Lagrangian Integrated Trajectory 4 (HYSPLIT-4) hourly during the observation period. Trajectories from HYSPLIT-4 were divided into four distinct categories: northeast Coastal, north inLand, Hongkong direction and southeast Sea surface direction. BC concentration is about 1.4 times of the average and there is a good correlation (R2=0.6342) between BC and CO when influenced by air masses from the northeast coastal area while the other three air masses have relative low BC concentrations and poor BC and CO correlations, which indicates that the short-range regional transport from the northeast is the main BC source area of this rural site.

P-2-050 $PM_{2.5}$ mass, chemical composition, and source in relation to PM_1 and PM_{10} in Seoul megacity

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Key words
PM_1 $PM_{2.5}$ PM_{10} aerosol mass OC EC SO_4^{2-} NO_3^- Seoul long range transport

The water soluble ions and carbonaceous compounds of PM_{10}, $PM_{2.5}$, and PM_1 were measured in Seoul, the capital of South Korea (N 37.59°, E127.03°) from March 2007 to June 2008. Ambient air was collected using sharp cut cyclones with 37mm Teflon and Quartz filters. Samples were collected once every six days for 24 hours. Concentrations of organic carbon (OC) and elemental carbons (EC) were determined by the IMPROVE thermal/optical reflectance method.
Recently, the new environmental regulation for $PM_{2.5}$ was enacted in South Korea. In this study, the attribute of $PM_{2.5}$ was investigated with regard to its mass, chemical composition, and source in relation to those of supermicron (PM_{10}) and submicron ($PM_{1.0}$) particles. The mean concentrations of PM_{10}, $PM_{2.5}$, and PM_1 mass were 52.4, 27.1, and 20.2 μg m^{-3}, respectively. In general, the mass and chemical constituents of $PM_{2.5}$ were reasonably correlated with those of PM_1 and PM_{10}. However, there were outliers deviating from the 95% confidence interval of the regression line between PM_{10} and PM_1. Therefore, the data were classified in the PM_1-PM_{10} mass domain into three groups: 1) the regions where PM_1 increased proportionally with PM_{10}, 2) PM_{10} dominant region, and 3) PM_1 dominant region. The groups 1 is characterized by high PM_{10} mass and represents the high PM episodes observed in Seoul including Asian dust and haze. Accordingly, Ca^{2+} and SO_4^{2-} were enhanced in PM_{10} and OC liberated at higher temperature were elevated in PM_1. In groups 2, the concentrations of NH^{4+}, NO^3-, and SO_4^{2-} were noticeably high and there was good correlations between PM_1 mass and SO_4^{2-}, OC2, pyrolyzed OC, and EC1. Particularly, the gaseous concentrations of NO_2, CO, SO_2, and 90th percentile of O_3 were the highest. The samples in group 3 showed the lowest concentrations of mass and chemical compositions under relatively stable atmospheric condition, but the mass fraction of OC and EC in $PM_{2.5}$ was prominently higher than those of other groups. In addition, NO^{3-} was better correlated with OC and EC than SO_4^{2-} was. These results demonstrate the Chinese influence was prevailing for high mass concentrations in Seoul (group 1 & 2), which was well indicated by $PM_{2.5}$ mass measurement. Without distinct enhancement in aerosol mass, the ratio of PM1/PM10 was elevated with higher contribution of carbonaceous species (group 3). This episode was not clearly distinguished by $PM_{2.5}$ mass, during which the local influence was more pronounced. The Positive Matrix Factorization analysis identified coal combustion, industrial and vehicle emissions, soil, sea salts, and secondary formation as aerosol sources.

P-2-051 Chemical weather forecast for south america: impact of urban emissions on local and regional scales

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Key words
CCATT-BRAMS Urban Emissions Ozone footprint

The larger cities in South America have a largest urban population growth ratio compared to Europe or North American continents. The increase of anthropogenic toxic trace gases emissions to the atmosphere and their oxidation products affect the quality of life in the cities and the surrounding areas. This local and regional impacts have been the subjects of observational and modeling studies, however, there are only a few studies about the South American cities regional influence in the atmospheric chemical composition. In context of SAEMC (South American Emissions, Megacities and Climate) program, it was proposed to study the regional impact of urban emissions on air quality and chemical weather forecast over South America using the CCATT-BRAMS (Coupled Chemistry Aerosol and Tracer Transport model to the Brazilian developments on the Regional Atmospheric Modeling System). For this purpose, we worked on the development of an urban vehicle emissions inventory for South America, based on the analysis and aggregation of available inventories for the major cities, with emphasis on its application in regional atmospheric chemistry modeling and the construction of future emission scenarios aiming of establish a relationship between regional air quality and climate change. The use of local emissions databases integrating specific information about urban centers into a broader database is shown to be relevant for a good representation of emissions in chemical weather forecasting models on local and regional scales. The applicability of this methodology in a broad range resolution, including the refinement with the inclusion of vehicular highways emissions and a more realistic distribution of emissions in the Metropolitan Area of São Paulo, provides a significant gain in chemical weather forecasts in finer resolutions. Using this tool and with extensive work on CCATT-BRAMS model, we showed that the larger South American cities has a significant influence on the ozone production in the surrounding area, and has distinct chemical regimes depending on their latitude location. Finally, we worked on the generation of urban emission inventories scenarios for ozone precursors in the period 2011-2030. The scenarios show a significant reduction of carbon monoxide and non-methane volatile organic emissions (around 37%) in 2030 relative to 2010, however, we estimated an increasing of 7% for the NO_X vehicular emission comparing the same period, suggesting that the current national programs for emission control, will not be enough to reduce the NO_X emission and ozone production in the next 19 years.

P-2-052 Comparison of CMAQ-simulated tropospheric NO_2 columns with Ozone

Monitoring Instrument (OMI)-retrieved NO_2 product over Japan

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Key words
Tropospheric NO_2 columns OMI CMAQ model REAS emission inventory NO_x emissions Averaging Kernel

Comparing model-simulated and satellite-observed tropospheric NO_2 column densities was implemented with an intention of evaluating the accuracy of bottom-up NO_x emissions (REAS) from Japan. For simulating tropospheric NO_2 columns, US EPA CMAQ model v4.5.1 was used with a grid-resolution of 30 km × 30 km, using REAS (Regional Emission Inventory in Asia), INTEX-B (Intercontinental Chemical Transport Experiment-Phase B) and CAPSS (Clean Air Policy Support System) emission inventories over Japan, China, and Korea, respectively. Satellite NO_2 products from Ozone Monitoring Instrument (OMI) were also used for this study. Comparison analyses between the two NO_2 columns were made over the entire Japan, and two major metropolitan areas, i.e., Greater Tokyo Area (GTA) and Greater Osaka Area (GOA), for 12 month period in 2006. For the analysis, it was found that CMAQ-simulated NO_2 column densities are well correlated to the OMI-retrieved NO_2 columns for seasonal averages over the entire domain, but are somewhat poorly correlated to the OMI-retrieved NO_2 columns, showing significant overestimation for the winter and spring episodes over the western parts of Japan. Such differences could be caused by trans-boundary error transport from the up-wind regions such as China and Korea. The differences in the two tropospheric NO_2 columns, after applying the averaging kernels (AK) of the OMI NO_2 products are also discussed in this study.

P-2-053 Seasonal and spatial variations of mono- and di-carbonyls in the atmospheric of nine cities in China

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Key words
carbonyls China

Carbonyls are volatile organic compounds in atmosphere, which play an important role in atmospheric photochemistry. They are the main components of photochemical smog and the precursors of free radical, ozone, peroxyacetyl nitrates (PAN) and secondary organic aerosol (SOA). Additions to being directly emitted by anthropogenic and natural sources, these compounds are also formed in situ as the photochemical oxidation products of gas-phase hydrocarbons in the atmosphere. In this study, carbonyl samples were collected by DNPH cartridge, and then analyzed by HPLC-UV based on the USEPA TO-11 method. Seventeen carbonyls in ambient air were quantified in summer (August 2010) and winter (January 2011) in nine cities in China (Beijing [BJ]; Cheungdu [CD]; Guangzhou [GZ]; Qinghai Lake [QH]; Shanghai [SH]; Tibet [TB];Wuhan [WH]; Xiamen [XM]; Yantai [YT]).

Formaldehyde was the most abundant carbonyls found in all cities, ranging from 0.07 ppbv to17.5 ppbv, with an average concentration of 5.07±4.11 ppbv in summer and 2.03±1.53 ppbv in winter. Acetaldehyde (0.01 – 9.46 ppbv, average 1.91±1.54 ppbv in summer and 1.41±1.19 ppbv in winter) was the second most abundant carbonyls. And these two species account for 77% and 72% of the total measured carbonyls in summer and winter, respectively. For dicarbonyls, the average concentration of methylglyoxal (summer: 0.06 ppbv; winter: 0.08 ppbv) was slightly higher than that of glyoxal (summer: 0.04 ppbv; winter: 0.06 ppbv), both accounting for $<$3.0% of total measured carbonyls. Generally, the total measured mono-carbonyls had higher concentrations in summer than in winter, except in TB and XM, indicating photochemical formation of mono-carbonlys play an important role in seasonal variation. However, there is no clear winter vs. summer variation for dicarbonyls.

Large variations of carbonyl concentrations were found among the nine cities; the total measured carbonyls ranged from 0.81 to 32.9 ppbv (average 9.03 ppbv) in summer and ranged from 0.27 to 17.0 ppbv (average 4.79 ppbv) in winter. In summer, the total measured carbonyls in rural and coastal cities (rural: QH and TB; coastal: SH, XM and YT) were lower than in urban cities. While in winter, the concentrations were the most abundant in central and southern urban China (i.e., CD, GZ and WH), and the lowest concentrations were observed in northern rural China (QH). Local emissions and meteorology (temperature, wind speeds and atmospheric conditions) may contribute to the high loadings of total measured carbonyls in urban cities. Because QH is located in a low-population lake region, low anthropogenic emissions may result in low loadings. Strong correlation among carbonyls in winter was observed due to low temperature, weak photochemical reaction, and strong primary emission sources. However, more complex sources exist in summer, resulting in no significant correlations observed.

All the information from this study of carbonyls is useful to thoroughly understand the distribution of carbonyls in the atmosphere, migration and transformation mechanism. Discussion on seasonal variation and spatial distribution are also helpful to control the emission sources and provides theoretical basis for the air quality.

P-2-054 Using heart rate variability to assess the effects of particulate and carbon monoxide exposures in different public transits for healthy young adults

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Key words
heart rate variability particulate pollution public transit

To evaluate the cardiovascular impact of traffic-related pollution and heat exposures on healthy young adults, the research team has collected the primary data of air pollutants and heart rate variability. In the current study, 20 young healthy adults (10 males and 10 females, aged 18-27 years) were recruited from campus dormitory in Taipei metropolitan area. In addition to electrocardiogram (ECG), personal exposures to air pollutants (PM and CO) and weather conditions (temperature and relative humidity (RH)) on campus, bus, and mass rapid transit (MRT) were monitored continuously. Heart rate variability (HRV) parameters including heart rate (HR), the square root of the mean of the sum of the squares of differences between adjacent NN intervals (r-MSSD), the standard deviation of all NN intervals (SDNN), the percentage of successive NN interval differences greater than 50 ms (pNN50), low-frequency power (LF), high-frequency power (HF), total power (TP), and LF/HF ratio were assessed. Since the collected data were repeated measurements and the relationship between time or weather and the outcome variables was nonlinear, we evaluated the association between traffic-related pollution and HRV parameters using appropriate and exact genernalized additive mixed model to adjust for personal and meteorological variables under single- and multi-pollutants models.

Personal exposures to all-sized PM and CO in public transits (including MRT and bus) were significantly higher than those on campus, while temperature and RH were significantly lower in MRT as compared to those on campus). Compared with the HRV parameters measured on campus, the percent changes in r-MSSD, SDNN, pNN50+1, LF, HF, and TP decreased when the subjects were in public transits, nevertheless the percent changes in HR and LF/HF increased.

After adjusting for all locations, 5-min moving averages of $PM_{2.5-10}$ and PM_1 significantly associated with the increase of the percent changes in HR and SDNN. Additionally, 5-min moving averages of $PM_{2.5-10}$ exposures significantly associated with the decrease of the percent change in HF, while associated with the increase of the percent change in LF/HF in significance. The reduction of the percent change in HR was also found to be associated with 5-min CO moving averages significantly. On the other hand, results of multi-pollutant models showed that weather condition was significantly associated with HRV parameters. The smooth function for temperature showed a nonlinear relationship between

temperature and all HRV parameters, while RH contributed significantly to the model fitted to most HRV parameters (except for SDNN and LF/HF).
Decreased trends in r-MSSD, SDNN, pNN50+1, LF, HF, and TP changes were found for increases with $PM_{2.5-10}$ moving averages for 5-60 min exposures.
Increases in 5-60 min $PM_{1-2.5}$ moving averages were associated with the increases in r-MSSD, pNN50+1, and HF, while associated with the decreases in HR and LF/HF. The changes of HRV indices varied with different size fractions of PM, as $PM_{2.5-10}$ had stronger effects on changing HRV as compared to $PM_{2.5}$.
In brief, current analyses have shown that all-sized PM and CO exposures in public transits might have significant changes of HRV indices for healthy young adults.

P-2-055 Source Apportionment of $PM_{2.5}$ in China

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Key words
source apportionment $PM_{2.5}$

With tightening $PM_{2.5}$ (particles with the aerodynamic diameter less than 2.5 micron) standard in the U.S. and the recognition of impacts of fine particulate matter on human health, visibility and climate change, there is increasing number of research focusing on the sources and composition of $PM_{2.5}$. It is critical to identify major contributors of $PM_{2.5}$ for formulating a better control strategy. This study aims to summarize published source apportionment work for $PM_{2.5}$ in China. China started source apportionment work relatively late compared to Europe in 1980s and America in 1970s. $PM_{2.5}$ source apportionment had been conducted in 24 cities mostly located in urban areas of the eastern China. The most studied cities included Beijing, Nanjing, Hong Kong, and Shenzhen. Methods of $PM_{2.5}$ source apportionment were divided into two categories based on the need for source profiles. Category 1 uses source profiles in the chemical mass balance model (CMB) while Category 2 just requires basic knowledge of sources with no demand for source profiles such as positive matrix factorization (PMF). The source apportionment method for $PM_{2.5}$ in China has been further summarized into six types with the first three types in Category 1 and the other three in Category 2: Type 1 (using inorganic species as tracers), Type 2 (organic species), Type 3 (a combination of inorganic and organic species); Type 4 (inorganic species), Type 5 (organic species), Type 6 (inorganic and organic species). Type 4 was the mostly used method in previous source apportionment work in China.

Different source types were identified with different methods, which varied greatly with input species. Results from Category 2 sometimes contain factors mixing several sources together, making it difficult to identify each specific source and its contribution. Sources from Type 2 and 5, which use organic tracers, often better identify sources from combustion processes (e.g., coal combustion, cigarette smoking, vehicular emission, biomass burning, and meat cooking). Based on these previous studies conducted in China, it was found that dust, vehicular emission, coal combustion, and secondary sources were major sources in $PM_{2.5.}$

P-2-056 Air Pollution Exposure to Different Public Transportation Modes in Beijing

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Key words
Transportation Mode pollution exposure Beijing

Beijing is a megacity with dense population and heavy traffic. Motor vehicle emission is a major contributor to air quality deterioration and significant source of toxic substances threatening human health. In this study, high time resolution measurements were conducted for two weeks in December 2011 to assess air pollution exposure to people on road or commuters using different transportation ways. Different public transportation modes were selected including walking, by bus (double deck/single deck) and by subway (underground/over-ground). Online

instruments were deployed to obtain information such as particulate number, $PM_{2.5}$ mass concentration, BC, CO, CO_2, and real time meteorological data. Background levels were simultaneously monitored on the roof-top of a building located inside the campus of Peking University. Results from this study showed particulate number and mass concentrations were highest in underground railway than other transportation ways. Emissions from buses with air-conditioning could enhance particulate matter exposure level by up to three times than those without air-conditioning in wintertime. Commuter exposure to particulate matter using bus or subway was higher than exposure levels by walking mode. Daily variation for exposure to CO and $PM_{2.5}$ by walking mode showed bimodal peaks with a significant peak in afternoon rush hours. Particulate number concentrations for walking mode were lower during rush hours than non-rush hours. This study revealed that level of exposure to particulate matter in Beijing was greatly influenced by transportation modes, the ventilation system, and traffic patterns.

P-2-057 VOC sources apportionment with meteorological analysis in an urban area under industrial emission influences

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Key words
VOCs source apportionment dynamic behavior

Although numerous groups have devoted to VOC studies with various purposes including identifying main sources and determining their contributions to local pollution, few studies have taken into account atmosphere dynamics to investigate the evolution of different source contributions with meteorological conditions. This approach is particularly important in urban zone surrounded by industries. Indeed, the complexity of studies on VOCs source apportionment in an urban area under industrial emission influences, results not only from the variety of industrial activities but also from the different emission modes. The objective of this work is to develop an innovative method which could help the source identification obtained with the use of a receptor model, by analyzing source contribution behaviors with some characteristics of atmosphere dynamics.

To do so, 85 C2-C10 VOCs, including 23 oxygenated VOCs, were collected by TD-GC-FID/MS every 90 minutes over two contrary periods in the industrial city of Dunkerque (North France). The meteorological parameter measurements were simultaneously performed by an ultrasonic anemometer. Among numerous physics parameters, the standard deviation of the fluctuations of wind speed, which characterize longitudinal, transversal and vertical turbulence respectively, were found to be critical to pollutants dispersion. Temperature and solar radiation were also found important. VOCs source apportionment was accomplished by running

PMF model with 1010 samples of 51 major VOCs (including 14 oxygenated VOCs). Ten factors were obtained and two of them mainly consist of oxygenated VOCs. The analyses of contributions with wind directions, vertical turbulence, temperature and solar radiation, led to separate sources at ground level from those in the upper part of surface layer but also to distinguish sources associated with industrial facilities from others and finally to identify secondary sources from local sources. In this complex study area, some factors were hardly attributed only with usual approach (source profiles and CPF analysis); for example, the two factors mainly consisting of oxygenated VOCs. In this case, the combination of usual approach with dynamical analyses of source contribution, led to allocate one factor to non-evaporative sources resulting from industrial facilities at ground level, and to link the other factor with aged air mass including primary and secondary pollutants from remote sources. The study of seasonal variation showed that different meteorological conditions in two contrary seasons have significant impacts on the contribution of remote sources, but slight influences on the factor associated with non-evaporative sources.

This work is, to our knowledge, the first experiment combining VOC source apportionment (via PMF use) with meteorological analysis of the optimized factors and will be useful in short term to optimize prediagnostic models (meteorology forecast module contained) of air pollution, and in long term to elaborate strategies in order to limit certain VOCs emitters according to meteorological conditions.

P-2-058 Impacts Of Rapid Urban Expansion On Regional Climate And Air Quality with WRF/CHEM over Pearl River Delta Region, China

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Key words
urban expansion   Pearl River Delta Region regional climate   air quality

The online Weather Research and Forecasting and Chemistry (WRF/CHEM) model, coupled with urban canopy (UCM) and biogenic-emission models, is used to explore impacts of urban expansion on secondary organic aerosols (SOA) and ozone formation. Two scenarios of urban maps are used in WRF/CHEM to represent early 1990s (pre-urbanization) and current urban distribution in the Pearl River Delta (PRD). Month-long simulation results using the above land-use scenarios for March 2001 show prominent differences in meteorological conditions and surface ozone concentrations. Urbanization increases both the day- and night-time 2-m temperatures by about 0.6°C and 1.4°C, respectively, while it decreases both the day- and night-time 10-m wind speed. Daytime reduction in the

wind speed by about 3.0 m s^{-1} is larger than that for the nighttime (0.5 to 2 m s^{-1}). Urbanization also increases both the day- and night-time boundary layer depths. The daytime increase in the PBL height (> 200 m) is also larger than the nighttime (50-100 m). The meteorological conditions modified by urbanization lead to detectable ozone and SOA concentration changes in the PRD: (1) Urbanization can increase monthly averaged temperatures by about 0.63℃, decrease monthly averaged 10-m wind speeds by 38%, increase monthly averaged boundary-layer depths by 80 m, and decrease monthly averaged water mixing ratio by 0.2 g/kg. (2) Changes in meteorological conditions can result in detectable concentration changes of NO_x, VOC, O_3 and NO_3 radicals. Urbanization decreases surface NO_x and VOC concentrations by a maximum of 4 ppbv and 1.5 ppbv, respectively. Surface O_3 and NO_3 radical concentrations over major cities increase by about 2 - 4 ppbv and 4 - 12 pptv, respectively; areas with increasing O_3 and NO_3 radical concentrations generally coincide with the areas of temperature increase and wind speed reduction where NO_x and VOC decrease. (3) Urbanization can induce 9% increase of SOA in Foshan, Zhongshan and west Guangzhou and 3% decrease in Shenzhen and Dongguan. Over PRD major cities, SOA from Aitken mode reduces by 30% but with more than 70% SOA from accumulate mode. Urbanization has stronger influence on SOA formation from Aitken mode. (4) Over the PRD, 55–65% SOA comes from aromatics precursors. Urbanization has strongest influence on aromatics precursors to produce SOA (14% increase), while there is less influence on alkane precursors. Alkene precursors have negative contribution to SOA formation under urbanization situation.

P-2-059 Patterns of Particle Number Size Distributions in The Spring of Beijing

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Key words
particle number size distributions new particle formation pollution dust

The seasonal patterns of particle number size distributions in Beijing can be quite different under the influences of meteorological factors and anthropogenic activities. The highest particle number concentration was observed in spring. In order to obtain the characteristics of particle number size distributions in spring of Beijing, measurements of particle number size distribution and Meteorological parameters (wind speed, wind direction, and relative humidity (RH)) were conducted at a urban atmospheric environmental monitoring station, at the campus of Peking University. Three cases including heavy pollution, dust and new particle formation (NPF) were analyzed in this study. More than 70% particle number concentration is contributed by the nucleation mode particles. Particle size distribution shows the geometry median diameter in the dominate mode is 6 nm. In heavy polluted day the nucleation , Aitken mode, and accumulation mode are

4100/cm^3, 8700/cm^3, and 6300/cm^3 ,respectively. The fine particle (< 1μm) volume concentration is four times higher than that of the NPF event day. However, during the dust period, an extremely low number concentration of accumulation mode (on average 400/cm^3) was observed. This is because accumulation mode is easily removed by strong wind. The contribution of coarse mode to total volume concentration is 93% during dust period. And then, because of the movement of accumulation mode particles, a NPF event was observed followed by dust. The features of mode distribution of particle number concentration reflect the evolution of particle formation. The shifts of three cases of mode distributions are unique characteristics of aerosol pollution in the spring of Beijing.

P-2-060 Spatial Patterns of $PM_{2.5}$ at Three Sites in the Pearl River Delta, China: Four-Year Observations

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Key words
Chemical speciation of $PM_{2.5}$ Regional Air Pollution

The Pearl River Delta (PRD) is among the most economically fast-developing regions in China. The region has been experiencing increasing levels of particulate matter (PM) pollution. In an effort of establishing long-term trend in chemical characteristics of $PM_{2.5}$ and understanding PM sources important at regional scale, filter-based samples have been collected at three sites in the PRD concurrently in one-in-six-day schedule since August 2007. We here report observation results of $PM_{2.5}$ and its major constituents over four-year period (August 2007-July 2011). The three sites include an urban downtown location in Guangzhou, Nansha, a rural receptor site at the mouth of the Pearl River, and Tsuen Wan, an urban background site in Hong Kong. Typically Guangzhou recorded the highest annual average $PM_{2.5}$ concentration, followed by Nansha and Tsuen Wan. Organic matter (OM) and sulfate are the top two constituents, accounting for ~70% of $PM_{2.5}$ mass. The annual average nitrate contributions were similar at Guangzhou and Nansha (~13%), but lower at Tsuen Wan (~7%). Inter-site correlations of $PM_{2.5}$ and major constituents indicate that Guangzhou strongly influenced ambient $PM_{2.5}$ levels at Nansha, but Guangzhou's influence on Tsuen Wan was much reduced. Sulfate, ammonium, and OM showed strong regional characteristics. To the contrary,

elemental carbon at the three sites had no correlations, suggesting a dominating local origin.

P-2-061 Aircraft-based measurements of size-resolved (0.1-10 μm) particles over the coastal and offshore area of China Yellow Sea

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Key words
aircraft measurement number concentration size distribution regional transport Yellow sea

1. Introduction
In order to investigate regional pollution and transport of atmospheric pollutants (NO_x, O_3, SO_2, CO, VOCs, PM10 and associated chemical components) in East Asia, an intensive aircraft-based observation campaign was carried out over the coastal and offshore area of China Yellow Sea in April, 2011. All flight missions were performed by a light aerotransport (Yun-12) in the daytime for nine days and each flight lasted about 3-4 hours at a cruising speed of approximately 180 km/hr. Flight routes were straight line and within the area with longitudes from 119 to 123 and latitudes from 35 to 38 at three altitudes of 800, 1600 and 2400 m above sea level. The base airport was in Qingdao, China. In this report, we mainly summarized number concentrations of size-resolved particles measured from 11 to 15 in April and focused on their spatial distribution characteristics.
2. Methods
A Passive Cavity Aerosol Spectrometer Probe (PCASP, model X2, DMT Corp., USA) based on the principle of light scattering was used to measure number concentrations of particles in the 0.10-10 μm range with a sampling frequency of 1 sec. A cyclone restricts inlet particles to the size range that can be measured. In order to avoid the influence of flight speed on sampling, an isokinetic sampling system was designed to minimize aerosol aerodynamic losses.
3. Results and discussion
During aircraft measurement period, total number concentrations and effective diameters of particles varied from 45 to 88031 cm^{-3} and 0.11 to 0.54 μm, respectively. In general, the particulate concentrations decreased with altitudes,

while effective diameters increased with altitudes. Concentrations fluctuated with larger deviations at altitudes of 800 and 1600 m, while those were relatively uniform at the altitude of 2400 m. The above characteristic distribution was mainly because regions at altitudes of 800 and 1200 m were in the mixing layer and were affected by local emission source distribution and meteorological conditions, whereas region at the altitude of 2400 m was in the lowest free troposphere and were affected by regional transport of pollutants. As to horizontal distributions, concentrations over the coastal area were higher than those observed over the offshore area, which indicated that the coastal area were affected by land emission sources much more than the offshore area. It was noteworthy that concentrations observed in April 11 were the highest during the measurement period. Those varied from 159 to 88031 cm^{-3} with the average of 34261 cm^{-3} which was nearly 2 times higher than those in other days. Especially, concentrations at the altitude of 2100 m were higher significantly than those at the altitude of 1500 m over the offshore area. With the help of the meteorological analysis and 24-h back trajectory analysis, it indicated that the polluted air mass over Asia land transported from northwest direction at the altitude of 2100 m. Number size distributions were characterized by typical exponential pattern. The highest concentration peak is at 0.12 μm and the lowest is at 0.5 μm.

P-2-062 Investigation of primary and secondary origins of increased roadside NO_2 concentrations in Hong Kong

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Key words
primary emissions secondary formation NO_2 VOC

Successful reduction of roadside and ambient NO_x (nitrogen oxides) levels have been recorded in Hong Kong due to the implementation of various emissions control measures in the last decade. However, no such decrease has been detected for nitrogen dioxide (NO_2) and there is even a rising trend over the last few years. Similar observations from long term monitoring data have also been found in other urban areas such as London, Amsterdam etc. As NO_2 is actively involved in the complex photochemical reactions with the participation of nitric oxide (NO), ozone(O_3) and volatile organic compounds (VOCs), a better understanding of the contributory factors affecting ambient NO_2 concentrations are of great interests to policy makers. The present study employs a novel approach of on-road plume chasing and analysis system (OPCAS) integrated with a real time ion molecule reaction –mass spectrometer (IMR-MS) to investigate the fresh emissions of NO/NO_2/VOCs and an array of particle instruments for particulate matter (PM)

measurements from different types of vehicles. The OPCAS platform actively selects those on-road vehicles and quickly determines the emission levels using fuel carbon balance method under their real world driving and maintenance conditions, thus avoiding the possible discrepancy resulted from inspection/maintenance program. The study results show that franchised buses and medium/heavy duty diesel trucks are the biggest contributor to the primary NO_x emissions and they have different ratios of NO_2/NO_x and contrast NO_x/BC emission profiles indicating the impact of different emission control policy and retrofitting after treatment devices on the primary emission levels. Mini-buses fuelled by liquid petroleum gas (LPG), however, emit significantly higher butane and other VOCs with high ozone formation potential which may have contributed to the titration of NO forming secondary NO_2. Based on the findings, a Community Multi-scale Air Quality (CMAQ) model is further implemented to simulate the impact of the previously underestimated VOC levels on the roadside NO_2 levels.

P-2-063 OH reactivity measurements as an aid for deriving instantaneous ozone production regimes and rates

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Key words
OH Reactivity ozone air quality

In this study air masses are characterized in terms of their total OH reactivity which is a robust measure of the “reactive air pollutant loading”. The measurements were performed during the DOMINO campaign (Diel Oxidant Mechanisms In relation to Nitrogen Oxides) held from 21.11.2008 to 08.12.2008 at the Atmospheric Sounding Station - El Arenosillo (37.1 N – 6.7 W, 40 m asl). The site was frequently impacted by marine air masses (arriving at the site from the southerly sector) and air masses from the cities of Huelva (located N.W of the site), Seville and Madrid (located N.N.E. of the site). OH reactivity values showed strong wind sector dependence. North eastern “continental” air masses were characterized by the highest OH reactivities (average: 31.4 ± 4.5 /s; range of average diel values: 21.3 - 40.5 /s), followed by north western “industrial” air masses (average: 13.8 ± 4.4 /s; range of average diel values: 7 – 23.4 /s) and marine air masses (average: 6.3 ± 6.6 /s; range of average diel values: below detection limit –21.7 /s), respectively. The average OH reactivity for the entire campaign period was ~18 /s and no pronounced variation was discernible in the diel profiles with the exception of relatively high values from 09:00 to 11:00 U.T.C. on occasions when air masses arrived from the north western and southern wind sectors. The measured OH reactivity was used to constrain both diel instantaneous ozone production potential rates and regimes. Gross ozone production rates at the site were generally limited by the availability of NO_x with peak values of around 20 ppbV O_3 /hr. Using the OH reactivity based approach, derived ozone production rates indicate that if NO_x would no longer be the limiting factor in air masses arriving from the continental north eastern sector, peak ozone production rates could double. We suggest that the new combined approach of in-situ fast measurements of OH reactivity, nitrogen oxides and peroxy radicals for constraining instantaneous ozone production rates, could significantly improve analyses of upwind point sources and their impact on regional ozone levels.

P-2-064 Study of CO and PM_{10} concentrations in Southern Chile (Temuco city 38° 45' S, 72° 38' O), using the Weather Research and Forecasting (WRF) modeling system

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Key words
CO and PM_{10} WRF-Chem METROPOLITAN AREA AIR-POLLUTION

Temuco city, located about 700 km south of Santiago, Chile (38° 45' S, 72° 38' O), suffers from severe particulate pollution events due to residential wood burning,

especially during the period from April to September (fall and winter in SH) each year. In fact, almost 70% of the population uses firewood for cooking or heating in winter, and it is estimated that 92% of PM_{10} winter emissions originates from residential wood combustion. For this reason, it was declared a nonattainment area for PM_{10} in 2005. In 2008, the records from the monitoring stations show high levels of PM_{10} concentration, with a 24-hr from 299 ug/m^3 reaching peak of 1,053 ug /m^3 (1^{-h}) despite the emissions reduction program recently implemented.
The primary goal of this study was to establish the relationship between air pollution and synoptic conditions using the Weather Research and Forecasting (WRF) with the chemistry model. The species included in this analysis were coarse particulate matter (PM_{10}) and carbon monoxide (CO).
The forecasting system is based on accurately simulating carbon monoxide (CO) as a PM10 surrogate, since during episodes and within the city there is a high correlation (over 0.87) among these pollutants with a lag of 1 hour, during 2008. The simulations were made with a 1 km resolution and four typical synoptic conditions for the period of study. The results were compared with concentrations and meteorological variables measurements in the monitoring stations of Las Encinas and Maquehue airport.
The results show that the concentrations of PM_{10} and CO are regulated in Temuco city by the meteorological condition, because these determine the emissions amount in this city.

P-2-065 Which emission sector is winning the mitigation competition?

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Key words
aerosols emission sectors

Attention has been drawn to black carbon aerosols, as a target for short-term mitigation of climate warming. Regulating soot emissions could, as a short-term action, potentially buy time by slowing global warming until regulations for long-lived greenhouse gases are set in place. The scientific community debates the impacts of such mitigation measures, and mitigation modelling studies show incoherent answers. One of the main reasons for the disagreement are semi-direct aerosol effects, that are neglected in some studies and included and dominating the overall results in others. In this study we apply the GISS/MATRIX model, a global climate model including detailed aerosol microphysics, to understand the single contributions of aerosol forcings and feedbacks. The study goes beyond black carbon mitigation by investigating the whole suite of aerosol sources and sectors of the CMIP5 emission data sets.
Our study finds a regionally divers picture. For example aerosol-cloud effects over the United States lead to reduced cloudiness through semi-direct effects and increased cloudiness by the indirect effect and the reversed phenomena is simulated over Europe. This response will be explained by the chemical

composition of the emission mix in the different regions and its impact on black carbon coatings. The most promising emission mitigation sectors differ greatly between geographically regions and even among industrialized countries.

P-2-066 Air quality in asia: status and trends, 2011 update

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Key words
air pollution PM2.5 monitoring

Majority of people living in Asian cities may be exposed to unhealthy levels of air pollution. Worldwide, it is estimated that around 3.1 million die prematurely each year due to air pollution with majority of victims in low and middle income countries (WHO, 2009). Effective management of air pollution first calls for an understanding and quantification of the problem. However, while information and knowledge on air quality management are increasingly generated in Asia, they are fragmented in different documents and websites. To ensure that decision-makers have better access to knowledge on clean air management that can assist in policy development, CAI-Asia has been releasing publications providing a comprehensive and comparative assessment of the status and challenges of air quality management in Asia.

For the 2011 update, we compared annual average ambient PM10, SO_2 and NO_2 concentrations for 2009 in about 270 Asian cities relative to international guidelines and assessed trends using data from 1993 to 2009. We also explored the relationship between city population size and air pollution levels using a correlation matrix. Results showed that while some improvements in air quality have been achieved, levels of PM10 and SO_2 continue to exceed World Health Organization (WHO) air quality guidelines (AQG). In 2009, only 2.2% of Asian cities surveyed (total: 271) had annual PM10 concentrations within WHO AQG of 20 $\mu g/m^3$. 52.8% of surveyed cities had PM10 levels above even the first interim target of WHO (70 $\mu g/m^3$). A weak positive correlation was found between air pollution levels and city population size.

Through this assessment, we also gained insight on limitations of air quality assessment in Asia. There is limited data to assess PM2.5 and ozone in several Asian cities. There is also a question regarding representativeness of data as several cities provide annual levels from stations with data for less than 50% days in a year. This highlights a need for an assessment of reliability of reported data and help Asian cities improve the quality of their air quality monitoring systems and infrastructure for effective AQM.

P-2-067 Characterization of submicron aerosols in a rural site in Northern China

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Key words
organic aerosol HR-ToF-AMS positive matrix factorization

Introduction

Northern China, especially Shandong Province and surrounding areas, has experienced fast development during the past two decades. With the rapid economic growth, high level of air pollutants becomes a more and more severe issue. In order to characterize in depth the pollution mechanisms and atmospheric chemical processes in this region, a campaign was carried out in spring 2011 near Lake Hongze. An Aerodyne High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS) was used in the campaign to measure the chemical composition and size distribution of non-refractory submicron particulate matter (NR-PM1)

Methods

Hongzehu supersite is located to the northwest of Lake Hongze in Jiangsu province, China, in the midst of North China plain, where air masses can have a long transportation process without being blocked by high mountains. The AMS deployed at the site operated under "V" and "W" mode alternately, to obtain mass concentrations, particle size distributions and high resolution mass spectra. The instrument was calibrated for inlet flow, ionization efficiency and particle sizing at the beginning and the end of the campaign following the standard protocols. Dataset measured by the AMS was calculated using the standard AMS data analysis software packages (SQUIRREL version 1.51H and PIKA version 1.10H) compiled and executed on Igor Pro 6.22A. Positive matrix factorization (PMF) analysis was conducted on the high-resolution mass spectra (HR-MS) (m/z 12-180) to identify major organic components.

Results and discussion

The mean measured PM1 mass concentration was 46.0 +/- 14.2 mu g m^{-3} during the campaign, with nitrate and organic aerosol (OA) being the two dominant species, accounting for 27.1% and 26.8%, respectively, followed by sulfate (22.1%), ammonium (15.4%), BC (6.0%, measured by an aethalometer) and chloride (2.6%). The average size distributions of the species were dominated by an accumulation mode peaking at around 550 nm. Calculations based on high-resolution organic mass spectra showed that on average, C, H, O and N contributed 53.3%, 6.0%, 39.5%, and 1.2% to the total organic mass, respectively. The average ratio of organic mass over organic carbon mass (OM/OC) was 1.88 +/- 0.11. Two components of OA were identified by PMF analysis, including SV-OOA and LV-OOA, which on average accounted for 40.0% and 60.0% of the total organic mass, respectively.

Conclusion

The relatively high organic matter to organic carbon ratio observed in this campaign (1.88 +/- 0.11) reflects the air masses reaching the site were rather aged, showing the nature of regional pollution. PMF result showed no primary OA, and in OOA, LV-OOA contributed 60.0% for its total mass, indicating strong secondary organic aerosol formation during the regional transportation processes.

P-2-068 EDXRF-FP method for rapid and simple multi-elements analysis in aerosol samples collected on QFF in five Asian megacities

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Key words

X-ray fluorescence spectrometry aerosol analysis quartz fiber filter trace metals

Introduction

Many cities around the world are adversely affected by air pollutants such as aerosol and hazardous chemical species. In particular, many kinds of metals and metallic compounds in aerosol are harmful and carcinogenic to humans. Variety of methods has been used for analyzing multi-elements in aerosols. This study shows a reliable, non-destructive, rapid and simple method using EDXRF for analyzing multi-elements in aerosols collected on QFF.

Experimental

Systematic experiments for elucidating the applicability of QFF for EDXRF have been carried out by comparing the analytical results obtained by EDXRF-fundamental parameter (FP) method to those obtained by ICP-MS and ICP-AES. Multi-elements analysis can be achieved by using EDXRF-FP quantification method as fast as 900 s (15min) per sample. Filter samples were analyzed by EDXRF without any pretreatment using the EDXL300 spectrometer manufactured by Rigaku Corp., Japan. The quantification of each element in aerosol samples was performed using a FP method. EDXL300 has a powerful FP algorithm called Rigaku Profile Fitting - Spectra Quant X (RPF-SQX). We selected aerosols samples collected on QFF in five Asian cities as the target samples. Beijing: The sampling site was Tsinghua University, located 15 km in a northwesterly direction from the center of Beijing city, China. We collected one sample each in summer and winter because air pollution level in summer and winter is quite different in Beijing. Hanoi: The sampling site was a private house, located 4km in a southwesterly direction from the center of Hanoi city, Vietnam. Kolkata: The sampling site was a private house in a center of Kolkata city, India. Tokyo: The sampling site was the University of Tokyo, located in a center of Tokyo metropolitan area, Japan. Nagoya: The sampling site was Nagoya University, located near a residential area in Nagoya city.

Results and discussion

EDXRF results for each element agreed well with the ICP-MS and ICP-AES results. We didn’t find an upper-limit of the concentration in terms of the agreement in the comparison results. This means that we don’t have to concern an overload of the particulate matter on the filter.

Conclusions

EDXRF-FP method is suitable for elucidating daily variations of multi-elements in aerosols. This study offers researchers much easy way to achieve a simultaneous measurement for carbonaceous and inorganic components in aerosols collected using QFF, by using EDXRF-FP method followed by further analytical procedures for the other components.

P-2-069 Evaluating the degree of oxygenation of organic aerosols during foggy days and hazy days in Hong Kong using High-resolution Time-of-Flight Aerosol Mass Spectrometry (HR-ToF-AMS)

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Key words
Organic Aerosol Oxygenation Aqueous-phase AMS

The role of aqueous-phase chemistry in the formation of secondary organic aerosols (SOA) is still poorly constrained because it is influenced by a number of factors. There are numerous laboratory studies concerning the effects of aqueous-phase chemistry, including that in cloud, fog and wet aerosol particles, on SOA formation, but only a few from field studies at limited locations. To understand whether or not these aqueous-phase processes affect the production of SOA, and eventually the properties of aerosol particles, more observation studies regarding the impacts of aqueous-phase chemistry are needed. Here we present observation results of the effects of fine particle liquid water content (LWC_fp) and other parameters on the degree of oxygenation of organic aerosols (OA) based on high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) measurements made at a coastal site in Hong Kong during a transient period from springtime to summertime in Apr.-May, 2011. Data from two foggy weeks with E-AIM predicted LWC_fp up to 100 μg/m^3 and one hazy week with PM$_1$ concentration (as measured by the HR-ToF-AMS) up to 60 μg/m^3 were chosen for analysis. The degree of oxygenation of OA, as indicated by several parameters including f44 (fraction of m/z 44 in total organic mass spectra) O:C and H:C ratios and the carbon oxidation state (OSc) were evaluated against odd oxygen (Ox) concentration, relative humidity (RH), LWC_fp, ionic strength, ratio of measured to predicted NH$_4$ (NH$_4$_m/p), and in-situ pH (pHis). Results showed that for the week of hazy period, the high concentration of OA (on average 8 μg/m^3) and the high degree of oxygenation were mainly due to gas-phase oxidation. During the foggy days with low photochemical activities, the degree of oxygenation of OA was as high as that in hazy days, consistent with previous field studies and further suggested the important role of aqueous-phase chemistry in wet particles in SOA formation. However, the evolution of OA behaved quite differently for the two weeks of foggy periods. The first foggy week in late April and early May had more LWC_fp and lower particle acidity (higher pHis), and the OA had more semi-volatile oxygenated organic aerosols (SV-OOA) as resolved by positive matrix factorization (PMF). The second foggy week in mid-May had lower LWC_fp and produced more low-volatility oxygenated organic aerosols (LV-OOA). This comparison suggests that even during similarly high RH and high liquid water content in fine particles, the aqueous-phase chemistry can be different due to compositional difference in the particle phase. Finally, results suggest that during this spring to summer transition in Hong Kong, OA concentration is highest in hazy days, followed by foggy days, and then by non-haze/non-fog days; in terms of degree oxygenation, OA in both hazy days and foggy days are more oxidized than non-haze/non-fog days.

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P-2-070 Aerosol optical properties observed by combined Raman-elasticbackscatter lidar in winter 2009 in Pearl River Delta,South China

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Key words
Raman lidar sunphotometer aerosol haze

Abstract: We present combined Raman and elastic backscatter lidar observations which were carried out in Zhongshan, PRD (Pearl River Delta) China during two periods in 2009: one haze pollution period and one moderate pollution period. The observed mean lidar ratio (extinction–to-backscatter ratio) from the Raman lidar was 60 sr for 355 nm, a bit higher compared to previous observations in this area. The largest lidar ratios, of the order of 80 sr are found in the upper boundary layer in 2-3 km during the haze period. Estimated values of the Angstrom and Depolarization ratio (DPR) are also used to help identify aerosol types. The Angstrom exponent, calculated from 355-532 wavelengths pairs exhibited high values around 1.82, indicating the presence of rather small particles. Four-day back trajectories in the haze period indicated that air masses in the lower layer were advected from the southeast coast of China, where incomplete combustion of carbonaceous fuels is frequently found in Shanghai during the heating period in winter. In the moderate pollution period, the air mass passed through western China indicating the combination of some pollution from the Qinghai-Tibet Plateau in case of strong convection and smoke from adjacent fire burning spots in PRD region.

P-2-071 Source apportionment studies on particulate matter in Beijing/China

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Key words
Particulate matter source apportionment modelling measurements

More than 15 million people in the greater area of Beijing are still suffering from severe air pollution levels caused by sources within the city itself but also from external impacts like severe dust storms and long range advection from the southern and central part of China.

Within this context particulate matter (PM) is the major air pollutant in the greater area of Beijing (Garland et al., 2009). PM did not serve only as lead substance for air quality levels and therefore for adverse health impact effects but also for a strong influence on the climate system by changing e.g. the radiative balance. Investigations on emission reductions during the Olympic Summer Games in 2008 have caused a strong reduction on coarser particles (PM_{10}) but not on smaller particles ($PM_{2.5}$).

In order to discriminate the composition of the particulate matter levels, the different behavior of coarser and smaller particles investigations on source attribution, particle characteristics and external impacts on the PM levels of the city of Beijing by measurements and modeling are performed:

-Examples of long term measurements of $PM_{2.5}$ filter sampling in 2010/2011 with the objectives of detailed chemical (source attribution, carbon fraction, organic speciation and inorganic composition) and isotopic analyses as well as toxicological assessment in cooperation with several institutions (Karlsruhe Institute of Technology (IfGG/IMG), Helmholtz Zentrum München (HMGU), University Rostock (UR), Chinese University of Mining and Technology Beijing, CUMTB) will be discussed.

- The impact of dust storm events on the overall pollution level of particulate matter in the greater area of Beijing is being assessed by the online coupled comprehensive model system COSMO-ART. First results of the dust storm modeling in northern China (2011, April 30th) demonstrates very well the general behavior of the meteorological parameters temperature and humidity as well as a good agreement between modeled and measured dust storm concentration variability at Beijing in the course of time.

The results show the importance of intertwine investigations of measurements and modeling, the analysis of local air pollution levels as well as the impact and analysis of advective processes in the greater region of Beijing. Comprehensive investigations on particulate matter are a prerequisite for the knowledge of the source strengths and source attribution to the overall air pollution level. Only this knowledge can help to formulate and to introduce specific reduction measures to reduce coarser as well as finer particulates.

P-2-072 Aqueous Reactive Uptake of Dicarbonyls as a Source of Secondary Organic Aerosols in the Pearl River Delta Region

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Key words
SOA CMAQ glyoxal methylglyoxal PRD

Organic aerosols (OA) are of essentially importance of air quality, climate forcing and public health. We used the regional scale chemical transport model CMAQ to simulate OA in the Pearl River Delta (PRD) region during 24 October and 6 November 2009. We modified the model chemical mechanism to include detailed gas phase chemistry of glyoxal and methylglyoxal, as well as reactive uptake of the dicarbonyls by aqueous aerosols and cloud droplets to form secondary organic aerosol (SOA). Improved description of gas phase dicarbonyl chemistry led to increases in the monthly mean glyoxal and methylglyoxal concentration by 16% and 13%, respectively, mostly due to increased production from glycolaldehyde and hydroxyacetone. We compared model results with measurements from the Aerodyne high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) in urban Shenzhen. Adding the dicarbonyl SOA increased the simulated monthly mean SOA from 2.20 μg m^{-3} to 5.13 μg m^{-3}, in better agreement with the observations (8.8 μg m^{-3}). Moreover, the simulated SOA with dicarbonyl contribution reproduced the observed strong correlation between SOA and sulfate ($r = 0.71$), as well as the observed diurnal patterns. Our analyses show that the aqueous reactive uptake of dicarbonyls can be an important source of SOA in the PRD region.

P-2-073 Influence of multi-air pollutants on the temporal variation of surface ozone in an urban site located in Deccan Plateau region

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Key words
air pollutants photochemical oxidation back trajectory modeling

The current generation dispossesses a healthy air-quality environment in urban areas because of the increase in concentration of air pollutants such as –SO_x, NO_x, CO, and O_3. The decrease in air-quality is a consequence of accumulation, dispersion and transformation of air-pollutants. Emissions from the automobile combustion, anthropogenic activities and rapid industrialization led to the raise in the air-pollutants. One of the main problems in urban areas is the pollution caused by photochemical oxidants such as O_3 and the NO_x. Anthropogenic sources are responsible for more than 95% of the O_3 in the lower atmosphere. Evidence has been found on the fact that photochemical smog may cause detrimental effects on

vegetation, human health and various materials at elevated concentrations.

To monitor these atmospheric trace gases an air monitoring station is established at Hyderabad, Andhra Pradesh, India (urban area). Trace gas analyzers were installed at Tata Institute of Fundamental Research–National Balloon Facility (TIFR-NBF) (17.47°N and 78.58°E). The vehicular traffic in Hyderabad is a major contributor to the urban air pollution load during dawn to dusk time. Inventories surrounding the observation site further added to the pollution load. Air samples were continuously analyzed for surface O_3, NO_x, CO and SO_2 (in ppbv). Along with the trace gases monitoring, meteorological variables were also continuously recorded. Diurnal/ monthly/ seasonal variations of the O_3 depicted the maximum mean during summer (57.5 ± 5.2 ppb) particularly in late noon hours (14:00 – 16:00). Photochemical oxidation of NO_2, CO and VOCs could the cause for build up during day-time surface O_3 levels.

Contrary to O_3 profile, NO_x, CO and SO_2 showed double peak pattern, relating to the busy traffic hours. Apart from the variation of these concentrations, remarked difference was observed between weekdays and weekends O_3 levels i.e. weekend effect of ozone. Daytime and nighttime atmospheric chemistry was established to understand the variations. Back trajectories were simulated to trace out the origin and path of air parcel transport. It was observed that in summer local/ regional sources contributed for surface level; in monsoon, wet deposition due to monsoon rain hailing from Arabian sea; and, in winter, indo-gangetic plain (IGP). Black carbon measurements were also made; the monthly diurnal pattern showed two peaks relating to traffic, domestic biomass burning and industrial sources.

Statistical analysis was performed using data sets of observed ambient air pollutants and meteorological parameters, which are the predictors for O_3 forecast. The calculated Pearson correlation values analyzed the strength between O_3 and different variables. Prediction of surface O_3 levels was made using two techniques - linear and non-linear modeling. Linear model was developed using Multiple Linear Regression (MLR), while non-linear model was developed using Artificial Neural Network (ANN). The non-linear model resulted in higher regression coefficient than the linear model signifying that the O_3 formation is non-linear phenomena.

In addition to the ground based observations, satellite inter-comparisons were also made during festive occasions. The ground data was corroborated with the satellite measurements, signifying source/sinks and transport of pollutants. In conclusion, temporal and spatial profile inter-comparison exercise made was useful to derive the dynamic behavior of measured air pollutants and checked with developed statistical models. These modeling aspects could be useful to understand air pollution aspects and change in climatic pattern. Further, these models would be helpful in the development of an environmental policy, in particular to green house gas (GHG) emission on a global scale.

P-2-074 Import of East Asian SO_2 into the Lower Stratosphere by a Warm Conveyor Belt event: First Measurements with the Research Aircraft HALO

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Key words
sulfur dioxide long-range transport

Atmospheric SO_2 is an important precursor of sulfate aerosol particles in the lower stratosphere (LS). The latter have an impact on the radiation field and ozone chemistry. Important sources of SO_2 to the LS are import from ground-level emissions, volcanism, and air traffic. An important SO_2 source region is East Asia with rapidly growing SO_2 emissions. An efficient vertical transport mechanism is warm conveyor belts (WCB) associated with mid-latitude cyclones. WCBs are very frequent in East Asia during winter. We present a case study of anthropogenic SO_2 pollution transport into the LS from East Asian source regions by a WCB event in November 2010. The pollution layer was detected over Central Europe using the new German research aircraft HALO. The measurements were performed with a chemical ionization ion trap mass spectrometer and a high spectral resolution Lidar. The pollution layer contained enhanced SO_2, HNO_3 and water vapor concentrations and caused an increased Lidar backscatter signal. Meteorological analysis and air mass transport and dispersion model simulations reveal that the detected pollutants were released from ground-based sources in East-China, South-Korea, and Japan. Our HALO measurements were performed 5 days after the WCB air mass uplift event, when significant parts of the Northern Hemisphere were already covered by the pollution plume. Accompanying trajectory chemistry and aerosol box model simulations indicate that H_2SO_4/H_2O aerosol droplets were generated in the SO_2-rich plume and grew to sizes large enough to explain the observed increased Lidar backscatter signal. Implications of this SO_2 transport pathway into the lower stratosphere will be discussed.

P-2-075 Air quality forecasting for Belgian cities

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Key words
forecast high resolution data assimilation

Notwithstanding improvements in the air quality over the last decade, European member states are still facing wide-spread exceedances of EU regulations particularly with respect to particulate matter (PM10), nitrogen dioxide (NO_2) and ozone (O_3) concentration levels. According to the European Environment Agency (EEA) on average 20% of EU-27 urban population is living in areas where PM10, NO_2 and/or O_3 concentration levels are higher than the air quality directive limit values. The situation is worst in the Benelux area, Northern Italy, and in new member states such as Poland and Hungary.

In its strategy to protect the European citizens' health the European Commission Air Quality Directive requires member states to assess the impact of air pollution on public health. Whilst previous Directives have based assessment and reporting mainly on measurement data, the new air quality directive places more emphasis on, and encourages, the use of chemical transport models (CTM). Compared to the observation data these models allow assessing air quality at a much higher spatial resolution and can be used to forecast air quality.

In our contribution we will present the results of an operational forecasting system based on the AURORA CTM that has been developed over the last years in the context of the European LIFE project ATMOSYS and the European Commission atmospheric GMES service, PASODOBLE (http://www.myair-eu.org). This system produces daily forecasts for the concentrations of O_3, NO_2 and PM10 for the major Belgian cities Brussels, Ghent, Antwerp, Liege and Charleroi at a 1 km resolution. Aspects we will address in our presentation are practical implementation using OGC standards and web services, the need for high resolution modeling when addressing air quality in cities and improvements to the model forecast results using Kalman filter-based bias correction.

P-2-076 Analysis of Carbon Monoxide Distribution in Megacities over East Asia from Satellite and Aircraft Observations

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Key words
Carbon Monoxide Megacities MOPITT MOZAIC

Carbon monoxide (CO) is a major pollutant and a precursor of ozone. The CO distributions in megacities have illustrated the combined natural and anthropogenic

influences. Global carbon monoxide has been monitored by the Measurements Of Pollution In The Troposphere (MOPITT) instrument on board NASA Terra satellite since 2000. In the meantime, airborne in-situ measurements of carbon monoxide from the Measurements of Ozone and Water Vapor by in-Service Airbus Aircraft (MOZAIC) program have been made. Combining the MOPITT (versions 4 and 5) and MOARIC data, we analyzed variations of carbon monoxide in megacities over East Asia in 2005, including Beijing, Shanghai, Hong Kong, and Tokyo. We found that at the bias between MOPITT and MOZAIC is generally the smallest at around 500 hPa, while MOPITT data usually show higher CO mixing ratios than MOZAIC data below 500 hPa except for near the surface where MOPITT cannot capture high CO concentrations observed by MOZAIC. The bias varies with cities, suggesting geographic influences from perspectives of local CO sources, boundary–layer meteorology conditions, and weather patterns. Seasonal variations and spatial distributions of carbon monoxide over East Asia will be discussed from an integral analysis of the both datasets.

P-2-077 Atmospheric chemistry research in China – a synthesis of results from several field studies

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Key words
ozone chemistry aerosol and cloud water

This paper gives a summary of the key results from several field studies which were conducted by our group in Hong Kong, other megacities, rural and remote areas of China. The results include trends, characteristics, and chemical and dynamic processes of ozone, ozone precursors, and aerosol and cloud water. A focus will be given to some unique chemical processes in China's atmosphere.

P-2-078 Observations of the optical properties of urban aerosols in Taipei, Taiwan

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Key words
urban aerosols optical parameters radiative properties

Tropospheric aerosols have been recognized as one of the critical components in the radiation balance of the earth-atmosphere system. Yet, to date, there is a substantial knowledge gap in the climate forcing mechanisms of aerosols. Consequently, the aerosol field was identified as the major source of the uncertainties in the simulation of climate. The uncertainties of the aerosol radiative forcing are due mostly to the variations in the amount and the physical/chemical

characteristics of aerosols on different temporal and spatial scales, particularly within the boundary layer. Besides, the uncertainties are also attributed to the deficiency in the fundamental understandings of the physico-chemical behavior of aerosols.

This study performed multi-wavelength measurements of the light scattering and absorption coefficients of aerosols at an aerosol observatory (TARO, 25.0°N, 121.5°E) in Taipei, Taiwan in December 2007. Spectrally resolved single scattering albedo (SSA) was retrieved from the data of light scattering/absorption coefficients.

The campaign averages of light scattering coefficient range from 149.9 ± 101.4 Mm^{-1} for 450 nm to 113.6 ± 76.4 Mm^{-1} for 550 nm and, in turn, 68.3 ± 46.0 Mm-1 for 700 nm. The averages of light absorption coefficient range from 26.1 ± 16.3 Mm-1 for 370 nm to 15.5 ± 8.9 Mm-1 for 520 nm and 7.4 ± 4.1 Mm^{-1} for 950 nm. Both show significant wavelength dependency, which are characterized by the Angstrom exponents As and Aa of 1.74 and 1.29, respectively. In addition to light scattering/absorption coefficients, it was found that the values of SSA exhibited also obvious wavelength dependency, which range from 0.878 ± 0.075 for 370 nm to 0.870 ± 0.070 for 520 nm and 0.830 ± 0.075 for 950 nm and was accordingly characterized with averaged Angstrom exponent of 0.06. The elevated light absorption capability and low SSA of urban aerosols observed in this study agree with those reported recently for several megacities worldwide, suggesting that the aerosols could have strong warming effects in urban areas. Given that the net aerosol radiative forcing is known being negative on global and regional scales, the sign reverse on local/urban scales necessitates further investigation upon the influences of aerosols in urban meteorology and regional climate changes.

P-2-079 Impact of HONO on global atmospheric chemistry calculated with an empirical parameterization in the EMAC model

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Key words
HONO Photochemistry EMAC model Global impact

The photolysis of HONO is important for the atmospheric HO_x ($OH+HO_2$) radical budget and ozone formation, especially in polluted air. Nevertheless, owing to the incomplete knowledge of HONO sources, realistic HONO mechanisms have not yet been implemented in global models. We investigated measurement data sets from 15 field measurement campaigns conducted in different countries worldwide. It appears that the $HONO/NO_x$ ratio is a good proxy predictor for HONO mixing

ratios under different atmospheric conditions. From the robust relationship between HONO and NO_x, a representative mean HONO/NO_x ratio of 0.02 has been derived. Using a global chemistry-climate model and employing this HONO/NO_x ratio, realistic HONO levels are simulated, being about one order of magnitude higher than the reference calculations, which only consider the reaction $OH+NO \rightarrow HONO$. The resulting enhancement of HONO significantly impacts HO_x levels and photo-oxidation products (e.g, O_3, PAN), mainly in polluted regions. Furthermore, the relative enhancements in OH and secondary products were higher in winter than in summer, thus enhancing the oxidation capacity in polluted regions, especially in winter, when the other photolytic OH sources are of minor importance. Our results underscore the need to improve the understanding of HONO chemistry and its representation in atmospheric models.

P-2-080 A review of operational, air quality ensemble forecasting modeling system for megacities (Beijing, Shanghai and Guangzhou) in China

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Key words
air quality ensemble forecast mega-cties in China chemical transport model

We have developed an air quality ensemble forecasting modeling system (EMS) in the past five years. The operational EMS system has successfully applied to the mega-cites including Beijing, Shanghai, and Guangzhou to afford air quality forecast service for the Beijing Olympics, the World Expo 2010, the Asian Games 2010. This ensemble system includes several air quality models (NAQPMS, Model3/CMAQ, CAMx, etc.), driven by them same meteorological forecast model (MM5) and several emission scenarios (perturbed NO_x or VOC etc.). Polynomial chaos method was applied firstly to analyze different uncertainty sources such as emission and wind field in chemical transport model, generating the evaluation of uncertainty for model outputs and the individual contribution from each sources. We also developed the ensemble Kalman Filter (EnKF) assimilation scheme to provide optimal initial conditions, optimal model parameters and then improved ensemble forecasts. The assimilation considering the model uncertainty into the EnKF shows that the ensemble spread now can maintain in an appropriate range and lead to much improvement of the forecasts. Highlight topics discussed in this paper include how regional chemical transport models are integrated into the EMS, how the performance of the each component model, how the ensemble results are formulated in Chinese mega-cites, the gaps of air quality ensemble forecast and potential priorities for future development.

P-2-081 Estimation of lifetime of carbonaceous aerosol from open crop residue burning during Mount Tai Experiment 2006 (MTX2006)

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Key words
lifetime of black carbon Organic carbon Biomass burning

Studying the emission ratios of carbonaceous aerosols (element carbon, EC, and organic carbon, OC) from open biomass burning helps to reduce uncertainties in emission inventories and provides necessary constraints for model simulations. We measured apparent elemental carbon (ECa) and OC concentrations at the summit of Mount Tai (Mt. Tai) during intensive open crop residue burning (OCRB) episodes using a Sunset OCEC analyzer. Equivalent black carbon (BCe) concentrations were determined using a Multiple Angle Absorption Photometer (MAAP). In the fine particle mode, OC and EC showed strong correlations ($r > 0.9$) with carbon monoxide (CO). Footprint analysis using the FLEXPART_WRF model indicated that OCRB in central east China (CEC) had a significant influence on ambient carbonaceous aerosol loadings at the summit of Mt. Tai. $\Delta ECa/\Delta CO$

ratios resulting from OCRB plumes were 14.3±1.0 ng/m3/ppbv at Mt. Tai. This ratio was more than three times those resulting from urban pollution in CEC, demonstrating that significant concentrations of soot particles were released from OCRB. ΔOC/ΔCO ratio from fresh OCRB plumes was found to be 41.9 ± 2.6ng/m3/ppbv in PM1. The transport time of smoke particles was estimated using the FLEXPART_WRF tracer model by releasing inert particles from the ground layer inside geographical regions where large numbers of hotspots were detected by a MODIS satellite sensor. Fitting regressions using the e-folding exponential function indicated that the removal efficiency of OC (normalized to CO) was much larger than that of ECa mass, with mean lifetimes of 27 h (1.1 days) for OC and 105 h (4.3 days) for ECa, respectively. The lifetime of black carbon estimated for the OCRB events in east China was comparably lower than the values normally adopted in the transport models. Short lifetime of organic carbon highlighted the vulnerability of OC to cloud scavenging in the presence of water-soluble organic species from biomass combustion.

P-2-082 Chemical characterization of $PM_{2.5}$ in a residential area of Beijing, China

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Key words
Aerosols chemical composition Beijing

Impacts of urban air pollution in Megacities are issues which will have major consequences to human welfare and quality of life. In view of the ongoing climate change – which will amplify environmental problems – it is important to convene with regional and local stakeholders in order to introduce suitable measures and to reduce and minimize air pollution levels and health impacts.

Beijing suffers from air pollution since the 1980s and ambient particle concentrations (PM) became a major problem in our days. A lot of emission reduction measures were performed to improve the air quality during the Olympic Summer Games in 2008, which has cut down coarse particles largely (see Suppan et al., this conference). The $PM_{2.5}$, which is more harmful than PM10, is still a problem and related investigation becomes more important.

To discuss typical aerosol characteristics during different seasons, two sequential high volume samplers (Digitel DHA-80, Hegnau, Switzerland) were used to collect $PM_{2.5}$ samples automatically at the campus of the CUGB which is located in the North West of Beijing from June 2010 to June 2011.

The comparisons of $PM_{2.5}$ mass concentrations with meteorological parameters provide some correlations with wind speed and relative humidity. It is also influenced by wind direction and precipitation and it is influencing visibility. Further, during the spring festival a real holiday situation exists each year in January/February.

Minor and trace elements are measured by PEDXRF (Polarized Energy Dispersive X-ray Fluorescence). Factor analysis is used to identify the sources of $PM_{2.5}$ in Beijing. The aerosols are originated from geogenic sources (found from Fe, Ti and Ba), which is most likely to be re-suspended road dust and dust storm, and also from anthropogenic sources (found from Zn, As and Pb), such as vehicle exhaust, chemical industry, coal combustion and fertilizer application.

P-2-083 Seasonal and diurnal variations of mono- and di-carbonyls in Xi’an, China

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Key words
Carbonyl compounds Seasonal and diurnal variation

Seventeen carbonyls in urban ambient air were quantified in summer (June 2009) and winter (January 2010) in an urban site located in Xi’an, China. Formaldehyde, acetaldehyde and acetone were the three most abundant carbonyls in the atmosphere with the concentration 6.54±2.38 ppbv, 2.08±1.07 ppbv and 2.74±1.14 ppbv in summer (from 14th to 24th June, 2009), respectively. In winter, the concentrations were 4.46±1.74 ppbv, 6.52±3.88 ppbv and 3.87±2.33 ppbv respectively from 4th January, 2010 to 10th January, 2010. Most carbonyls had higher concentrations in winter than in summer. And majority of the species had higher concentrations in daytime than in nighttime, indicating photochemical oxidation/human activities played an important role in diurnal variation. Formaldehyde/acetaldehyde ratios (F/A) in summer of 2.14 was much higher than 0.47 in winter, showing significant effect of photochemical oxidation in the urban air during summer. Acetaldehyde/propionaldehyde (A/P) average ratio was 12.2 in wintertime, implying anthropogenic emission was the major source of carbonyls in Xi'an. In addition, the ratio of acetone to methylglyoxal (A/M) is used to determine the impact of photochemistry in the atmosphere. The average acetone/methylglyoxal ratio (10.3±2.3) in summer was lower than that in winter (21.3±5.1) in Xi'an. Strong correlations among some carbonyls imply that they came from the same pollution sources. Formaldehyde and acetaldehyde play a very important role in photochemical smog formation. Methylglyoxal and glyoxal also have significant contribution to ozone formation potential.

P-2-084 Emission inventory to estimate co-benefit of Low carbon Society policy in Asian countries.

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Key words
Emission inventory Co-benefits

In order to prevent global climate change, a lot of low carbon society scenario and countermeasures are proposed in national scale to city scale in Asian countries. These low carbon society policies also have a large co-benefit which decreases the air pollution impact to human health, agriculture and eco-systems. In this study, the emission estimation model of the air pollutant from the industry, transportation, power generation, residential and commercial section with the detailed spatial distribution was developed to estimate the co-benefit of low carbon society policy to air pollution quantitatively.

Especially, for the emission from transportation sector, we developed two quantification methods to estimate current and future emission from road transportation depending on the spatial scale.

One method was developed for country scale. In this scale, we used a top-down estimation technique from various international and national statistics for transportation and social activities. Estimated traffic amount for each mode were down-scaled to local administrative boundary and major road segments. We applied this method to all East Asian countries and estimated traffic relate CO,

PM, NO_x and NMVOC on 1 km scale.

Another method was developed for city scale. In this scale, we used a bottom-up estimation technique. All individual trips were generated in the model according to the various information, such as population distribution, vehicles ownership, commuting area and so on. When the traffic census or person trip survey exists for the target area, it is used to adjust to consistent with them. We applied this method to Johor state, Malaysia as a case study of LCS scenario development.

P-2-085 Contribution of garbage burning to chloride and PM2.5 in Mexico City

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Key words
garbage burning chloride

The contribution of garbage burning (GB) emissions to chloride and PM2.5 in Mexico City is investigated during the period from March 24 to 29 in association with the MILAGRO-2006 campaign using the WRF-CHEM model. When the MCMA (Mexico City Metropolitan Area) 2006 official emission inventory without biomass burning is used in simulations, the WRF-CHEM model significantly underestimates the observed particulate chloride in the urban and suburban area of Mexico City. GB emissions remarkably improve the simulations of particulate chloride and contribute more than 60% of the observation, indicating GB is a major source of chloride in the city. GB yields up to 3 pbb HCl at the ground level in the city, which is mainly caused by the burning of polyvinyl chloride (PVC). GB is also an important source of PM2.5, constituting about 3-30% simulated PM2.5 mass on average. More work is needed to evaluate the GB contribution to hazardous air toxics, such as dioxin, which is found to be released highly from PVC burning in lab experiments.

P-2-086 Determining Aerosol Hygroscopicity through Airborne In-situ and Remote Sensing Observations in an Urban Environment during NASA DISCOVER-AQ

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Key words

Aerosol Measurement Hygroscopicity

The DISCOVER-AQ (Deriving Information on Surface Conditions from Column and Vertically Resolved Observations Relevant to Air Quality) mission conducted its first field deployment in Washington D.C./Baltimore region. The overarching goal is to better understand the remote sensing column measurements for diagnosing the near surface air quality. To this end, aerosol optical properties (scattering and absorption) are the basis for complex remote sensing retrievals and their relationships with chemical composition can bridge the gap between satellite observations and air quality measurements. Aerosol hygroscopicity, the propensity of a particle to take up water, is an important parameter for understanding how particles scatter visible light. Both hygroscopicity and absorption enhancements are inherently related to the chemical composition of particles; changes to that composition (photochemical aging) can result in increased hygroscopicity and absorption. This results in a complex vertical and spatial distribution of aerosol properties in urban environments.

To achieve the science objectives, the DISCOVER-AQ project adopted a sampling strategy involving two NASA aircraft, i.e., P-3B and UC-12, for highly coordinated vertical profiling. The P-3B was dedicated for in-situ observations and repeatedly spiraled over the 6 MDE (Maryland Department of the Environment) ground sites. The UC-12 was used to conduct remote sensing measurements over the region of the interest in coordination with the P-3B sampling. The P-3B aircraft was equipped with comprehensive aerosol measurements for microphysical, optical and chemical properties. We present aerosol scattering, absorption, and hygroscopicity observations from the P-3B airborne observations. The dry aerosol scattering coefficient was measured by a TSI nephelometer and the absorption coefficient was determined by a particle soot absorption photometer (PSAP). A closure analysis has shown high level of consistency between the observed dry scattering coefficient and Mie-theory calculation based on independently observed aerosol number size distribution. The aerosol hygroscopicity is derived from dry (relative humidity (RH) $< 40\%$) and humidified (RH > 75 %) scattering coefficients measured by two additional nephelometers. The derived hygroscopicity was then assessed by comparing the in-situ observed aerosol extinction coefficients (sum of scattering and absorption coefficients) corrected to ambient humidity with the extinction profile measured by HSRL (high spectral resolution lidar) system, which is conducted under ambient conditions. Comparisons are made over for vertical profiles in close proximity in space and time with the HSRL measurements. Among the nearly 250 NASA P-3B vertical profiles flown during the DISCOVER-AQ field deployment, a total of 85 cases were indentified for the comparison. The cases cover an AOD range of less than 0.1 to 0.7 and RH value range from 2 to 96%. A close agreement was found in the comparison which provides a high level of confidence for both the hygroscopicity model and for our ability to accurately measure AOD.

P-2-087 Kathmandu and Beyond: Addressing Urban and Regional Air Pollution in the Himalaya

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Key words
Himalaya Kathmandu black carbon

The Kathmandu Valley in Nepal has a severe and worsening air pollution problem, with contributions by local emissions, long-distance import, bowl-shaped topography, and local meteorology. Broad rural areas of the Himalaya have also seen rapidly worsening air pollution in the past decade, with impacts on health, visibility and local/regional climate. At least part of the rapid melting of Himalayan snow and ice can be attributed to the air pollutant and climate forcer black carbon.

The International Centre for Integrated Mountain Development (ICIMOD), headquartered in Kathmandu, is an eight-country intergovernmental organization with a mandate to address issues that affect the mountain people of the Greater Himalaya. Three initiatives have recently commenced at ICIMOD to address these issues.

The first, SusKat (“Sustainable Atmosphere for the Kathmandu Valley”) is a joint project between the Institute for Advanced Sustainability Studies (IASS) in Potsdam, Germany, and ICIMOD. It will improve scientific understanding of the atmospheric chemistry and physics and of air pollution health impacts in the valley through an intensive field campaign in late 2012, followed by modeling exercises. This will be followed by an analysis of mitigation and policy options.

The second initiative, “Reducing the Impacts of Black Carbon and other Short-lived Climate Forcers” will build up in-house scientific expertise at ICIMOD, including establishing atmospheric observatories and a data and modeling center; it will also analyze and pilot ways to reduce emissions from brick kilns, cook stoves, and vehicles, contribute to regional policy, and to the dissemination of knowledge.

The third initiative, “Atmospheric Composition and Pollution Transport in the Annapurna Himalaya” is led by the University of Virginia with local support from ICIMOD and Nepal Wireless. Measurements of CO, BC, ozone, AOD and meteorology at stations north and south of the Annapurna Himalaya, and in the Kali Gandaki Gorge connecting the two sides will contribute to the quantification of the transport of air pollutants from South Asia towards the Tibetan Plateau.

P-2-088 Observation of a Prominent Phase Lag between Ambient SO_4^{2-} and SO_2 Concentrations during Two Long Range Transport Events:

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Key words

Sulphur Dioxide (SO_2) Sulphate (SO_4^{2-}) Plume Transport OH radical Oil Fire

Simultaneous measurements of ambient sulphur dioxide (SO_2) and sulphate (SO_4^{2-}) aerosol (PM2.5) have been made from two sites (i) Shillong (25.67°N, 91.91°E, 1064 m ASL) – a high rainfall site in northeast India and (ii) Mt. Abu (24.6°N, 72.7°E, 1680 m ASL) - a high altitude site in the semi arid region of western India, during two major long range transport events. The variability in the molar ratio of SO_4^{2-} to total sulphur compounds ($SOx = SO_2 + SO_4^{2-}$) termed S-ratio - an indicator of the atmospheric oxidation efficiency of SO_2 – is also studied. At Shillong, in March 2009 when a major transient long range transport of SO_2 containing plumes was reported (Francis, 2011) during a dust storm event, from source regions in Russia, the median S-ratio obtained was unusually low (0.03) compared to that for January 2010 (0.32) - when major long range transports were absent. The time series SO_4^{2-} and SO_2 concentrations also differed significantly between the two sampling periods in the relative 'phase lag' between the SO_4^{2-} and SO_2 peaks. During the days 9th, 10th and 11th March 2009, when the SO_2 variability could not be explained by the PBL height effects and governed solely by the long range transport, the SO_4^{2-} and SO_2 concentrations were almost in anti-phase which is explained by the differences in the mobility and loss mechanisms of SO_4^{2-} (particulate matter) and SO_2 (gas) to cause a delayed arrival of SO_4^{2-} from the source regions at far distances. From 12th March 2009 onwards the SO_2 variability was found governed by the PBL height variations and not by the fluctuations in the long range input. Hence for the days 13th to 20th of March 2009, the observed anti correlation between SO_4^{2-} and SO_2 can be explained by a depleted OH radical concentration in the atmosphere – a major portion of which might got utilized during the initial days for the oxidation of large amounts of SO_2 - to lead to a suppressed oxidation efficiency. During 21st to 26th March 2009, when the SO_2 concentrations reduced significantly from the initial days, the SO_2 and SO_4^{2-} variations came in-phase with each other indicative of a regain of the oxidation capacity of the atmosphere via sufficient enough build up of OH radicals. Similarly, at Mt Abu during November 2009, when an SO_2 plume transport from a major oil fire event in Jaipur (26.92°N 75.82°E) was reported (Francis, 2012), the SO_4^{2-} peaks which were in phase with SO2 during the initial sampling days slowly started moving out of phase and became anti-phase with each other by the second half of the sampling period. This is again explained by the different transit times and loss mechanisms of SO_4^{2-}- and SO_2 as well as a depleted OH radical concentration in the atmosphere. These two instances are suggestive of using the 'phase lag' between SO_4^{2-} and SO_2 peaks as an 'indicator' for the 'presence' of long range transported plumes over sampling sites.

P-2-089 Seasonal distributions and characteristics of airborne particulate dicarboxylic acids, ketoacids and a-dicarbonyls in Xi'an, China

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Key words
PM_{10} diacids winter summer characteristics

PM_{10} aerosols in winter and summer in 2009 in Xi’an, China were studied for dicarboxylic acids, ketoacids and a-dicarbonyls, as well as inorganic ions, water soluble organic carbon (WSOC), organic carbon (OC) and related carbonaceous species. Diacids were characterized by a predominance of oxalic acid (C2) (1162 ng m^{-3} in winter and 767 ng m^{-3} in summer) followed by t-phthalic (tPh) in winter (250 ng m^{-3}) and phthalic (Ph) in summer (103 ng m^{-3}), which was different from other Asian cities where C4 was the second abundant specie. High concentrations of tPh, Ph and C2 in winter suggest extensive emissions from coal burning and plastic wastes incineration. SO_4^{2-} (56 μg m^{-3} in winter and 41 μg m^{-3} in summer) was the major ions and WSOC took up 56% and 78% of OC in winter and summer, which implied pollutants had been seriously aged from coal combustion, vehicle exhausts and industries. Ratios of C3/C4, C6/C9, Ph/C9, F/M together with the correlations between C2 and C3 and C4 indicated aged aerosols in Xi'an, while different seasonal ratios were interpreted as enhanced photochemical oxidation in summer. Correlation between C2 and C2 provided evidence for photochemical process of C2 in summer. Diacids-C/TC ratios and correlation between diacids and ketoacids also showed aerosols were photochemically aged in summer. High loadings of OC, EC, WSOC, WSIC and inorganic ions were ascribed to huge amount of primary emissions and unique meteorological conditions. Correlations of C2 and WSOC, SO_4^{2-} and C2 combined with RH characteristic demonstrated formation of C2 through aqueous oxidation in winter.

P-2-090 An Investigation of Winter Haze Episode in Beijing: Analysis Based on Online Measurements for Physical and Chemical Properties

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Key words
episode size distribution diurnal variation inorganic species air masses

This study examined the physical and chemical properties of an air pollution episode with visibility less than 3 km and high $PM_{2.5}$ concentration on December 13, 2011 in Beijing. This campaign was conducted from December 10 to 23 at three different ambient sites in Beijing area. Concentrations of $PM_{2.5}$, BC and other chemical components in bulk and single particles as well as particle number concentration and size distribution were measured with online instruments and offline PM samplers. Three high time resolution instruments, including Aerosol Chemical Speciation Monitor (ACSM), Single Particle Aerosol Mass Spectrometer (SPAMS) and Fast Mobility Particle Sizer (FMPS) were employed to study detailed chemical composition and particulate size distribution during this haze episode. Comparison of characteristics between the episodic and a typical non-episodic (clean) day in Beijing revealed that particles during haze day were different in (1) size distribution: size distribution from FMPS measurements shifted from peaking in 10-40 nm in the clean day to larger size (about 100 nm) during the haze episodic day, (2) diurnal variation of major components of fine particles: concentrations of sulfate-nitrate-ammoniums (SNAs) from ACSM in the episodic day increased gradually after 10 am, reached the highest concentration around 2 pm and remained relatively constant until 11 pm. However, organics based on ACSM measurements increased from 60 ug/m3 around 5 pm to about 110 ug/m^3 during 7-8 pm, suggesting the haze episode was first dominated by SNA and later OC played a major role, (3) inorganic species (elements): concentration of Pb from SPAMS measurements was also higher during the episodic day than a non-episodic day, indicating higher PM in the haze day was contributed from multiple components and sources besides SNA and organics, (4) air masses: back

trajectory analyses were performed by the HYSPLIT model and the results indicated air mass was mainly from the South of Beijing during the episode day, while air mass in the clean day was primarily originated from the North. The comparison of sources and formation mechanisms of this episode with other haze episodes in Beijing will be also discussed and presented in this research.

P-2-091 New trends in air quality management policies in China: Prospects and challenges

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Key words

New trends air quality management policies China Prospects challenges

This article surveys major recent changes in China's air quality management policies and assesses their significance, prospects, and challenges. Inadequate implementation of existing policies is one longstanding and well understood problem in China. Some new policies are intended to strengthen implementation, as well as address other structural problems, which have been recognized by policy makers, such as the increasing domestic transboundary nature of air pollution and increasing overall complexity of air pollution in terms of the number of pollutants and their interactions. In 2010, the "Guiding Opinion on Promoting the Joint Prevention and Control of Atmospheric Pollution to Improve the Regional Air Quality," based in part on the experience with the Beijing Olympics, set up a regional system to coordinate air pollution management between provinces, which, in the long run, could lead to greater coordination from the center. It also introduced a multi-pollutant management approach. These measures are in line with the National 12th Five-Year-Plan which also strengthened the targets and regulation for air quality including the emissions of SO_2, NOx, PM_{10}, and volatile organic compounds (VOC). Several related policies have also been issued, or are under the discussion. The paper concludes that the overall trend is towards steady strengthening of the policies, and that they are also aimed at strengthening implementation. However, the research also showed that the legal linkages between the policies are very complex, and the initial efforts to develop these policies as well as to prepare for their implementation has required considerable coordination efforts. Therefore, it remains to be seen how successful these policies will actually be in strengthening implementation, and in any case, it will require considerable coordination efforts between different levels of government.

P-2-092 Satellite observations of air quality in China's megacities

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In the last three decades, air pollution has become a major environmental issue in metropolitan areas of China as a consequence of fast industrialization and urbanization, and the rapid increase of the vehicle ownership. Now in China there are 3 megacities (Beijing, Shanghai and Guangzhou) in existence. A recent study of Asian megacities showed that they cover less than 2% of the land area, hold more than 30% of the population and produce about 10% of the anthropogenic gas and aerosol emissions. Therefore, it is important to qualify and understand current air pollution distribution and development in and around the megacities of China. Satellite observations provide unique insight into the regional air quality around megacities and air pollution transport from surrounding areas.

In this work, we present an investigation of air quality over Beijing, Shanghai and Guangzhou combining satellite and ground-based measurements. Aerosol optical thickness (AOT), precursors of ozone (notably NO_2 and CH_2O), and SO_2 are observed from space. The operational GOME-2 trace gases products developed at German Aerospace Center will be used. To observe the characteristics of aerosol over megacities, AOT acquired from the MODIS instruments onboard NASA's Terra and Aqua satellite are analyzed. Moreover, near surface concentrations of particular matter (PM), NO_2 and SO_2 in Beijing, Shanghai and Guangzhou are investigated.

The effect of air pollution transport from neighboring areas to megacities will be researched using satellite measurements. Long-term NO_2, SO_2 and CH_2O data recorded from GOME, SCIAMACHY and GOME-2 are analyzed to investigate possible trends in air pollutants for China's megacites. Initial comparison between satellite and ground-based measurements of air pollutants in Beijing, Shanghai and Guangzhou will be shown. We will present the relationship between AOT and PM concentrations in megacities. The use of AOT, tropospheric NO_2 and CH_2O columns for air quality applications will also be shown.

P-2-093 Addressing Science and Policy Needs with Community Emissions Efforts

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Key words
Emissions GEIA ECCAD CIERA

We present an overview of the community-driven emissions efforts within the

Global Emissions InitiAtive (GEIA, http://www.geiacenter.org/), a joint IGAC/iLEAPS/AIMES initiative of the International Geosphere-Biosphere Programme. Since 1990, GEIA has served as a forum for the exchange of expertise and information on emissions. GEIA's mission is to quantify anthropogenic emissions and natural exchanges of trace gases and aerosols; and facilitate the use of this information by the research, assessment, and policy communities.
GEIA supports a worldwide network of over 1200 developers and users in international scientific projects, providing a solid scientific foundation for atmospheric chemistry research. Moving forward, GEIA is broadening its role to serve the scientific, regulatory, and operational emission communities. GEIA intends to demonstrate the potential for improving emission information by promoting the interoperability of datasets and tools and by making use of near-real-time observations. As a first step toward these goals, two new programs are being linked with GEIA:
- ECCAD (Emissions of chemical Compounds & Compilation of Ancillary Data, http://eccad.sedoo.fr/) is GEIA's new interactive emissions data portal, providing consistent access to emission inventories and ancillary data with easy-to-use tools for analysis and visualization. - CIERA (Community Initiative for Emissions Research & Applications, http://ciera-air.org/) is a new GEIA community project to develop interoperability in emissions datasets and tools, support evaluations of inventories, communicate emissions information in innovative ways, and connect the emissions development and user communities. We will present the conclusions of the 2012 GEIA Conference and how these might inform future emissions research directions. We invite the scientific and policy community to join the GEIA network and build partnerships to improve emissions information.

P-2-094 Satellite-based estimates of reduced CO and CO_2 emissions due to traffic restrictions during the 2008 Beijing Olympics

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Key words
CO_2 emissions transportation sector

We present estimates of the reductions in CO and CO_2 emissions resulting from the control measures on the Beijing transportation sector taken during the 2008 Beijing Olympics. This study used MOPITT (Measurements Of Pollution In The Troposphere) multispectral satellite measurements of near－surface CO along with WRF－Chem (Weather Research and Forecasting model with Chemistry) simulations for Beijing during August, 2007 and 2008 to estimate changes in CO due to meteorology and emissions. Using fractional changes in the emissions inventory transportation sector along with a reported CO/CO_2 emission ratio for Beijing vehicles, we find the corresponding reduction in CO_2 emissions. We then compare this reduction to target CO_2 emissions in the RCP (representative concentration pathway) scenarios being considered for the IPCC AR5 (Intergovernmental Panel on Climate Change, 5th Assessment Report). Our result suggests that urban traffic could play a significant role in meeting target reductions for global CO_2 emissions, even for the most aggressive control scenario (RCP2.6).

P-2-095 Temperature threshold of biogenic isoprene emission in subtropical urban and suburban areas

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Key words

Secondary organic aerosols (SOA) Isoprene Temperature threshold Vehicular tracer

Measurement of speciated volatile organic compounds (VOCs) was conducted in a subtropical metropolis to investigate the characteristic of biogenic and anthropogenic isoprene, which deserves attention due to its extremely strong photochemical reactivity in producing ozone and secondary organic aerosols (SOA). Biogenic emissions possess a significant fraction of isoprene in the summertime, especially for subtropical cities. During nighttime, the observed isoprene showed a pretty good correlation with traffic-related tracer-1,3-butadiene, indicating that nighttime isoprene is mostly anthropogenic. Thus, the observed ambient isoprene in the subtropical city was compared with 1,3-butadiene to estimate the biogenic fraction vs. the anthropogenic fraction during daytime. Furthermore, there seems to exist a temperature threshold, beyond which the biogenic fraction increases with ambient temperatures. The threshold varied with different areas and mainly depended on local plant species.

P-2-096 Carbonaceous Aerosols over the Eastern Mediterranean and Black Sea: Primary versus secondary sources

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Key words

Organic Carbon Elemental Carbon water-soluble organic carbon Primary sources Secondary sources Eastern Mediterranean Black Sea

Carbonaceous material constitutes an important fraction of aerosol mass. However its role in the Mediterranean and Black sea atmosphere is yet to be unravelled. To

fill this gap daily aerosol samples were simultaneously collected at two remote (Finokalia, Crete, Greece and Gökçeada, Turkey), two highly populated urban centers (Athens, Istanbul) and two rural sites in Turkey (Sinop, Erdemli), from April 2009 to February 2010. Approximately 1500 samples were collected and analyzed for organic and elemental carbon (OC and EC) and water-soluble organic carbon (WSOC).

The average OC and EC concentrations at Istanbul were much higher than those measured at the other sampling sites, due to the abundance of anthropogenic sources. OC at the urban and rural sites demonstrate a clear seasonal variation with a minimum during summer and a maximum during winter. On the other hand no clear seasonal variability was observed at the remote areas of Gökçeada and Finokalia. In the case of EC, a seasonal cycle similar to OC is observed only at Istanbul and Erdemli. In Gökçeada and Finokalia, the opposite tendency is observed with a clear summer maximum, due to absence of local sources and wash-out of OC and EC during winter-time. The OC to EC carbon ratios are used to identify the presence of primary organic aerosols. At the remote locations of Finokalia and Gökçeada (OC/EC ratios of 6.2 and 5.3, respectively) OC is mostly of secondary origin, whilst in Istanbul and Athens the lower OC/EC ratios (2.1 and 2.5) underline the role of traffic-related or/and industrial originated sources. A good correlation between WIOC and EC (r^2=0.99) in Istanbul indicates the influence of primary emissions. In addition, the significant correlation of OC and EC with no-sea-salt potassium, nitrate, oxalate, and sulfate indicated that OC and EC sources in Istanbul are mainly due to fossil-fuel combustion.

Secondary Organic carbon (SOC) determined based on the relation SOC = OC-(OC/EC) primary are higher in Finokalia, Sinop, Gökçeada and Erdemli (average 77% of total OC) than in Istanbul's (average 52%), reflecting the abundance of primary sources of pollution at Istanbul.

Regarding the carbonaceous composition, organic matter constitutes a significant part of the total mass of PM10 (25.5±12.9% for Particulate Organic Matter and 4.5±3.4% for EC). The percentage of WSOC ranges from 34-38% of the OC for the rural regions to 50-67% for the background areas, while for the urban sites it constitutes 30.3±5.1% of OC and 7.0±3.6% of the total PM10 mass. At all sites the WSOC to OC ratio presents a clear seasonal variability with higher values during summer. The decrease during winter is due to the lack of photochemistry and to wet depositional losses of WSOC.

P-2-097 Air quality in the east mediterranean: impacts of emissions, temperature and mitigation

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Key Words

East Mediterranean Emissions Mitigation

High concentrations of air pollutants are often observed in East Mediterranean due to the local anthropogenic emissions, long-range transport and high background levels from natural sources. Major urban centers in the studied region include Istanbul megacity (GIA) and the Greater Athens Area (GAA). Urban centers are mainly exposed to elevated primary gaseous pollutants like CO and SO_2 as well as particulate matter, whereas the downwind locations experience high secondary pollutant concentrations like O_3 and inorganic aerosols. In the present study, we investigate the summertime levels of gaseous and particulate pollutants in the East Mediterranean using a mesoscale WRF/CMAQ modeling system. A number of scenarios have been simulated to quantify the contributions of local and regional anthropogenic and biogenic emissions to the air quality in the area as well as the sensitivity of the natural emissions and pollutant concentrations to potential increases in ambient temperatures.
The results show that Istanbul and Athens emit large amounts of precursors that lead to secondary pollutant production downwind. The local anthropogenic emissions are largely responsible for the particulate matter levels in the cities while they lead to suppressed levels of O_3. Impacts of regional anthropogenic emissions on the domain-mean pollutant levels (up to 17% for summertime O_3 and 52% for wintertime $PM_{2.5}$) are much higher than those from GIA and GAA anthropogenic emissions together (~1% for O_3 and ~6% for $PM_{2.5}$, respectively). The concentrated emissions in the hot spot regions are shown to enhance NO_x emissions impact on the domain mean levels of NO_x and HNO_3 during winter, whereas this is not the case for summertime emissions. On the other hand, small but no significant environmental gain is computed for $PM_{2.5}$ for the entire region by the concentration of pollutant emissions in the hot spots. Biogenic emissions are found to have a limiting effect on the aerosol formation in the region during

summer while in winter, the impact is negligible due to considerably low biogenic emissions. The temperature sensitivity simulations show that O_3 levels in the Eastern Mediterranean are expected to increase by 1 ppb K^{-1}, particularly due to elevated biogenic emissions. PM levels are subject to smaller changes with high spatial variations. Mitigation of anthropogenic emissions results in domain-mean O_3 mixing ratios by up to ~1%. Computed increases are large in Istanbul urban areas (up to 13%) while they are up to 2% in Athens urban areas. Domain-mean $PM_{2.5}$ levels are projected to decrease by up to ~10%. Overall, the results suggest improvements for most major pollutants in the urban areas, leading to decreases in the exceedences and the human exposure. The results show that the impact of long range transport is significant in the region and therefore, should be considered while quantifying and evaluating the impacts of emissions and mitigation.

P-2-098 Work Plans for Model Inter-Comparison Study - Asia Phase III (MICS-Asia III)

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In order to obtain common understanding of model performance and uncertainties in Asia, MICS-Asia Phase III following Phase I (1998-2000) and Phase II (2004-2009) was discussed, originated and carried out from 2008 to now. MICS-Asia Phase III is financially and technically supported by IAP and ACAP through Joint International Center on Air Quality Modeling Studies (JICAM). The work plans of MICS-Asia III has been discussed and determined preliminary in the Third Workshop on Atmospheric Modeling in East Asia in Chengdu in September 2011. And the work plans were further discussed and finalized at the International Workshop on MICS-Asia III in Beijing in July 2012.

In this project, three topics are included: Topic 1: Model Inter-comparison (Leader: Zifa Wang, IAP); Topic 2: Development of reliable emission inventories in Asia (Leader: Jung-Hun. Woo, Konkuk University); Topic 3: Air Quality/Climate Change (Leader: Greg Charmichael, Iowa University).

The objectives of the three topics have been defined as following: Topic 1: To evaluate strengths and weaknesses of current multi-scale air quality models and provide techniques to reduce uncertainty in Asia; Topic 2: To develop a reliable anthropogenic emission inventories in Asia and understand uncertainty of bottom-up emission inventories in Asia; Topic 3: To provide multi-model estimates of radiative forcing and sensitivity analysis of short-lived climate pollutants.

In Topic 1, the key questions include three parts: (1) Assessing the ability of models to reproduce pollutants concentrations under highly polluted conditions (e.g. Regional Haze); (2) Quantifying uncertainties of each process and model resolutions and model resolutions. In particular, uncertainties in key boundary layer parameters need to be addressed; (3) Investigating the air quality responses to specific emissions perturbations in a common case.

In this topic, current global (CHASER, GEOS-Chem, CACTUS, GEATM, MOZART, etc.) and regional (CMAQ, CAMx, NAQPMS, RAQM2, WRF-Chem, etc.) air quality models would be chosen. The models should include three common domains covering the whole East Asia (10S-50N; 90E-150E) with a coarse resolution of 45 km, Northeast Asia including Northeast and Southeast China, Korea and Japan with a resolution of 15km, main megacities in East Asia with a fine resolution of 9km, respectively. Long time simulation from 2008 to 2010 will be carried out for seasonal variation and inter-annual variability of wet deposition and atmospheric concentrations. Typical high air pollutant concentration cases in 2010 will be chosen and simulated. All models should use the same emission fields including anthropogenic emission, natural emission, and they should use the same meteorological model (WRF) to drive. The models should use the same boundary conditions from a global model during 2008 and 2010. For evaluating abilities of each model in Asia air quality modeling, all participants should perform base year model simulation and compare model results of air quality and meteorology with the best available observation data. For obtaining source-receptor relationship and future projection, all participants should perform model simulation for air quality responses in various areas to specific emission perturbations. And the chemical mechanism, transportation, deposition, boundary layer process and model resolutions should be compared for each model to evaluating the impact of uncertainties of the key process on model results.

In Topic 2, to evaluate strengths and weaknesses of current emission inventory and provide techniques to reduce uncertainty and improve performance in Asia, reliable emission inventories in Asia would be developed. Anthropogenic emission datasets, natural emission datasets covering the whole Asia would be developed for emissions inter-comparison study in 2008, 2010 and 2030. In this Topic, a modeling emission inventories would be developed and provided to the air quality models. And at last, the discrepancies between bottom-up and top-down emission inventories should be investigated.

In Topic 3, the objective is to evaluate strengths and weaknesses of current Air Quality/Climate Change models and ensemble estimates of Air Quality/Climate Change quantities to reduce uncertainty of impacts. The participants should provide estimates of SLCF distributions and deposition for use in health, ecosystem, and climate studies, and estimates of radiative forcing and regional responses for base case and selected emission scenarios. Based on the results, multi-model estimates would be obtained. And the analysis of the sensitivity of estimates to key processes or inputs would be provided in this topic.

Now, the MICS-Asia III is carried out step by step. And it would provide more reliable simulation of air quality in Asia.

P-2-099 Contribution of long-range transport to Eastern Mediterranean air pollution levels: A 3-D modeling study

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Key words
long-range transport Eastern Mediterranean basin Air pollution Ozone PM

The Mediterranean, located at the boundary between the tropical and mid-latitudes is a crossroad of air masses coming from Europe, Asia and Africa. In this region, anthropogenic emissions, mainly from Europe, Balkans and the Black Sea, meet with biomass burning emissions and with naturally emitted Saharan dust. During the last decades, the region has experienced a rapid growth in urbanization, reflected by increasing pollutant emissions to the atmosphere. The Eastern basin of the Mediterranean includes megacities (i.e. Cairo and Istanbul) as well as several large urban centers (like Athens). Air pollution is a challenging environmental problem in the Eastern Mediterranean basin (EM) since both ozone and aerosol air quality limits are often exceeded during the year. Air quality abatement strategies require quantification of the contribution of the various sources to the pollutant levels in the area.

For this purpose, the contribution of long-range transport (LRT) to air pollution levels in the EM has been investigated using the global chemistry-transport model TM4-ECPL. The model simulates ozone chemistry including non-methane volatile organic compounds and all major aerosol components, including secondary aerosols like sulfate, nitrate and secondary organic aerosol. It accounts for multiphase chemistry that is producing secondary aerosols in clouds and in the aerosol water. TM4-ECPL is driven by the European Center for Medium-Range Weather Forecasts Interim re-analysis project meteorology.

The model is able to simulate air pollutant levels in the rural and remote atmosphere in the EM as recorded by surface-based and satellite observations. It reasonably reproduces the tropospheric O_3 column maximum in the EM observed by satellites and high levels in the North East of the Black Sea and in the South East of the Persian Gulf. According to TM4-ECPL model calculations, LRT significantly contributes to the O_3 burden in the EM. Eastward transport of O_3 within the free troposphere from upwind locations is calculated to contribute almost 80% to the total O_3 sources in the EM. Furthermore, downward transport from the free troposphere is estimated as the major source of O_3 into the boundary layer of the EM (~60%), whereas the contribution of local anthropogenic sources to boundary layer O3 levels is only limited.

Overall, the simulations show that air quality in the EM is strongly affected by sources other than the local anthropogenic sources. A three-dimensional budget analysis quantifies the EM air-pollution footprint and documents inter-country influences and natural contributions. These findings suggest that improvements in air quality in the region require coordinated efforts between the countries surrounding and located upwind of the basin.

P-2-100 Seasonal Interactions between East Asian Summer Monsoon Circulation and Direct Effects of Anthropogenic Aerosols in Model SensitivityExperiments

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Using a regional climate model coupled with a chemical model (RegCM4.1), interactions between anthropogenic aerosol direct effects and East Asian summer monsoon (EASM) circulation has been examined. In order to explore those interactions, two experiments are conducted. One is the control experiment without anthropogenic aerosols (SO_2, BC, OC), and the other is with the aerosols. Modeled and observed EASM circulations are firstly compared, and it is found that RegCM4.1 can well reproduce the climatology of the variations of the EASM circulation. The model results suggest that the aerosols play different roles during different stage of the EASM circulation. When the EASM is in south, the aerosols are favorable for the EASM marching north; however, when the EASM is in north, the aerosols weaken the EASM circulation. This phenomenon can be explained by the feedback mechanism between the EASM circulation and the aerosol direct effects; the EASM changes the distribution of the aerosols, and the aerosols hence alter the stability of the atmosphere, which is (or not) favorable for the development of the EASM circulation.

P-2-101 The concentration of Elemental and Organic Carbon in $PM_{2.5}$ during and after the 2011 Shenzhen UNIVERSIADE

ZHANG Yang, ZHANG Yuanxun, WANG Yuqin

Carbonaceous matter is a major component of ambient atmospheric aerosol being mainly associated to particulate matter with diameter less than 2.5 micrometer ($PM_{2.5}$). The daily samples of $PM_{2.5}$ were collected simultaneously at two sites in Shenzhen during (August 12-23) and after (August 24 to September 4) the 2011 UNIVERSIADE. One sampling site was located on the roof of the 4-story Building on the campus of Shenzhen Graduate School of Peking University (S1). The other site was located on the roof of a 31-story Building about 1 km far from the Main Stadium of UNIVERSIADE Center (S2). The distance between the two sampling sites is about 45 km. The concentration of elemental carbon (EC) and organic carbon (OC) were obtained with an OC/EC analyzer (Sunset Lab). The mean ± standard deviation of OC and EC at S1 were 4.26±1.93 μg m^{-3} and 2.83±1.63 μg m^{-3} (n=36) during the UNIVERSIADE, whereas their concentrations after the UNIVERSIADE were 15.30±7.29 μg m^{-3} and 4.33±2.26 μg m^{-3} (n=35), respectively. The mean ± standard deviation of OC and EC at S2 were 5.85±1.67 μg m^{-3} and 2.98±0.89 μg m^{-3} (n=36) during the UNIVERSIADE, whereas their concentrations after the UNIVERSIADE were 13.09±2.57 μg m^{-3} and 3.60±1.14 μg m^{-3} (n=36). These results show that the levels of OC and EC during the UNIVERSIADE were lower than after the UNIVERSIADE, presumably due to the restrictions on sources and reduction traffic volume during the UNIVERSIADE.

In addition, meteorological parameters (temperature, atmospheric pressure, relative humidity, wind speed and wind direction) were obtained simultaneously. The atmospheric transport characteristics in Shenzhen were discussed via using the backward trajectory model. Furthermore, the speciation analyses of organic matter in $PM_{2.5}$ were conducted by GC-MS.

P-2-102 **Evaluating NOx Emissions Using Satellite Observations**

FROST Gregory, KIM Si-Wan, BRIOUDE Jerome, TRAINER Michael, MCKEEN Stuart, LEE Sang-Hyun, HSIE Eirh Yu, ANGEVINE Wayne, GRANIER Claire, RYERSON Thomas, PEISCHL Jeff, FEHSENFELD Fred, HECKEL Andreas, HILBOLL Andreas, RICHTER Andreas, BURROW John, GLEASON James, and BOERSMA Folkert

P-2-103 **Measurements of NO_2 mixing ratios with Topographic Target Light Scattering-Differential Optical Absorption Spectroscopy System and Comparisons to Point Monitoring Technique**

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Key word:
ToTaL-DOAS atmospheric optical atmospheric trace gas

A Topographic Target Light Scattering-Differential Optical Absorption Spectroscopy (ToTaL-DOAS) system is developed for measuring averaging concentrations along a known optical path and studying surface-near distributions of atmospheric trace gases. The telescope of the ToTaL-DOAS system is pointed to targets which are located at known distances from the measuring device and illuminated by sunlight. Averaging concentrations with high spatial resolution can be retrieved by receiving sunlight reflected from the targets. Erna Frins et al first discussed the principles of ToTaL-DOAS and analytical methods in 2006. The group explored the spatial resolution of ToTaL-DOAS using three targets, which were 146m, 196m, 280m apart from the instrument in 2008. A ToTaL-DOAS system which is built for experiment, includes the telescope with small field of view (FOV) and large aperture and a two-dimension rotating platform. A filed measurement of NO2 concentration was performed with the ToTaL-DOAS system in Shijiazhuang city of china in the autumn of 2011. The measurement data are compared with concentrations from the point monitoring technique at the same site. The results show ToTaL-DOAS is sensitive to describe variation of NO_2 concentrations along the optical path.

P-2-104 **Aromatic hydrocarbons is the Pearl River Delta (PRD) region from 2007 to 2009: emission sources and contribution to ozone formation**

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Introduction

Aromatic hydrocarbons (AHs) as an important class of VOCs are not only hazardous air pollutants but also dominant anthropogenic precursors of ozone and secondary organic. The rapid economic growth in the PRD region has resulted in fast increase in anthropogenic emissions of air pollutants in this region. With the industry in the PRD region hard-hit due to financial crisis, it is a unique opportunity to assess the role of industrial emission from ambient monitoring before and after the outbreak of financial crisis, and in the processes of economic recovery. The purpose of this study is to better understand the emission sources of aromatic hydrocarbons (AHs) and their contribution to ozone formation in the region for more efficient and accurate control strategies to mitigate regional ozone pollution. The field sampling site Wanqingsha (WQS, 22.711ºN, 113.549ºE), a small town southernmost of Guangzhou in the centre of the PRD, is a good regional monitoring station to characterize the air pollution in inner PRD.

Methods

Ambient VOCs samples were collected from Nov. to Dec. in the year of 2007, 2008 and 2009 using cleaned and evacuated 2-Liter electro-polished stainless steel canisters. The air samples were analyzed using a Model 7100 preconcentrator (Entech Instruments Inc., California, USA) coupled with an Agilent 5973N gas chromatography-mass selective detector/flame ionization detector (GC-MSD/FID, Agilent Technologies, USA).

Results and discussion

AHs shared of 44.2% in average in 2007, while in 2008 it decreased to 22.1% after a hard-hit by the financial crisis from the second half year of 2008, however, with the economy recovery the proportion of AHs increased to 25.6% in 2009. The contributions of AHs to total OFPs in 2007 (60.3%) were significantly larger than those in 2008 (25.8%) and 2009 (26.4%). Source apportionment by positive matrix factorization (PMF) revealed the predominantly sources of benzene are combustion of fuels and biomass, the contributions of vehicle exhaust source (including gasoline and diesel) increased from 67% in 2007 to 75.2% in 2008 and 75.6% in 2009 accompanying with the vehicle numbers increase. For toluene and C8-aromatics (including ethylbenzene and xylenes), two major sources, industrial emissions and vehicle exhausts, were identified with contribution over 90% in the three years. Contributions of industrial emissions to toluene and C8-aromatics dropped from 46.3% and 65.9% in 2007 to 29.3% and 15.7% in 2008, rebounded to 57.4% and 58.8% in 2009. Contributions to OFP by different sources based on OFP-PMF results the industrial emission shared the largest part in 2007 (40.8%), while, increased OFP percentages from vehicle exhaust and LPG were observed in 2008 and 2009. Further investigation into local industry output statistics suggested that the plummet of AHs in 2008 should be attributed to small enterprises, which contributed largely to ambient AHs due to their huge numbers and non-existent emission treatment, but were much more influenced by the FC.

Conclusion

Our study revealed the important role of industrial emission to AHs in the highly industrialized PRD region, the dominant contribution of toluene and xylenes from industry sources due to their widely use as solvents, and the need of emission control for small factories in the PRD region in reducing AHs for the reduction of secondary pollutants like ozone and secondary organic aerosols in ambient air.

Poster Presentations, Session 3

P-3-001 Toward a combined SAGE II-HALOE aerosol climatology

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Key words
stratospheric aerosol climate SAGE II HALOE

Herein, the Halogen Occultation Experiment (HALOE) aerosol extinction coefficient data is evaluated in the low aerosol loading period after 1996 as the first necessary step in a process that will eventually allow the production of a combined HALOE/SAGE II (Stratospheric Aerosol and Gas Experiment) aerosol climatology of derived aerosol products including surface area density. Based on these analyses, it is demonstrated that HALOE's 3.46 micron data is of good quality above 19 km and suitable for scientific applications above that altitude. However, it is increasingly suspect at lower altitudes and should not be used below 17 km under any circumstances. The 3.40 micron is biased by about 10% throughout the lower stratosphere due to the failure to clear NO_2 but otherwise appears to be a high quality product down to 15 km. The 2.45 and 5.26 micron aerosol extinction coefficient measurements are clearly biased and should not be used for scientific applications after the most intense parts of the Pinatubo period. Many of the issues in the aerosol data appear to be related to either the failure to clear some interfering gas species or doing so poorly. For instance, it is clear that the 3.40 m aerosol extinction coefficient measurements can be improved through the inclusion of an NO_2 correction and could, in fact, end up as the highest quality overall HALOE aerosol extinction coefficient measurement. It also appears that the 2.45 and 5.26 micron channels may be improved by updating the Upper Atmosphere Pilot Database which is used as a resource for the removal of gas species otherwise not available from direct HALOE measurements. Finally, a simple model to demonstrate the promise of mixed visible/infrared aerosol extinction coefficient ensembles for the retrieval of bulk aerosol properties demonstrates that a combined HALOE/SAGE II aerosol climatology is feasible and may represent a substantial improvement over independently derived data sets.

P-3-002 The Stratospheric Aerosol and Gas Experiment on the International Space Station

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Key words
SAGE III ISS ozone aerosol

The Stratospheric Aerosol and Gas Experiment (SAGE) III is anticipated to be installed on the International Space Station (ISS) sometime after summer 2014. This is the fourth generation of visible/near-IR solar occultation instruments operated by the National Aeronautics and Space Agency (NASA), the first coming under a different acronym, to investigate the Earth's upper atmosphere. Three flight-ready SAGE III instruments were built by Ball Aerospace in the late 1990s and delivered to NASA Langley Research Center. One was launched aboard the former Russian Aviation and Space Agency Meteor-3M (M3M) platform in 2001 and continued until the platform lost power in 2006. Another of those instruments was manifested for the ISS in the 2004 time frame, but was delayed due to budgetary considerations and placed into storage at NASA Langley. Fortunately, that SAGE III/ISS mission was restarted in 2009 with a major focus upon filling an anticipated gap in ozone and aerosol observations in the second half of this decade. An important component of this exciting mission is the European Space Agency contribution of the Hexapod pointing platform, which allows SAGE III to be in the correct position prior to gathering its high precision measurements. The 52-degree inclined orbit of the ISS is well-suited for solar occultation and provides near-global observations on a monthly basis with excellent coverage of low and mid-latitudes, similar to that of the SAGE II mission (1985-2005). The nominal science products include vertical profiles of trace gases, such as ozone, nitrogen dioxide and water vapor, along with multi-wavelength aerosol extinction. An advancement of the SAGE III instrument over the SAGE II model is a system with a large dynamic range (~million) enabling lunar occultations on a routine basis to augment the solar products. The standard lunar products, ozone, nitrogen dioxide & nitrogen trioxide, were demonstrated during the SAGE III/M3M mission. The operational flexibility of the SAGE III instrument facilitates main goal of producing ozone and aerosol extinction profiles, while allowing exploration of new possibilities for the occultation technique, such as night-time aerosol extinction profiles or other trace gases not measured by SAGE in the past. We present an overview of the mission architecture, its implementation, the data produced by SAGE III/ISS including their expected accuracy, coverage and plans for data quality evaluation (aka validation).

P-3-003 SLCF Co-benefit Scenario in East Asia

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Key words
Co-benefit Co-control East Asia
SLCF Co-benefit Scenario in East Asia

Introduction

Co-benefit, co-control approach for air pollution and climate change mitigation is of particular importance in East Asia, which consists of many rapidly growing developing countries. Targeted SLCF (SLCP) are generally agreed to be black carbon (BC) and ozone (O_3). However, there is a different view between EU/US and Asia on which precursor(s) should be controlled for O_3 reduction. EU and US approaches exclusively prioritized CH4 mitigation for O_3 reduction. We propose here prioritized NOx/VOC reduction rather than CH_4 for East Asia. Regional O_3 pollution in East Asia is much more serious than in EU and US, and are highly concerned from human health impact. Reducing NOx/VOC is more efficient for reducing regional O_3 concentration, which can justify the co-benefit, co-control policy in East Asia.

Method

The three IIASA reference scenarios for 2030 and base year emission of 2005 are used in the present study. Starting from the “CO2-eq. 450 ppm stabilization scenario”, CLE (Current Legislature Scenario) and MFR (Maximum Feasible Reduction Scenario), climate and air quality (O_3 and $PM_{2.5)}$ changes are evaluated in 2030. A Global Chemical Climate Model, CHASER-MIROC-4.5, and Regional Chemical Transport Model, WRF(v.3.1.1)-CMAQ(v.4.7.1) were used for model evaluation of global climate and air quality change, and regional detailed air quality change in East Asia, respectively. The former provided boundary conditions to the latter as well.

Results and Discussion

As for climate change, SLCF (CH_4+O_3+BC) reduction gives direct negative forcing by ca. 0.1 and 0.5 W m^{-2} for the “450 ppm” and MFR scenario, respectively, in 2030, which are almost compensated by direct positive forcing by SO_4^{2-} due to reduction of SO_2. Large positive indirect forcing in 2030 is predicted due to the reduction of SO_4^{2-} and other aerosol.

As for air quality change, the “450 ppm Scenario” does not reduce much of O_3 in 2030 in East Asia. Although it reduces $PM_{2.5}$ substantially in China, but much more stringent control is needed to meet better air quality.

From these results, it is concluded that in East Asia more stringent air quality scenario for NOx/VOC is necessary to justify co-benefit approach.

P-3-004 Hygroscopic Properties of Internally Mixed Ammonium Sulfate and Phthalic Acid Particles

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Key words
hygroscopicity internal mixture ion-molecule interactions

The hygroscopicity of aerosol particles is a property with profound implications. Hygroscopicity affects the particle size and phase, and then influences many physicochemical characteristics of the aerosol, including optical properties, atmospheric lifetime and chemical reactivity. Changes in aerosol hygroscopicity can therefore lead to changes in both direct and indirect radiative forcing of climate. Aerosol particles typically consist of inorganic and organic components. While the water uptake properties of inorganic salts as a function of relative humidity (RH) are generally well-known, the hygroscopic properties of organics are not yet fully understood. Moreover, organics are typically found to be internally mixed with inorganic compounds, which may alter the hygroscopic behavior of inorganic components. Phthalic acid (PA) has been identified as the main species of aromatic acids existed in the atmospheric fine particles. However, the effects of PA on the hygroscopic properties of inorganic salts by homogeneously internally mixed and heterogeneously coated modes have not been well-characterized in earlier studies.

A Hygroscopicity Tandem Differential Mobility Analyzer (H-TDMA) system has been designed to investigate the hygroscopicity of PA and ammonium sulfate (AS) homogeneously internal mixing and heterogeneously coating particles. The FTIR-ATR method has been used to monitor the molecule structural changes on the deliquescence processes of AS/PA homogeneous internally mixed particles.

The results show that the PA particles start absorbing small amount of water at low RH and exhibit continuously growth with increasing RH. In addition, over the size range studied (50, 100 and 200 nm), no size dependence is observed to the water uptake by PA. Homogeneously internally mixed AS/PA particles display abrupt deliquescence transitions at 70% RH. Above the DRH, mixed droplets show successive hygroscopic growth, however, the growth factor is obviously reduced relative to pure AS, and the growth factor increment shows a significant diminution with increasing PA fraction. Similar to homogeneously internally mixed particles, heterogeneously coated particles consisting of AS core and PA shell deliquesce at 70% RH, followed by hampered consecutive hygroscopic growth with increasing RH. The relative growth factor increment presents a significant reduction with increasing PA shell thickness. The FTIR-ATR spectroscopy spectra of the homogeneous internal mixing of AS and PA indicate that there are interactions between sulfate ions and PA molecules in the mixture. It is proposed that the interactions probably lead to the mixed particles deliquescence at 70% RH and hamper hygroscopic growth above 70% RH. These results indicate that the presence of phthalic acid significantly affects the water uptake by atmospheric ammonium sulfate aerosols.

P-3-005 A modeling study about the combined effect of aerosols and its influence on summer monsoon in China

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Key words
Aerosol Climatic effects China Summer monsoon

A coupled regional climate-chemistry modeling system (RegCCMS) is applied to investigate the distribution, radiaitive forcing as well as the influence of aerosols on summer monsoon in China. In this study, parameterizations for cloud droplet number concentration and precipitation autoconversion rate are introduced into RegCCMS to investigate the indirect effect of aerosols. Modeling results show that sulfate, nitrate, BC and OC aerosols are mainly distributed in East and North China as well as Sichuan basin because of industrial distribution and terrain, while dust is mainly distributed in Northwest China. The annual average of total radiative forcing of aerosols at the top of atmosphere is -4.43W/m^2. Owing to the combined effect of aerosols, the countrywide annual average changes of surface air temperature and precipitation are -0.59K and -0.31mm/d, respectively, showing significant differences in seasons and regions. Climate effects of aerosols are stronger in summer than in other seasons. As a result, summer monsoon is weakened in eastern China and the subsequent precipitation is decreased in most part of China.

P-3-006 Development and application of analytical system of Perfluorocyclocarbon tracers for Lagrangian aircraft experiments

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Key words
PFCs tracer Lagrangian SHIVA

Lagrangian experiment is a powerful tool in atmospheric investigations of the

evolution of various airborne species. It is crucial to evaluate and confirm the validity of Lagrangian experiments and improve their performance by tagging the air mass of interest with a tracer. Perfluorocyclocarbons (PFCs) are rapidly becoming the exotic tracers of choice because of their favorable properties. They are inert, non-toxic, non-scavenged by water, and contribute neither to stratospheric ozone depletion nor to photo-chemically induced air pollution. The global warming impact of PFCs is negligible at present, due to their extremely low atmospheric background concentrations. The analytical system with high selectivity and sensitivity to detect PFCs in the atmosphere has been developed by using thermal desorber (TD) based on two steps of desorption and gas chromatography (GC) equipped with negative ion chemical ionization mass spectrometry (NICI-MS), due to PFC's high reactivity with free electrons. It allows a dynamic range of perfluorocarbon tracer concentration measurements of over 3 orders of magnitude. The background concentration of PFCs in the atmosphere was determined by this analytical system. In Aug of 2011, PFC tracer has been released at Hohenpeißenberg by a prototype PFC Release Unit designed and constructed by DLR. Total 42 available tracer samples have been collected by aircraft tracer sampler TDS-another prototype instrument of DLR-in the research aircraft Falcon. Furthermore, tracer experiment was carried out during

SHIVA campaign in Malaysia in Nov of 2011. The background determination of PFCs verifies the feasibility of this analytical method. The measurement results of tracer experiments of Hohenpeißenberg and Malaysia SHIVA will be shown. Initial comparison with Hybrid Single Particle Lagrangian Integrated Trajectory Model (Hysplit 4) forecast result will also be presented.

P-3-007 Preliminary results of an ongoing year-long study characterizing physical and chemical properties of PM in one of the world's most PM-polluted cities, Ulaanbaatar, Mongolia

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Key words
aerosols field campaign outdoor air pollution urban air pollution particulate matter black carbon regional climate

Lagrangian experiment is a powerful tool in atmospheric investigations of the evolution of various airborne species. It is crucial to evaluate and confirm the

validity of Lagrangian experiments and improve their performance by tagging the air mass of interest with a tracer. Perfluorocyclocarbons (PFCs) are rapidly becoming the exotic tracers of choice because of their favorable properties. They are inert, non-toxic, non-scavenged by water, and contribute neither to stratospheric ozone depletion nor to photo-chemically induced air pollution. The global warming impact of PFCs is negligible at present, due to their extremely low atmospheric background concentrations.

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P-3-008 Management of plant genotypes and soil C-N ratio as biological mitigation option for nitrous oxide emission from wheat (Triticum aestivum) agriculture

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Key words
Nitrous oxide Wheat genotypes Soil C-N ratio Mitigation

Nitrous Oxide (N_2O) is a potent greenhouse gas which contributes significantly to global climate change and agriculture is one of the important sources of N_2O emission. In view of its increasing concentration in the atmosphere, emphasis has been laid on exploring suitable ways to mitigate N_2O emission from agricultural sources. Two parallel experiments were conducted during the wheat growing season from December 2010–April 2011 at Tezpur University Campus, to study the effect of genotypes and soil C-N ratio management on N_2O emission. Three genotypes of wheat viz. Sonalika, DBW 39 and K 0307 were grown in a randomized block design and N_2O emissions were recorded from the soil planted with these cultivars. For soil C:N management, the field with cultivar Sonalika was treated with organic fertilizers having different C: N viz cow dung (25:1), rice straw (41:1) and poultry manure (15:1) and N_2O emission was compared with nitrogen fertilizer treatment. Gas (N_2O) sampling was done at weekly interval by using the Static Chamber Method and analyzed in a gas chromatograph to determine its concentration.

N_2O emission from these genotypes ranged from 12.90-286.53 μg $N_2O\text{-}Nm^{-2}h^{-1}$. Highest seasonal integrated N_2O flux (Esif) was recorded from DBW 39 (124.84 mg N_2Om^{-2}) and lowest from Sonalika (103.25 mg N_2Om^{-2}). N_2O emission was found to be directly associated with plant morphophysiological characters like, plant height, leaf area, and leaf and tiller number in both the studies. Scanning Electron Micrographs of stomatal frequency of the adaxial leaf surface and xylem size of culm have revealed that high N_2O emitting varieties have higher stomatal frequency and larger size of xylem. Low emitting cultivar (Sonalika) recorded higher grain yield, indicating that, efficiency of photosynthate partitioning between source and sink organs may be instrumental in determining cultivar differences in N_2O emission from wheat. Different quantities of N_2O emission ranging from 13.24 – 175.53 μg $N_2O\text{-}Nm^{-2}h^{-1}$ were recorded in soil treated with different C-N ratios. Lowest Esif (89.28 mg N_2Om^{-2}) was recorded in cow dung (C:N – 25:1) and highest (144.83 mg N_2Om^{-2}) in poultry manure (C:N-15:1). Organic fertilizer with optimum C:N (25:1) was identified to reduce N_2O emission whereas low C:N (15:1) resulted in higher N_2O emission in comparison to inorganic nitrogen fertilizer (NPK) application. Highest yield was recorded from the soil applied with poultry manure (30.22Q/ha) and lowest with rice straw (14.04Q/ha). Soil organic carbon, soil nitrate and soil moisture content had substantial influence on N_2O emission. These experiments revealed that selection of suitable variety and C-N ratio management of organic fertilizers may provide effective and environment friendly mitigation options for N_2O emission.

P-3-009 Czech Globe – the national contribution to global atmosphere monitoring and research efforts

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Key words

Global climate change Long-term monitoring global carbon cycle long-range transport

The CzechGlobe project aims towards the construction of a spatially distributed research infrastructure, including research sites in various ecosystems, monitoring network and atmospheric research station. These components will provide the necessary background for research focused on the impacts of global climate

change on the atmosphere, terrestrial ecosystems and human society. This presentation is focused on the atmospheric segment of above mentioned project – atmospheric station Křešín u Pacova. The essential part of atmospheric station aimed to provide long-term monitoring, a 250 m high atmospheric mast (assigned the function of National Monitoring Point – NMP) is being constructed right next to the Košetice Observatory, operated by the Czech Hydrometeorological Institute. The Observatory was established in 1988 as a background station specialized in air quality monitoring and research (geographical co-ordinates: 49 35′ N; 15 05′E; 534 m a.s.l.) and represents Czech Republic in activities under Convention on Long-range Transboundary Air Pollution (Co-operative Programme for Monitoring and Evaluation of Long-Range Transmission of Air Pollution in Europe, Task Force on hemispheric Transport of Air Pollution and International Cooperative Programme on Integrated Monitoring), World Meteorological Organization (Global Atmosphere Watch) and in several international monitoring and research projects (EUSAAR, ACTRIS, ACCENT). The most important research and monitoring activities of the NMP include long-term measurement of greenhouse gases concentrations and their exchange dynamics under the Integrated Carbon Observation System (ICOS) project. ICOS aims to provide long-term observation required to understand the present state and predict future behaviour of the global carbon cycle and greenhouse gas emissions. Further, it monitors and assesses the effectiveness of carbon sequestration and/or greenhouse gases emission reduction activities on global atmospheric composition levels, including attribution of sources and sinks by region and sector. The ICOS monitoring programme will include continuous measurements of carbon dioxide, carbon monoxide, methane and meteorological parameters. Besides ICOS, the NMP will be also used for investigating the impacts of global climate change on air quality and long-range transport of air pollution. The research will be focused on pollutants with a high potential for hemispheric transport (tropospheric ozone, ultrafine particles and mercury). Special attention will be devoted to atmospheric aerosols which have besides greenhouse gases an important impact on global climate change processes. The measurements of absorption and diffusion of solar radiation by atmospheric aerosols are currently implemented at a limited number of ACTRIS (Aerosols, Clouds, and Trace gases Research Infrastructure Network) superstations, however, not yet in the Czech Republic. The instrumentation is based on ACTRIS recommendations. The research will also focus on the health features because negative impacts of atmospheric particles in the atmosphere on human health belong among the most serious environmental problems nowadays.

P-3-010 Unanticipated side effects of aerosol emissions on regional precipitation

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Key words

Ultrafine particles aerosol cloud interaction precipitation

Aerosol interactions in the climate system are among the main uncertainties in climate projections into the future. Aerosol effects are considered to be counteracting global warming by direct interaction with solar radiation and reductions of aerosol emissions are thus adding to current radiative forcing. Less well known are effects of aerosols on the water cycle due to aerosol - cloud microphysics interactions. Current directives to clean emissions lead to a significant change in the aerosol size distributions with large reductions of coarse and fine mode aerosol and a strong amplification of ultrafine particle numbers. These ultrafine particles act indirectly on the radiation budget. A possibly important side effect of these indirect processes involving clouds is the modification of the water and precipitation cycle.

We performed regional scale measurements of aerosol size distributions and characteristics of major aerosol sources in areas which are characterized by reductions in precipitation within the last decades while concurrently the water vapor (dewpoint) in the boundary layer in these areas was increasing. Such a mismatch between the available water vapor and precipitation is difficult to explain by modification of advection patterns and could be due to regional scale change in aerosol emissions. Data are presented from field campaigns in Australia and Germany and the possibility to trace back changes in precipitation spatial and temporal distributions to changes in aerosol emissions in pristine and moderately polluted conditions will be discussed.

P-3-011 Semi-direct and indirect climate effects of fossil fuel black carbon aerosol over China

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Key words

Black carbon Radiative forcing Climate effects Regional climate and chemistry model

A regional climate chemistry modeling system was applied to investigate spatial distribution of black carbon (BC) loading, radiative forcing (RF) as well as its climatic effects over China. Results show that high levels of BC distribute in southwest and central to eastern China. Simulated aerosol optical depth is large with maximum of 0.11. Annual mean cloud droplet number concentration, effective radius and auto conversion rate changed by +70.89 cm^{-3}, -0.154 μm and -7.51 10-6 g kg^{-1} s^{-1} due to BC indirect effects. Owing to the absorption, BC exerts a positive RF at the top of atmosphere (TOA) with the regional mean of +0.81 W

m^{-2}. Strong BC first indirect RF distributes in south, northeast China and north India, with the regional averaged value of -0.95 W m^{-2}. Combined (direct plus first indirect) RF at TOA over the region is -0.15 W m^{-2}. Climate responses to BC direct effect show significant decreases of low level cloud amount and liquid water path in whole east China, with regional average of -0.017 % and -0.104 g m^{-2}. Since more solar radiation was absorbed (+1.223 W m^{-2}), surface became warming and drying (Changed by 0.042 K and -0.003 mm d^{-1}). For total indirect effects, annual mean cloud amount, liquid water path, and total absorbed solar radiation over the region changed by 0.114%, 1.42 g m^{-2} and -2.02 W m^{-2}. Second indirect effect is more significant compared to BC first indirect effect. Surface cooling and drying appear in north India and south to east China, yielding regional averaged changes of -0.135 K for surface temperature and -0.06 mm d^{-1} for rainfall. Due to the combined effects of BC, annual mean cloud amount, cloud liquid water path and the radiative fluxes are changed by -0.025%, 1.28 g m^{-2}, -0.733 W m^{-2}. Considerable annual mean surface air temperature decrease in center and south China, with the maximum of -0.7 K. Total precipitation is inhibited in most parts of China (regional mean of -0.09 mm d^{-1}).

P-3-012 Air pollution, atmospheric oxidation capacity and haze-clouds in China: results from the IPAC-NC field campaign

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Key words

regional air pollution atmospheric oxidation capacity pollution acid coating haze-clouds aerosol indirect effect

North China (Huabei in Chinese) is a geographical region located between 32°-42°N latitude in eastern China, including several provinces and large municipalities, e.g., Beijing and Tianjin. In the past decades the region has experienced dramatic changes in air quality and climate. Among the multiple causes, regional photochemical smog and haze pollution has been expected to play a dominant role. To investigate this, a field measurement campaign was performed in April-May 2006 as part of the project - Influence of Pollution on Aerosols and

Cloud Microphysics in North China (IPAC-NC). This is the first aircraft measurements of atmospheric trace gases, aerosols and clouds over this part of China, a region strongly affected by both natural desert dust and pollution smog.

It appeared that strong pollution emissions from urban and industrial centers tend to accumulate in the lower atmosphere over the central area of Huabei. We observed very high concentrations of gaseous air pollutants and haze particles, partly together with non-precipitating stratiform clouds. The clouds were characterized by numerous droplets, much smaller than in a less polluted atmosphere. The high pollution concentrations were found to be associated with enhanced levels of OH and HO_2 radicals, calculated with a chemical box model constrained by the measurements. In the upper part of the boundary layer and in the lower free troposphere, high CO and SO_2 compete with relatively less NO_2 in reacting with OH, being efficiently recycled through HO_2, preventing a net loss of HO_x radicals. In addition to reactive hydrocarbons and CO, the oxidation of SO_2 causes significant ozone production over Huabei (up to ~13% or 2.0 ppbv h^{-1} at 0.8 km altitude).

The high OH levels tend to promote the formation of semi- and low-volatile species such as inorganic and organic acids through the oxidation of SO_2, NO_2 and VOCs. In spring, the atmospheric load of primary aerosols over Huabei is very high due to strong emissions from both natural and anthropogenic sources (e.g., dust and black carbon), providing a large surface area for the condensation of these oxidation products. The condensation rate of sulfuric acid was estimated to be 2-8 micro g cm^{-3} h^{-1} during IPAC-NC, with a maximum at about 0.8 km altitude. Our analyses reveal that the highly efficient coating of dust particles by pollution acids provides the predominant source of cloud condensation nuclei (CCN). The pollution-enhanced activation of dust particles into droplets is shown to be remarkably efficient so that clouds even form below 100% relative humidity. Contrary to previous analyses, we find that the haze particles influence the spectral shape of the cloud droplet size distribution such that the indirect climate cooling effect of aerosols on clouds is increased.

Our results indicate that the lower atmosphere over Huabei is not only strongly polluted but also acts as an oxidation pool, where pollutants undergo very active photochemistry, leading to haze-clouds and exerting an unusually cooling forcing on climate.

P-3-013 Spectrally-resolved measurements of aerosol extinction at ultraviolet and visible wavelengths

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Key words

Aerosol absorption optical properties

Aerosols play an important role in the Earth's radiative budget. Aerosol extinction includes both the scattering and absorption of light, and these vary with wavelength, aerosol diameter, and aerosol composition. Historically, aerosol absorption has been measured using filter-based or extraction methods that are prone to artifacts. There have been few investigations of ambient aerosol optical properties at the blue end of the visible spectrum and into the ultraviolet. Brown carbon is particularly important in this spectral region, because it both absorbs and scatters light, and encompasses a large and variable group of organic compounds from biomass burning and secondary organic aerosol.

We have developed a laboratory instrument that combines new, high-power LED light sources with high-finesse optical cavities to achieve sensitive measurements of aerosol optical extinction. This instrument contains two broadband channels, with spectral coverage from 360 – 390 nm and 385 – 420 nm. Using this instrument, we report aerosol extinction in the ultraviolet and near-visible spectral region. We have measured the extinction cross-sections between 360 – 420 nm with 0.5 nm resolution for different sizes and concentrations of polystyrene latex spheres, ammonium sulfate, and Suwannee River fulvic acid. Fitting the real and imaginary part of the refractive index allows the absorption and scattering to be separately determined.

P-3-014 Atmospheric aerosol impact on diffuse radiation and the Amazon biosphere

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Key words
aerosol Amazon biosphere radiation deforestation biomass burning

Over the past few decades a substantial increase in the Amazonian forests land carbon sink has been observed, with estimates equivalent to almost 10% of the anthropogenic fossil fuel emissions. The reasons behind this important carbon sink are still unknown, with a variety of possible mechanisms based on changes in temperature, carbon dioxide, precipitation, clouds, or solar radiation being suggested. The present work focuses on one such mechanism, namely the increase in canopy photosynthesis due to changes in the diffuse/direct radiation fraction caused by atmospheric aerosols from deforestation fires. We hypothesise that this mechanism, which has not yet been quantified over the Amazon basin, plays a significant role in the observed carbon storage increase. A recent global modelling study estimated that increased diffuse radiation due to anthropogenic aerosols has led to a 25% increase in the global land-carbon sink.

P-3-015 Addressing Air Pollution and Climate Change Together: A Science Policy Dialogue

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Key words
Air pollution Climate change

Air pollution is projected to be the world’s top environmental cause of premature mortality worldwide by 2050, ahead of dirty water and lack of sanitation. Current climate change mitigation actions will not be enough to prevent the global average temperature from exceeding the internationally agreed goal of 2˚C above pre-industrial levels by 2050. The International Geosphere-Biosphere Programme (IGBP) and the International Global Atmospheric Chemistry (IGAC) initiative on Air Pollution and Climate builds on current efforts calling for mitigation of short-lived climate forcers (SLCFs) by facilitating a science-policy dialogue that examines the multiple implications of existing and proposed mitigation policies that address air pollution and climate change in the near and long-term. In a recent statement, Time to Act: The Opportunity to Simultaneously Mitigate Air Pollution and Climate Change, the initiative went beyond addressing just SLCFs and examined linkages between air pollution and climate change, including how reductions of some air pollutants that lead to cooling, such as sulfur dioxide, will uncover warming from carbon dioxide already emitted and how climate change may render air pollution control management strategies less effective. It also called for an integrated approach to addressing air pollution and climate change in order to slow the rate of climate change and protect human health, food/water security and ecosystems.

Addressing air pollution and climate change together provides a unique opportunity to simultaneously achieve both air quality and climate policy goals in the near-term. However, addressing this issue would benefit from the development of a holistic framework to integrate air pollution and climate change solutions into economic development and broader decision processes in various local, national, regional, and global contexts. The integrated framework would be informed by scientific research that cuts across traditional disciplines to develop mitigation strategies adapted to the physical, economic, political and social contexts within a given nation or region. Possible strategies for ways forward outlining potential concepts will be discussed as part of the development of a strategic plan for an inter-disciplinary program on Air Pollution and Climate Change.

P-3-016 Concentrations measurements of selected trace gases such as CFCs and SF_6 in urban area of Krakow, Poland

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Key words
trace gases chlorofluorocarbons CFCs SF_6 ozone depleting substances greenhouse gases ECD measurements in the urban area

The concentrations of chlorofluorocarbons (CFCs) and sulphur hexafluoride (SF_6) in the atmosphere are on the ppt level. CFCs compounds are synthetic, stable, and they contribute to ozone depletion in the stratosphere. CFCs and SF_6 also participate in intensification of the greenhouse effect.

The measurements of CFCs and SF_6 in air are usually carried out at places situated outside of urban areas influence (“clean stations”). In Europe such clean station is Mace Head (Ireland), which participates in AGAGE program since 1987 and in InGOS program since 2011. This kind of research is also conducted in Central Europe, in densely populated area of Krakow (Poland) since 1997. Actually, measurement station of Krakow has opportunity to calibrate standards and to compare measurement data with results from e.g. Mace Head. The aim of these investigations is to determine an influence of Krakow area on CFCs and SF_6 concentration.

To obtain concentrations of measured compounds the mathematical procedure has been used. Concentrations were calculated using a five points Lagrange’s interpolation. Based on temporary measurement data, were determined daily arithmetic means and their standard deviations, and then values of monthly weighted averages. Also, it was made Gaussian filtration, to estimate trend of the base line of individual compounds.

It is observed, that after 1.07.2002, when the Montreal Protocol legislations were implemented in Poland (The Journal of Laws No. 52), frequency of seasonal variability of CFCs concentration pollution events are diminishing. The concentration of freon F-11 ($CFCl_3$), F-12 (CF_2Cl_2), F-113 (CCl_2FCClF_2), chloroform ($CHCl_3$), 1,1,1-trichloroetane (CH_3CCl_3), carbon tetrachloride (CCl_4) has a tendency to decrease, whereas sulphur hexafluoride (SF_6) tend to increase. This suggests that the most observed sources of CFCs have been localized to the territory of Poland (the wind rose and air mass movements have still the same statistical character in Krakow region, as before legislation implementation).

The work presents results from 15 years of research of air pollution in Krakow (in the years 1997-2012) by selected halocarbons and SF6. It also presents comparison of CFCs and SF_6 concentrations and trends in the atmosphere of Krakow with results from European measurement stations. Additionally the work discuss meteorological characteristics of Krakow region and their influence on observed concentration of measured compounds.

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P-3-017 An Overview of the Puerto Rico African Dust and Clouds Study (PRADACS) – Aerosol and Cloud Measurements at a Caribbean Tropical Montane Cloud Forest

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Key words

Tropical Montane Cloud Forest Aerosol-Cloud Interactions Long range transported African dust PRADACS

Aerosol-cloud interactions and precipitation are key aspects for tropical climate research, but lack of data limits our understanding of processes occurring in pristine regions of the world, like the Caribbean. As part of the Puerto Rico African Dust and Clouds Study (PRADACS), we aimed to determine how the presence of long-range transported African dust (LRTAD) impacts cloud properties and processes. Aerosols and cloud water were studied in a tropical montane cloud forest at Pico del Este (PE) research station (18°16' N, 65°45' W, 1051 masl) during dust and non-dust events in summers 2010 and 2011.

With HYSPLIT back-trajectory analysis and SAL images, the origin of air masses at PE was determined in order to identify dust and non-dust events. Bulk chemical composition (pH, conductivity, organic content and inorganic species – metals and water-soluble ions) of aerosols and cloud water collected in the presence and absence of LRTAD events were assessed and correlated to cloud physical properties (i.e., liquid water content - LWC, particle surface area, effective radius - Re, and cloud droplet size distribution). The results were complemented with online particle chemical analysis from by particle mass spectrometry (Aerosol Time of Flight Mass Spectrometer - ATOFMS, Aircraft based Laser Ablation Mass Spectrometer - ALABAMA, and Aerosol Mass Spectrometer - AMS) and aerosol morphology measurements using an Aerosol Particle Spectrometer with Depolarization (APSD).

Preliminary results showed that during dust events: 1) the number concentration of droplets smaller than 8 μm increased, leading to a decrease in Re (from fog monitor measurements, FM-100) 2) bulk cloud water chemical analyses showed an increase in pH and conductivity, the presence of non-sea salt particles containing iron and aluminum, and higher organic content; 3) single particle analysis from the ATOFMS revealed titanium and iron, indicating the presence of African dust; 4) aerosol particles measured with the APSD appeared to have mixtures characteristically similar to urban and Azores dust, depending on the air mass origin, and no interstitial particles above 5 μm were observed which could suggest a possible cloud removal mechanism; and 5) a non-decreasing particle absorption of the drop residues was observed, although residual particle number decreased substantially during the dust event, implying that the absorption does not come from black carbon.

Overall, differences in the studied physicochemical properties of aerosols and clouds during dust and non-dust events were observed. Our preliminary results show that LRTAD events have an impact on cloud properties and processes. Demonstrating that LRTAD particles produce changes in the chemical and

physical properties of aerosols and clouds at the tropical montane cloud forest of Pico del Este can develop a better understanding of the impact of African dust particles on clouds and climate to reduce the uncertainties in terms of their radiative forcing. More results regarding the physicochemical properties of aerosols and clouds studied will be presented at the meeting.

P-3-018 Effects of the past climate changes on regional air quality in East Asia

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Key words
Air pollution meteorology Climate change Ozone Secondary inorganic aerosol

East Asia is one of the largest source regions of gaseous pollutants and aerosols due to the rapid industrialization, urbanization and the population growth and has experienced air quality deterioration and climate changes over the past. Increases in anthropogenic emissions are a main cause for the air quality degradation that is also sensitive to changes in climate conditions. In this study we examine the effects of the past climate changes on regional air quality in East Asia using the 3-D global chemical transport model (GEOS-Chem) simulations for the period 1985-2006. The model was driven by the GEOS assimilated meteorology with the emission estimates from the Streets et al. inventory with annual scale factors of Regional Emission inventory in Asia. The analysis of simulated results shows significant effects of climate changes on ozone and sulfate-nitrate-ammonium concentrations. Springtime ozone concentration has been generally increased over the past two decades mainly due to increases in anthropogenic precursors emissions but concurrent decreases of cloud covers further enhance ozone increases in East Asia. On the other hand, increases in temperature and large-scale precipitation over the past two decades slow down the increases of sulfate-nitrate-ammonium concentrations in summer. However, decreases in large-scale precipitation in winter results in enhancement of sulfate-nitrate-ammonium aerosols concentrations. Overall, the effects of past climate changes on ozone and sulfate-nitrate-ammonium aerosols concentrations are highly sensitive to individual meteorological variables that affect chemical and physical processes in the atmosphere. We analyze relationships between individual important meteorological variables, and ozone and aerosols concentrations based on the past changes, which can be interpreted for future air quality projection due to climate changes in East Asia.

P-3-019 Heterogeneous Uptake and Temperature Dependence of Hydrogen Peroxide on Mineral Oxides

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Key words
mineral oxides hydrogen peroxide heterogeneous uptake temperature dependence

The interaction of mineral oxides (Al_2O_3, MgO, Fe_2O_3 SiO_2 and $CaCO_3$) with hydrogen peroxidewas investigated using the Knudsen cell reactor. The initial reactive uptake coefficients for the commercially available powders are measured as $(1.00\pm0.11)\times10^{-4}$ for Al_2O_3, $(1.66\pm0.23\times10^{-4}$ for MgO, $(9.70\pm1.95)\times10^{-5}$ for Fe_2O_3, $(5.22\pm0.9)\times10^{-5}$ for SiO_2, and $(3.71\pm0.74)\times10^{-4}$ for $CaCO_3$. These metal oxide powders exhibit some catalytic behavior toward the decomposition of hydrogen peroxide excluding SiO_2 and $CaCO_3$. H_2O_2can be destroyed on Fe_2O_3 surface and O_2 is formed. The heterogeneous kinetic processes of hydrogen peroxide on silicon dioxide and calcium carbonate have also been studied over the temperature range from 253K to 313K and the functions of temperature were obtained. The results indicated that hydrogen peroxide could be adsorbed on SiO_2 and $CaCO_3$ reversibly in this temperature region and the initial uptake coefficient increased evidently with temperature decreasing. The quick uptake on these mineral oxides, expsicially at low temperature, provided an active surface for further reaction. The experimental results suggest that the heterogeneous loss on mineral surface can represent an important sink of hydrogen peroxide.

P-3-020 Long-term observations of saccharides in marine aerosols from the western North Pacific: A comparison between 1990-1993 and 2006-2009

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Key words
marine organic aerosol sugar compounds seasonal variation

We obtained two sets of four-year record of saccharides in total suspended particles (TSP) collected at a remote Chichi-Jima Island (Japan) in the western North Pacific from 1990 to 1993 and from 2006 to 2009 on a biweekly and weekly basis, respectively. Anhydrosugars (galactosan, mannosan and levoglucosan), sugars (xylose, fructose, glucose, sucrose and trehalose) and sugar alcohols (erythritol, arabitol, mannitol and inositol) were measured in the aerosol

samples using a capillary gas chromatography/mass spectrometry (GC/MS). Total concentrations of anhydrosugars, the biomass burning tracers, were 0.01-5.68 ng m^{-3} (average 0.76 ng m^{-3}) during 1990-1993 versus 0.02-11.7 ng m^{-3} (0.96 ng m^{-3}) during 2006-2009. Their seasonal distributions were characterized with maxima in winter/spring and minima in summer/fall. Such a pattern can be interpreted by the long-range atmospheric transport of terrestrial organic matter (such as natural biogenic detritus and soil dust) from Asian continent in winter/spring seasons, when westerly winds prevail over the western North Pacific. Sugars and sugar alcohols showed different seasonal patterns. The mean concentrations of erythritol, arabitol, mannitol, inositol, fructose, glucose and trehalose were found to be highest in spring/summer and lowest in fall/winter during both 1990-1993 and 2006-2009, indicating an enhanced biogenic emission source in warm seasons. Interestingly, total concentrations of saccharides gradually decreased from 1990 to 1993 but increased from 2006 to 2009. The decrease in early 1990s was probably caused by changes in regional meteorology (such as E1 Niño-Southern Oscillation).

P-3-021 How does tropospheric OH influence the transport of SO_2 from the surface to the stratosphere? – A conceptual study

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Key words
Trajectory study sulfur transport

The stratospheric sulfate aerosol layer is a key element in the climate system as it affects both the chemistry of the stratosphere and the absorption and scattering of radiation in the stratosphere. In the absence of major volcanic eruptions, the stratospheric aerosol layer is maintained by transport of sulfur containing species, predominantly carbonyl sulfide (COS) and sulfur dioxide (SO_2), from the troposphere, through the tropical tropopause layer (TTL), to the stratosphere. The lifetimes of these species, both in the troposphere and in the TTL, are largely determined by hydroxyl (OH) concentration. The product of the OH oxidation, SO_3, is highly soluble and is removed by rainout or deposition on ice. It is therefore the non-soluble fraction of these sulfur containing species that determines the flux of sulfur entering the lower stratosphere and contributing to the stratospheric aerosol abundances. The use of Lagrangrian trajectories to study transport processes from the troposphere to the stratosphere through the TTL is well established. In this study a Lagrangrian (trajectory based) chemistry transport

model (ATLAS) is used to investigate the sensitivity of SO_2 entering the stratosphere to tropospheric OH concentrations. Recent observations of very low ozone concentrations in the tropical western Pacific implied very low OH concentrations in that region. Using (i) the OH concentrations derived from ozonesonde measurements carried out during the Transbrom cruise with the Research Vessel Sonne in October 2009 and (ii) an OH field derived from the GEOS-Chem chemistry transport model, as initial concentrations, trajectories were initialized in the tropical lower stratosphere (440K) and then followed backward. Only trajectories that extend back to the boundary layer were used for further study. The breakdown of COS and the conversion of SO_2 into highly soluble SO_3 is calculated along these trajectories. The fraction of the initial COS and SO_2 abundances that reaches the stratosphere has been determined. The implications of reduced tropospheric OH concentrations on sulfur transport into the stratosphere will be shown. The results suggest that reduced OH over the Pacific warm pool (the main entry region for stratospheric air masses), enhances the fraction of SO_2 reaching the stratosphere and suggest that direct emissions of SO_2 into the troposphere from anthropogenic activity in South East Asia or from small scale volcanic activity in that region can have a larger effect on the stratospheric sulfur budget than would be derived from standard OH fields.

P-3-022 Determining Earth crustal material in particulate matter (PM)

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Key words
crustal matter weather conditions SEM-EDX

Introduction

Earth crustal matter is an integral part of the total particulate load in the atmosphere. Depending on site and weather conditions, it is one of the most important natural components. In respect to meteorological conditions, crustal matter is believed to be influenced more strongly than particles from anthropogenic sources. Quantification of the contribution of crustal matter to PM is mostly based on bulk chemical measurements using Si as a marker element. However, Si cannot be determined by ICP-MS since PM samples need to undergo an extraction step. A reliable alternative method is needed to quantitatively determine the Si concentration.

Methods

The ongoing study aims to determine the crustal component in size-segregated impactor samples. Samples are taken at three different sites on an event basis: one urban background station (DD) and two rural sites (M and OBB) in Eastern Germany. At all three sites, 24 hour samples are taken concurrently, and depending on the large scale synoptic weather conditions. Samples are analyzed by ICP-MS. In addition, a scanning electron microscope coupled with an energy dispersive X-ray detector (SEM-EDX) is used for single particle analysis, and for determination of the bulk Si-content. A method has been developed to determine the bulk Si-concentration from the bulk Al-concentration using the Si/Al ratio measured with SEM-EDX.

Results & Discussion

First results show that the developed method is applicable. The Si/Al ratios determined by SEM-EDX can be used to calculate bulk Si-concentrations. The Si/Al ratios are different for the three sites. DD shows the highest ratios (~3), OBB the lowest (~1). Depending on the size of the particles and the concentration of the crustal component, the uncertainties of the method vary. The resulting Si-concentrations are used to calculate the abundance of crustal matter. Crustal elements as Si, Al, Fe or Ti are influenced by air mass origin and weather conditions. Source identification based on single particle analysis reveals that the contributions of different sources fluctuate. Additional sources for the so-called crust elements can be fly ashes from combustion processes (e.g., coal burning, metallurgy, waste incineration).

Conclusions

Where other analytical methods are not applicable, SEM-EDX can be used to quantify Si in PM samples, even if only very small sample volumes are available. The resulting Si/Al ratios are essential to determine the abundance of crustal matter on the size-segregated aerosol samples. It is thus possible to investigate the relation between the crustal component concentrations and weather conditions. In addition, single particle analysis enables the distinction between anthropogenic fly ashes and natural crust material. In bulk chemical analysis, these two components are always merged in one group that is erroneously interpreted as being of completely natural origin.

P-3-023 Evaluation of modeled vertical aerosol distributions over Europe using in-situ and satellite data

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Key words

regional modeling aerosol properties satellite observations short-lived climate forcers

As part of the EU ECLIPSE project, which aims to quantify the climate impact of short-lived climate forcers (SLCFs), including aerosols, black carbon and ozone, regional models are being used to evaluate global model performance for specific case studies. Here, we present results using regional WRF-Chem simulations run with different aerosol schemes over Europe. Results are compared to satellite data and field campaigns which took place in spring and summer 2008.

The aim is to evaluate the ability of the models to simulate the aerosol physical, chemical and optical properties, with a focus on pollution layers over source regions and during transport downwind. The radiative impact of such layers over Europe is also examined as a function of their relative positions to clouds. The WRF-Chem regional model was run using MOZART gas phase chemistry and different aerosol schemes and evaluated against the measurements. The model was run using anthropogenic and fire emissions for 2008, while boundary conditions were specified using the fields from a global chemical transport model.

The radiative impact of pollution aerosol layers has already been investigated but less is known about the influence of vertical layering in the atmosphere. Such layers might have different radiative impacts whether they are below or above clouds and in that sense, a better understanding of their spatial extent is critical. Information about pollution aerosol layers and clouds optical properties and positions over Europe are determined using satellite-based remote-sensing measurements (CALIPSO lidar). The radiative impact of these layers has been evaluated and compared to the observations.

In addition to satellite observations, data providing information on aerosol physical, optical and chemical properties from 2008 measurements campaigns over Europe (e.g., EUCAARI and POLARCAT-France) have been used to evaluate the simulations. In this study, we assess, for exemple, aerosol total number concentrations and size distributions simulated by the model. The aerosol aging is also evaluated bu examining the ratio between elementary and organic carbon (EC:OC), while the aerosol origins and sources are investigated using Lagrangian back-trajectories and observed chemical compositions, respectively. Specific attention is given to the simulation of observed pollution aerosol layers identified by CO and aerosol total number concentration enhancements.

P-3-024 Size patterns and CCN activity of less hygroscopic aerosol particles observed in the urban atmosphere during summer

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Key words
urban aerosol cloud condensation nuclei HTDMA

Primary organic carbon and elemental carbon (EC) can be emitted from anthropogenic sources in the urban environments. It is plausible that at least some of the particles containing these primary components are hydrophobic in the beginning, and that the soluble fractions of the particles increase by aerosol aging. Hence, to assess the role of urban aerosols to cloud formation, it is important to understand the chemical characteristics and the ability to act as cloud condensation nuclei (CCN). In this study, we obtained the size patterns (i.e., the estimated size distributions with an approximation) of less hygroscopic particles in the urban air and compared them with the chemical composition of PM1. Further, CCN in these particles was analyzed and the possible change in the abundance of soluble fractions is discussed.

Aerosol measurements were performed in August 2011 in Nagoya, Japan. For the analysis in this study, the data collected from 16 to 25 August were used. The sampled urban aerosol was introduced to a hygroscopicity tandem differential mobility analyzer (HTDMA) and classified according to the particle hygroscopic growth factor (HGF) at 85% relative humidity. The number fraction of CCN in the particles selected using the HTDMA were obtained based on the measurement using a cloud condensation nuclei counter (CCNC) and the condensation particle counter in the HTDMA. The size-resolved number fraction of CCN in the particles with HGF of unity was obtained under 1% and 0.5% supersaturation (SS) conditions, and the fraction in the particles with HGF of 1.1 was obtained at 1%SS. The HGF distributions of particles with the dry mobility diameter of 150 nm were also obtained. The chemical composition of non-refractory PM1 was obtained using a high resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS). The mass concentrations of EC and organic carbon in PM1 were measured using a semi-continuous carbon analyzer.

The median and mean diameters of the estimated size patterns of less hygroscopic particles with HGF of unity varied with time. Whereas the temporal variation in the mass concentration of hydrocarbon-like organic aerosol (HOA) shows relatively high positive correlation with the less hygroscopic particles in the small size range (71-102 nm), the variation in the mass concentration of EC shows relatively high positive correlation of with less hygroscopic particles in the large size range (175-359 nm). The result suggests the size dependence of the chemical composition of less hygroscopic particles, i.e., both HOA and EC exist in the less hygroscopic particles and HOA exists in the diameter range that is smaller than that of EC. The size-resolved number fraction of CCN in the less hygroscopic particles also varied with time, suggesting the changes in the amount of hydrophilic components in the particles. The causes of the change of the CCN fraction may include the enhanced emissions of fresh primary particles during rush-hours and the temporal changes of air masses at the studied area. The result suggests the complexity of the chemical composition and the CCN activity of less hygroscopic particles in the urban atmosphere.

P-3-025 Characterization of the size distribution of organic aerosol components over a forest site in Japan

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Key words
HR-ToF-AMS Biogenic SOA PMF analysis

Oxidation of volatile organic compounds (VOCs) from terrestrial vegetation forms biogenic secondary organic aerosol (BSOA). Despite the large contribution to organic aerosols on the global scale, their formation mechanisms, the physical and chemical properties, and the evolution processes are not understood well. Biogenic VOCs and their oxidation products have been studied using traditional off-line techniques in the forests of East Asia; however, characterization of biogenic SOA from a real-time measurement (e.g., using an aerosol mass spectrometer) at a forest site in the East Asia is not reported to our knowledge. This study aims to characterize the size distributions of organic aerosol components in the forest atmosphere in East Asia based on on-line aerosol mass spectrometry.

A field measurement of atmospheric aerosol particles was conducted at a mid-latitude forest site in Wakayama, Japan (34.07 degrees N, 135.52 degrees E), from 20 to 30 August 2010. The mass concentrations and the size distributions of chemical components in non-refractory submicron particulate matter (NR-PM1) were measured using a high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS). The number size distributions of aerosol particles were measured using a scanning mobility particle sizer (SMPS).

On the days when new particle formation (NPF) was not evident (from 20 to 25 August 2010), the size distributions of organics (or sulfate) were mainly peaked at ~500 nm (or ~600 nm) in vacuum aerodynamic diameter both during daytime and nighttime. On NPF event days (from 26 to 30 August 2010), a prominent diurnal evolution of the size-resolved mass concentrations of organics was observed, particularly in the small size range (below 200 nm) during the daytime. By contrast, the size distribution of sulfate was almost constant and it mainly peaked at ~450 nm. The average size-resolved vapor condensation sink (CS) that derived from particle number size distributions showed bimodal distributions on NPF event days: one peak in the Aitken mode and the other in the accumulation mode. The occurrence of NPF and the subsequent growth of new particles accompanied the increase of CS in the Aitken mode size range, which presumably enhances the mass concentration of organics in this size range as a result of the gas-to-particle conversion of biogenic VOCs. The size distribution of two organic components derived from the deconvolution of organic mass spectra with positive matrix factorization (PMF), being assigned as a highly oxidized, low-volatile oxygenated organic aerosol (LV-OOA) and a less oxidized, semivolatile OOA (SV-OOA), were analyzed using a least square approach. Whereas the average size distribution of LV-OOA was similar to that of sulfate both on non-event and NPF event days, SV-OOA distributed in the size range that is ~100 nm smaller than that of LV-OOA. Possible explanations include the enrichment of SV-OOA in the smaller size range as a result of gas-to-particle conversion, and the smaller vacuum aerodynamic diameter of externally mixed SV-OOA with relatively low density. The results from this study suggest the important role of secondary organic aerosol formation in the size distribution of organics in the forest atmosphere.

P-3-026 How Stratospheric are Stratospheric Air Intrusions?

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Key words
ozone stratospheric air intrusion water vapour

The change in atmospheric composition is partly driven by changing atmospheric dynamics, in potential relation to climate change. A prominent example is doubling of the stratospheric influence on ozone at the summit station Zugspitze (2962 m a.s.l., Garmisch-Partenkirchen, Germany) between the mid-seventies and 2000. Systematic efforts for identifying and quantifying this influence have been made since the late 1990s (e.g., H. E. Scheel in: http://www.forst.tu-muenchen.de/EXT/LST/METEO/staccato) based on data filtering ozone, relative-humidity (RH) and 7Be measurements. The stratospheric impact is much lower at the neighbouring site Wank (1780 m a.s.l.). In recent years we have refined the data filtering approach, based on 7Be and relative humidity, which will lead to a revision of the analysis. The quality of the results, however, significantly depends on the degree of mixing of the stratospheric air tongues with tropospheric air. It is important to note that the humidity measurements with our differential-absorption water-vapour lidar, carried out since 2004, show that even in very thin intrusion layers arriving at low altitudes the minimum RH mostly stays in the range between 0 and 2 %. Given the long distance between the arctic source regions and the Alps, this result suggests that free-tropospheric mixing is rather slow. It is also difficult to explain the characteristic strong drop in ozone in the subsiding stratospheric ozone layers in the time series of our ozone lidar to typically 50 to 80 ppb by mixing. We think that our observations in the Northern Alps to some extent map the vertical trace gas distributions in the lower stratosphere of the Arctic source regions. A smooth transition from tropospheric to stratospheric air in the source region is also consistent with the small to moderate drop of the Zugspitze CO mixing ratio during intrusion periods. In addition, the average CO in direct stratospheric intrusions has slightly grown since the beginning of the measurements in 1990, probably indicating increased input into the lower stratosphere from regions with growing air pollution, such as Asia.

P-3-027 Impacts of different canopy schemes on urban climate and air pollution modeling in Yangtze River Delta, China

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Key words
urban canopy layer climate air polltion WRF/CHEM YRD

WRF/Chem model is used to study the impacts of urbanization on regional climate change in the Yangtze River Delta, China, in this study. In our simulations, Sensitive experiments are designed to investigate the urban canopy impacts on climate change and atmospheric environment. The MOIDS land use data used in d03 is to represent the urbanization process. To qualify the impacts of urbanization on regional climate change, the parameterizations set in the model except for the canopy layer schemes are the same. The canopy scheme used in base case is SLAB scheme, in case 1 we use UCM scheme and in case 2 we use BEP scheme, respectively. July of 2010 is selected and the differences between these experiments are analyzed. The results show that for 2m-temperature prediction, the UCM scheme captures the trend well with the mean bias of -0.3°C, the mean bias between BEP simulation and observation is -0.9 °C. When the radiation reaches the urban canopy layer, the BEP scheme considers the interaction between the buildings, and the width of the street and building is also have significant

impacts on energy storage, the shape of the city such as the building height, will shelter the radiation from reaching to the ground. For 2m-relative-humidity simulation, the BEP scheme better predict the performance with the mean bias of 1.6%. The urban canopy significantly influences the wind speed over the urban area. The mean bias of wind speed predicted by BEP is -1.0m/s, while that predicted by UCM and SLAB scheme are 0.4m/s and 1.2m/s, respectively. The canopy layer will increase the roughness, and decrease the wind speed. The BEP scheme is well predict the performance of the inner city such as Nanjing, Hefei and Hangzhou, but under-estimate the wind speed in coast city Shanghai and Pudong sites. It maybe implicates that BEP scheme is poorer to simulate the sea breeze.

Impact of urban canopy on air pollutions is more complicated. The differences of temperature, relative humidity, wind field and boundary layer could influence the pollutants' chemical interaction and transportation. The SLAB scheme simulated primary pollutants such as SO_2 and CO, is about ~20ppb and 400ppb, respectively. But the difference between the BEP and SLAB case, UCM and SLAB case, is rare, the emission of SO_2 sharply decrease due to the control strategy. The CO concentration of BEP-SLAB is similar to that of UCM-SLAB, the magnitude is ~300ppb, and the high concentration center locates in Shanghai city. It may be due to the emission of CO is larger in Shanghai, and the wind speed significantly decrease that is not conducive to diffusion. The SLAB case predicted PM_{10} and $PM_{2.5}$ concentration is ~100 ug/m^3 and ~80 ug/m^3 over the urban area, the differences of PM_{10} concentrations in both BEP-SLAB and UCM-SLAB are ~-10 ug/m^3 in Nanjing and ~10 ug/m^3 in Shanghai city. The simulated RH2 in the urban area in both BEP and UCM case are higher than that in SLAB case. The high RH2 and low wind speed in the urban area may be the main cause of this phenomenon.

In this study, we primary investigate the urban canopy impacts on climate and air pollution using different canopy schemes, and make some reasonable results. In future study, temporal and special distributions of anthropogenic heat and surface albedo should be considered to investigate the influence of urbanization on climate change.

P-3-028 Evaluation of Black Carbon Aerosol from Energy Combustion on Its Radiative Forcing Effect

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Key words
Black carbon Mixing state Mass absorption coefficient Radiative forcing

Black carbon (BC), acting as the main light absorbing aerosols in atmosphere, it can change the global radiative forcing in many ways and contribute to the brownish color in the sky. It had been estimated that next to Carbon Dioxide, BC is potentially the second major contributor to the observed twentieth century global warming[Jacobson, 2001]. Because of their short lifetimes (around 1-4 weeks), BC has strong warming effect regionally, particularly large over Asia, Africa and the Arctic [Ramanathan, 2007]. The reduction in its emissions can result in rapid climate benefits due to its short lifetime and strong climate impact.

Optical properties of BC particles have been the research focus for decade because of its global and regional climate impact. The great uncertainties in the estimation of radiative forcing are still difficult to minish due to its complexity. One of the main reasons is the large uncertainty (factor of 2 or more) in the current estimates of the emission of the organic and elemental carbon [Bond et al., 2007]. In addition, the mixing state of BC particles affects the light absorption intensity significantly [Jacobson, 2001], which leads to the change of absorption property in visible spectrum. So far the quantitative measurement of soot light absorption is also still a challenge. The traditional filter based methods (e. g., Aethalometer) would have many measurement artifacts due to particle deposition and concentration on filters; In situ methods (e. g., Single Particle Soot Spectrometer) measure particles in their natural suspended state but it still has to take the conversion between the BC mass concentration and light absorption, which would also have error since the conversion coefficient is not constant.

In our research, we used Single Particle Soot Spectrometer and Photo Acoustic Soot Spectrometer with three wavelengths to measure the BC particles concentration and absorption coefficient simultaneously with the same $PM_{2.5}$ µm size cutter. The purpose of this study is to find the relationship between the mixing state and mass absorption coefficient, and analyze the absorption property in different wavelengths. Our results indicate that the average internal mixing state ratio is 24.2%, the mass absorption coefficient has the same diurnal variation as the internal mixing state ratio. If we use the constant coefficient to calculate the absorption value, the error will be up to ±50%. That will significantly affect the estimation of radiative forcing.

P-3-029 Regional data assimilation of multi-spectral MOPITT observations of CO over North America

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Key words
inverse modeling carbon monoxide

We assimilate new multi-spectral retrievals of CO from the Measurement of Pollution in the Troposphere (MOPITT) instrument using the GEOS-Chem model to constrain North American CO emissions. The multi-spectral MOPITT retrievals (version v5) provide greater sensitivity to lower tropospheric CO over land and can capture pollution signals in the planetary boundary layer. We use the nested version of the GEOS-Chem model to conduct a high-resolution (0.5x0.67) inversion analysis over North America from March 2006 to April 2007 to quantify urban and regional CO emissions and their seasonal variations. We also use emission estimates of isoprene, inferred from formaldehyde (HCHO) data from the Ozone Monitoring Instrument (OMI), to help independently quantify the anthropogenic and biogenic sources of CO in the inversion. The MOPITT inversion results are validated using independent in situ surface and aircraft observations over North America.

P-3-030 Lagrangian Backward Dispersion Modeling and its aplication in understanding seasonal, inter-annual and trend of ozone in South China

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Key words
Ozone Monsoon Long-range Transport

Tropospheric ozone (O_3) plays important roles in air quality, atmospheric chemistry and climate change. In the past decades, China, especially its eastern and southern part, has undergone rapid urbanization and industrialization, which caused sharp increase of pollutant emissions and could have changed chemical compositions of the atmosphere (including O_3). To document the change, long-term measurements of O_3 and carbon monoxide (CO), an O_3 precursor and tracer of combustion source, were carried out at the Hok Tsui regional background station in Hong Kong since 1994. Previous study (Wang et al., 2009@ACP) reported a strong positive

trend of O_3 recorded during 1994-2007 at the site. In this study, we present some preliminary results from a Lagrangian backward dispersion modeling, which was conducted to improve the understanding of roles of Asian

Monsoon on the seasonal, inter-annual and long-term trend of O_3 in South China.

P-3-031 Impacts of climate change and ozone recovery on the tropospheric oxidizing capacity

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Key words
ozone recovery and climate change oxidizing capacity

Ozone recovery and climate change are both anticipated to influence the composition of the troposphere, particularly its ozone burden and the oxidizing capacity. Increasingly, links between tropospheric chemistry and stratospheric changes are being recognized; these include transport of ozone-rich air into the troposphere, attenuation of UV radiation driving tropospheric chemistry by stratospheric ozone, and tropospheric meteorology and circulation and its deep coupling with stratospheric dynamics. However, to date few studies have been published that adequately account for these links, by using a whole-atmosphere chemistry mechanism. We will present results from a study using the UKCA whole-atmosphere model, assessing the consequences of climate change and stratospheric ozone recovery on tropospheric chemistry.

P-3-032 Multi-decadal variability of aerosol optical depth over China estimated from visibility measurements at ground meteorological stations

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Key words
AOD visibility

Multi-decadal variability of aerosol optical depth over China estimated from visibility measurements at ground meteorological stations

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Aerosols are important for global and regional climate change due to its direct radiative forcing and indirect effects on the hydrological cycle. The effects are subject to significant temporal and spatial variability due to the short aerosol lifetime, warranting a systematic analysis of their long-term variations. Of first concern is the variability of aerosol optical depth (AOD), a primary optical property of aerosols. Current dedicated satellite (e.g., Multi Axis Differential Optical Absorption Spectroscopy (MODIS)) and ground based (e.g., Aerosol Robotic Network (AERONET)) measurements of AOD are limited by temporal and/or spatial coverage, not allowing for a multi-decadal analysis. Long-term surface visibility measurements at a large number of ground meteorological stations, albeit subject to large uncertainties, provide a unique opportunity to study multi-decadal variations of AOD and its climate importance. As a first step, this study proposes a novel method to estimate AOD over China based on visibility data from hundreds of ground stations. The widely used chemical transport model (CTM) GEOS-Chem is used to provide temporally and spatially varying profiles of aerosol extinction coefficient for conversion from visibility to AOD. Results for 2006 are evaluated with data from the China Aerosol Remote Sensing Network (CARSNET), the Aerosol Robotic Net-work (AERONET), and MODIS. Reasonable consistency is found between visibility-deduced AOD and CARSNET and AERONET in regions where the ground stations are close in distance. The preliminary analysis thus suggests the reliability of visibility-derived AOD for studying long-term variability of aerosols and its climate impacts.

P-3-033 Model Simulation of Processes of Aerosol and Gaseous Air Pollutants in Long-range Transport in East Asia

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Key words
Modeling Long-range Aerosol

Regional atmospheric air pollution affects human health as well as climate system of earth. To improve the understanding of the regional air pollution situation, two intensive monitoring campaigns including both surface, ship and aerial observation to study the distribution, transport and transformation mechanism has been conducted during the period during Dec. 9, 2010 to Dec. 17, 2010 and Mar. 15 to April 10, 2011. In this work, the CMAQ ver.4.7 model has been used to simulate the transport, transformation of air pollutants such gaseous SO_2, NO-NO_2, CO, O_3 and primary and secondary aerosol for the same periods. The chemistry of NO_x-VOCs-O_3 system in long range transport have been discussed. The aerosol characteristics and composition in long-range transport are also discussed. Furthermore, we also simulated the change of extinction of aerosol and AOD due to the chemical transformation during the long range transport. The modeling work currently uses the academic emission inventory of INTEX-B as the anthropogenic emission, while MEGAN is implemented to generate biogenic VOC emission. The comparisons of modeling results with the observation data have been conducted to validate the model and to identify the uncertainties of the model.

P-3-034 Climate system impacts associated with Kalman Filter optimized time-varying black carbon emissions

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Key words
black carbon climate change kalman filter data assimillation emissions

Black Carbon (BC) impacts the climate system by scattering and absorbing solar radiation, leading to a cooling effect at the surface and a warming effect in the atmosphere. Other primary and secondary anthropogenic aerosols, such as Organic Carbon (OC), sulfate (SO_4) and various mixtures scatter solar radiation, leading to a cooling at the surface. Thus, it is important to understand the distribution and processing of these species to better understand atmospheric and climate effects such as changes in the large-scale dynamics, temperature, and precipitation.

Present knowledge of BC emissions and mixing state are poorly understood. While large amounts of BC are emitted in urban areas, a significant amount is also emitted due to biomass burning and agricultural fires: leading to a distribution variable both geographically and temporally. Rapid growth in amount and type of emissions tends to not be well represented by present-day models, especially over regions undergoing rapid economic development, that have a high population density, and diverse sources of biomass burning and fires. Finally, these issues are compounded by the fact that many present global models use simplified aerosol-climate chemistry, physics, and couplings.

To address these issues, this work describes the results of a time-varying Kalman Filter (tvKF) to better quantify the amount, spatial, and temporal distribution of BC emissions. The tvKF is constrained using data from more than 60 AERONET

stations and more than 20 surface monitoring stations, all of which have at least 3 years of continuous data. The modeling environment used for the tvKF is based on the CAM3 GCM, coupled with two-moment, 7-mode, core-shell aerosol mode, an interactive radiation model, and an urban-scale chemical and physical process model. Previous results with this modeling setup have shown good agreement with measurements, and scientifically that quantifying both the BC mixing state and secondary formation of SO_4 are important for studying the climate response.

The optimized emissions, and underlying sensitivities computed for the tvKF runs, are then run in interactive climate mode, using the model coupled to a mixed-layer ocean model. This allows the long-term equilibrium climate impacts and their sensitivities to be quantified as a function of the optimized time-varying BC emissions. This end result included how change in aerosol amounts, mixing state, interactions with the other aerosols, and time-variation of emissions, impact the column burdens, and how these in turn impact the climate response.

The results show that the global emissions of BC are roughly twice the value commonly used in the atmospheric sciences community, and that much of this change is associated with statistically relevant increases in emissions from East, South, and South East Asia. The optimized emissions will be presented in terms of their error ranges, and underlying potential physical causes. Finally, the climate response will be explained, with the results showing that while the optimized emissions lead to small changes in regional surface temperature, they lead to more significant change in the global distribution of precipitation, with a large amount of these changes occurring over monsoon Asia.

P-3-035 Does size matter? Studies with three different versions of the UM-UKCA model.

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Key words
Atmospheric chemistry mechanisms Coupled chemistry-climate simulations UT-LS Ozone

After hundreds of man years spent on developing models of atmospheric chemistry there are still many open questions. One of the fundamental questions we must ask ourselves is how complex does our model formulation need to be to answer the questions we ask of the model?

In this presentation we compare three configurations of the UK community atmospheric chemistry and aerosols coupled climate model (UM-UKCA). A tropospheric chemistry only configuration (CheT), a stratospheric chemistry configuration (CheS) and a combined model of stratospheric and tropospheric chemistry (CheST).

A series of nearly identical model simulations are performed, with the main difference being the different representations of chemistry used in the different model configurations. These simulations are used to assess the impact of the different model configurations on the budgets of key gases in the troposphere, UTLS and stratosphere; the oxidising capacity of the troposphere and surface and tropospheric column ozone.

P-3-036 High level of pollution transported up to 5000 m asl in the Southern-Himalayas: continuous observations since 2006 at NCO-P GAW global Station

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Key words
Ozone Black carbon Biomass burning Himalaya South Asian Monsoon

In the framework of the UNEP-ABC and EvK2CNR-SHARE projects, the Nepal Climate Observatory – Pyramid (NCO-P) GAW-WMO global station is operative since March 2006 in the high Himalayas (5079 m asl). The instrumental setup allows to characterize aerosol mass and size distribution, optical properties (especially absorption coefficient, corresponding to black carbon concentration), ozone and halocarbon concentrations. In particular black carbon and ozone are considered important Short Lived Climate Forcers (SLCFs), reducing which concentration can lead to fast climate benefits.

Besides obtaining a continuous characterization of the atmospheric background, the observation programmes running at NCO-P allow to monitor the intensity and the frequency of pollution transport events at such altitude. The seasonal variation of atmospheric conditions is influenced both by the local mountain wind system, and by the large-scale Asian monsoon circulation. The annual variations of the main synoptic circulation can also modulate the diurnal cycles characterizing the local mountain weather regime.

The mean average values of ozone and equivalent BC concentration over the first five years of measurements were 48 ± 12 ppbv and 210 ± 369 ng m^{-3} (± 1 sigma).

On average BC accounted for 8% of PM_1 (particulate matter with Dp < 1 m). Concurrently, the aerosol mass concentration (PM_{10}) as derived by OPC was 5.3 ± 10.2μg m^{-3}, the coarse fraction accounting for about half of aerosol mass with a larger variability with respect to the accumulation fraction. These low concentration values indicate a large occurrence of “clean” conditions at the site. Nevertheless, acute pollution episodes with ozone and BC concentrations up to 80 ppb and 5 μg m^{-3} as well as with high values of PM_1, were regularly observed during the pre-monsoon seasons from 2006 to 2010. A different behavior has been pointed out during the pre-monsoon 2011, when lower levels of pollutant affected NCO-P observation, probably due to significantly different broad-scale meteorological conditions with respect to previous years.

In this work we will provide an overlook of the frequency and intensity of pollution transports up to NCO-P GAW-WMO global station during the first 5 years of continuous monitoring activity, also investigating the possible role of biomass burning emissions on the observed variability.

P-3-037 Aerosol radiative forcing from forest fires over Siberia

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Key words
Biomass Burning Smoke aerosol Radiative forcing

Aerosols have potentially large impacts on the climate change as they absorb and scatter the solar radiation, affecting the earth’s energy budget. Main sources of aerosols include fossil fuel combustion and biomass burning. The latter in particular has great uncertainty and hence the climatic effect of smoke aerosol has been poorly quantified. In this study, we examine the climatic effects of smoke aerosols from Siberian forest fire using a global 3-D chemical transport model (GEOS-Chem) together with the past four years of in-situ observations of aerosols at the ZOTTO tall tower in Siberia. We also evaluate two most widely used biomass burning emission inventories, Fire Locating and Modeling of Burning Emissions (FLAMBE) and Global Fire Emissions Databases version 3 (GFEDv3). We conduct the model simulations with both emission inventories and compare the results with observations at the ZOTTO site (60.8 N, 89.35 E). A comparison of the model with aerosol volume observation data from September, 2006 to January, 2010 shows that the model reproduces the observed monthly variation of aerosols concentrations including peak concentrations during fire periods. We find that the model with the FLAMBE captures episodic high smoke aerosols better than the GFEDv3 although the former overestimates the observed aerosol concentrations. So, we reduce the FLAMBE by 50% in the model and then use the optimized model results to estimate the monthly variation of radiative forcing of smoke aerosols that is extensively examined for climate implication over Siberia.

P-3-038 A dichotomy in primary marine organic aerosol-cloud-climate system

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Key words
marine aerosol organic matter cloud climate

Organic matter has been observed to significantly contribute to particulate matter in every environment including pristine remote oceans. A significant if not dominant contribution of insoluble organic matter to marine aerosol has been proved to be of biogenic origin. High time resolution measurements of marine organic matter have demonstrated a dynamic system with regular organic matter plume events occurring during summer as well as frequent open ocean particle formation events. High-time resolution measurements of primary marine organic sea-spray physico-chemical properties reveal an apparent dichotomous behavior in terms of water uptake: specifically sea-spray aerosol enriched in organic matter possesses a low hygroscopic Growth Factor (GF~1.25) while simultaneously having a cloud condensation nucleus/condensation nuclei (CCN/CN) activation efficiency of between 83% at 0.25% supersaturation and 100% at 0.75%. Simultaneous measurements of Cloud Droplet Number Concentration (CDNC) during primary organic aerosol plumes reveal CDNC concentrations of 350 cm^{-3} in newly formed marine stratocumulus cloud for boundary layer organic mass concentrations of 3-4 micro g m^{-3}. It is suggested that marine hydrogels are responsible for this dichotomous behavior which has profound impacts to aerosol-cloud-climate system along with a better understood process analysis of aerosol formation by sea-spray. A hydrophobic character of organic matter dominated aerosol in sub-saturated conditions should have significant implications for direct radiative effect while effectively forming cloud condensation nuclei should have significant contribution to indirect effect.

P-3-039 Applying Hilbert-Huang Transform to Greenhouse Gas Time Series Data

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Key words
greenhouse gas climate indice time series Hilbert-Huang Transform

With the availability of new sources of data, time series analysis has been playing an ever-increasing role in atmospheric and climate studies. Time series analysis of greenhouse gas concentrations in the atmosphere brings out different patterns of variability of these atmospheric concentrations (e.g. diurnal, synoptic, seasonal, annual, and other concealed time scales). Traditional spectral analysis is generally based on certain assumptions of the data, such as its linearity and stationary nature, for example the Fourier Transform (FT), which assumes that any time series can be decomposed into a set of linear components. When decomposing a nonlinear time series, this will often produce physically meaningless harmonics. In order to accommodate the variety of data generated by nonlinear and non-stationary processes, data analysis methods need to become adaptive.

The Hilbert-Huang transform (HHT) is a recent method for the analysis of non-stationary signals in an adaptive manner. This allows the frequencies and amplitude inherent in a time series to be evaluated. The method consists of two parts: (1) the empirical mode decomposition (EMD); and (2) the Hilbert spectral analysis. The key part of this method is contained in the first step. In contrast to FT, the EMD does not assume a time series is linear nor stationary prior to analysis, it effectively allows the data driven approach, as it is adaptively decomposed into a set of abstracted patterns (or components) called intrinsic mode functions (IMF), together with a residual component, which represents the underlying trend in the time series. When the IMFs and residual are summed together they form the original time series. Subsequently, the second step, the Hilbert transform (HT) is then applied producing a localized time-frequency spectrum and instantaneous (time-dependent) frequencies of these modes. Since atmospheric and climatic phenomena are highly non-stationary and nonlinear, HHT would potentially provide more insights into their long term records.

The HHT is carried out using greenhouse gas baseline time series data from Cape Grim, Australia and 4 NOAA stations distributed in both Northern and Southern hemispheres. The results from the analysis are presented in terms of changes in variance with time in different time scales, instantaneous frequencies, and trends. The relationships between stations in different time scales are also compared. For the monthly greenhouse gases data, almost all the stations exhibited an annual cycle and inter-annual cycles with periods approximately 3 to 20 years. Relationships between greenhouse gases and climate indices are analyzed. Work is in progress to further quantify the relationship between the IMFs of greenhouse

gases and climate indices in different locations.

P-3-040 Biomass burning emissions from Robbins Island fire, Tasmania, Australia 41ºS

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Key words
biomass burning emissions

VOCs affect climate by driving formation of tropospheric ozone and secondary organic aerosol, (SOA) while aerosol has both direct and cloud albedo climate effects (through cloud condensation nuclei, CCN). The degree of radiative forcing from aerosol and tropospheric ozone is highly uncertain. Biomass burning is an important source of VOCs and aerosols in Australia. It is therefore crucial to understand the composition of smoke plumes and the processes within plumes that govern aerosol formation.

Measurements of reactive gases and aerosol were made at the Cape Grim Baseline Air Pollution Station from the 10 February to the 1 March 2006, as part of the Precursors to Particles (P2P) campaign. For four days the station was impacted by a biomass burning plume from a fire on Robbins Island 20km east of the station. This fire gave a unique opportunity to examine the affect of the smoke plume on the composition of the air in this otherwise very clean marine boundary layer environment.

P-3-041 Eastern Pacific Emitted Aerosol Cloud Experiment (E-PEACE)

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Key words
aerosol-cloud interactions particle activation droplet composition

Aerosol-cloud-radiation interactions are widely held to be the largest single source of uncertainty in climate model projections of future radiative forcing due to increasing anthropogenic emissions. The underlying causes of this uncertainty among modeled predictions of climate are the gaps in our fundamental understanding of cloud processes. There has been significant progress with both observations and models on addressing these important questions, but quantifying them correctly is nontrivial and limits our ability to represent them in global climate models. The Eastern Pacific Emitted Aerosol Cloud Experiment (E-PEACE) 2011 was a targeted aircraft campaign with embedded modeling studies, using the CIRPAS Twin Otter aircraft and the Research Vessel Point Sur in July and August 2011 off the coast of Monterey, California, with a full payload of instruments to measure particle and cloud number, mass, composition, and water uptake distributions. E-PEACE included (a) using three emitted particle sources to separate particle-induced feedbacks from natural variability, namely (i) combustion particles from container ships with dry diameters between 50 and 100 nm, (ii) shipboard smoke-generated particles with dry diameters between 100 nm and 1 μm, and (iii) aircraft-based milled, coated salt particles with dry diameters between 3 and 5 μm; (b) comparing large eddy simulations and aerosol-cloud parcel modeling studies to measured plume dispersion and interactions to form the basis for interpreting the aircraft observations; and (c) analyzing satellite images of the effects of these emitted particles to quantify warm cloud microphysics.

P-3-042 Modification of physiochemical characteristics of dust particles in spring of 2010

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Key words
dry deposition individual particle ESEM-EDX morphology elemental composition

In order to investigate the interaction between dust particle and continental air pollution, dry deposition of dust particles were collected during the dust storm in Match, 2010 in Qingdao. Morphologies and elemental compositions of individual particles were analyzed using an environmental scanning electron microscope coupled with an energy dispersive X-ray spectrometer (ESEM-EDX). Diameters of the detected particles were mainly in the range of 0.2-20 μm, and the medium diameter was around 0.5 μm, 1.5 μm and 1.0 μm in the pre-, during and post-dust storm period. The aspect ratio showed that the particles almost fall in the range of 1.0-1.5, with the frequency of 73%, 64%, and 68% in the three periods. The modes of circularity factor were around 0.7 and 0.9 for the particles in the pre-dust storm and post-dust storm, respectively. The particles during the dust storm mainly distributed in the range of 0.65-0.95. Those result indicated that the particles were more irregular during the dust storm. Al and Si were the main elemental compositions in the analyzed particles. The detected frequency of Al and Si was much lower in the pre-dust storm because of the mixing of dust particles and the local existence particles. The Al-Si-Ca triangle diagrams demonstrated that the Ca weight ratio increased with the approach of the dust storm, and then decreased with the departure of the dust storm. Fe element did not exhibit obvious variations during the dust storm episode. The sulfuration of the dust particles was higher in the pre-dust storm than in other periods implying that the physiochemical characteristics of dust particles modified due to interaction with anthropogenic air pollution along dust storm approach.

P-3-043 Deep Convective Clouds and Chemistry over the Central U.S.

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Key words
clouds and chemistry upper troposphere composition

The Deep Convective Clouds and Chemistry (DC3) field campaign is taking place in May and June 2012. Its goals are to quantify 1) storm dynamical, physical, and electrical characteristics, as well as convective transport of chemical constituents in active thunderstorms, and 2) changes in chemistry and composition of the upper troposphere (UT) after active convection. DC3 aircraft, based in Salina, Kansas, are sampling storms in three regions, northern Alabama, west Texas to central Oklahoma, and northeast Colorado, allowing the DC3 team to contrast storms and airmass composition in environments from remote continental to moderately polluted air quality scenarios.

To examine the change in UT convective outflow composition 12-30 hours after the air has been lofted from the boundary layer, high-resolution Weather Research and Forecasting (WRF) model forecasts with boundary layer (BL) tracers from each sampling region were conducted for the 2011 DC3 dry run and are being conducted during the 2012 field campaign. Simulations from the 2011 DC3 dry run showed that BL air from northeast Colorado mostly traveled north and eastward to the Mississippi River Valley and Midwest, 1000 km or more from northeast Colorado. The BL tracer from Oklahoma was transported in a variety of directions, sometimes quite widespread (from the Great Lakes to the Gulf of Mexico), reaching distances of 1000-1700 km from southwest Oklahoma. The BL tracer from Alabama was transported to the east or northeast (1000-1400 km downwind of northern Alabama) when convection was more associated with fronts or to the Gulf of Mexico (~1000 km downwind) when airmass thunderstorms were present. This presentation will include results from both the 2011 DC3 dry run and the 2012 DC3 field campaign.

P-3-044 The Stratospheric Contribution to Tropospheric Ozone Variability: Observational Constraints and Impacts of a Changing Climate

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Key words
tropospheric ozone chemistry-climate

Tropospheric ozone, as both a pollutant and a regionally-varying greenhouse gas, is at the nexus of atmospheric chemistry and climate change. Understanding the natural variability of ozone and how this variability will change in a changing climate is key to developing mitigation strategies to reduce ozone and improve air quality while reducing radiative forcing. We present diagnostics based on satellite observations that constrain the contribution of stratospheric ozone to tropospheric ozone variability on monthly and interannual timescales. Using chemistry-transport model output, we show that correlations in monthly and interannual anomalies in stratospheric and tropospheric ozone are a meaningful measure of the stratospheric contribution to tropospheric ozone variability. These diagnostics are readily derived from satellite observations and easily applied to chemistry-climate models, and we demonstrate our approach using output from the CAM3.5 and WACCM models to test how well they capture the spatial and temporal patterns of stratospheric influence. Furthermore, we examine changes in the stratospheric contribution to tropospheric ozone variability in a changing climate, and find that for the models we examined, the increase in the stratospheric circulation contributes to mean increases in tropospheric ozone, but changes in variability in both regions result in a decreased contribution of the stratosphere to tropospheric ozone variability.

P-3-045 Sensitivity analysis of aerosol feedbacks on chemistry and climate at urban and regional scales

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Key words
aerosols ffedbacks data assimilation

Black carbonaceous aerosol (BC) is an important component of atmospheric particulate matter because of its dual roles in health and climate impacts. In this paper a fully-coupled meteorology-chemistry-aerosol model (WRF-Chem) is used to assess the impacts of BC and other aerosols on radiative forcing, weather and atmospheric chemistry. A series of simulations from different aerosol - cloud environments ranging from heavily polluted cities in East (Beijing) and South (Delhi) Asia to relatively clean coastal areas with permanent cloud cover (VOCALS experiment off the west coast of Chile/Peru) are simulated. The results are further analyzed using adjoint sensitivities of meteorological and cloud parameters calculated using a new adjoint model of WRF-Chem. We also present new results testing a data assimilation technique to constrain modeled aerosol concentrations using satellite retrievals of cloud optical depth (COD) and liquid water path (LWP).

P-3-046 Estimating global-annual aerosol indirect effects in global climate model simulations by controlling for meteorology to reduce the impact of natural variability.

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Key words
aerosol clouds climate indirect

Although greenhouse gas emissions constitute the largest component of the anthropogenic radiative forcing, aerosol particle emissions represent the largest sources of uncertainty. Unlike greenhouse gases that are well mixed and have easily measured radiative properties, aerosol particles are highly variable in space and time, have varied radiative properties, and have complex interactions with cloud microphysics. Aerosol particle concentrations are strongly impacted by natural modes of variability on many timescales that influence not only their circulation, but also cloud processes important to their production and removal. As a result, long simulations are typically required to statistically isolate the anthropogenic aerosol forcing in pairwise climate model experiments with and without pollution. In this case, the two simulations not only have different emissions, but also produce unique weather patterns and may be in different large-scale climate states. The largest source of noise in estimates of aerosol indirect effect is variability in the liquid water path (LWP), such that statistically significant differences due to aerosol perturbations can only be detected by integrating over the dominant modes that influence LWP natural variability. Here we present results from an alternate approach, which implements Newtonian relaxation ("nudging") to constrain the two simulations toward observations with identical meteorology, thus reducing differences in natural variability and dampening feedback responses, to isolate the calculation of radiative forcing. This provides a more stable global estimate of the aerosol indirect radiative forcing on shorter time scales and increases the statistical significance of the signal over large parts of the world. The approach is suggested as a strategy to bring the enhanced physics of new computationally intensive aerosol enabled climate simulation technology, for which long simulations remain prohibitive, to bear on the aerosol indirect effect problem.

P-3-047 The relevance of column integrated measurements in the Baltimore-Washington area during DISCOVER-AQ.

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Key words
in situ measurements aerosol optical properties

Satellite measurements of aerosol and trace gas species represent an important tool in expanding the global observational dataset. These measurements are often challenged near urban areas by complex spatial and vertical structures arising from the combination of intense local emissions sources and long-range pollution transport. In-situ aircraft observations provide a detailed picture of vertical aerosol structure, and are thus essential for bridging the gap between satellite observations and ground based sensors. We present detailed measurements of aerosol optical, chemical, and microphysical properties measured aboard the NASA P-3B aircraft in the Baltimore-Washington area during the July 2011 DISCOVER-AQ (Deriving Information on Surface Conditions from Column and Vertically Resolved Observations Relevant to Air Quality) mission. Over two-hundred aircraft spiral ascents and descents were performed in the vicinity of six ground sites equipped with Aerosol Robotic Network (AERONET) sun photometers. The payload of the P-3B included a comprehensive suite of aerosol instrumentation including humidified and dry TSI 3-wavelength integrating nephelometers, a 3-wavelength Radiance Research Particle Soot Absorption Photometer (PSAP), a custom scanning mobility particle sizer (SMPS), a Droplet Measurement Technologies Ultrahigh Sensitivity Aerosol Spectrometer (UHSAS), and a TSI Aerodynamic Particle Sizer (APS). The relevance of column-integrated P-3B observations of aerosol single scattering albedo, Angstrom exponent and size distributions are evaluated. The implications for remote sensing retrievals will be discussed.

P-3-048 Observations of Regional-Scale Impacts on Cloud Microphysics from Anthropogenic Aerosols and Warm Sea Surface Temperature over the East China Sea

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Key words

aerosol cloud

Cloud microphysical properties and aerosol concentration were measured aboard an aircraft over the East China Sea and Yellow Sea in April 2009 during the Aerosol Radiative Forcing in East Asia (A-FORCE) experiment. We sampled stratocumulus and shallow cumulus clouds over the ocean in 9 cases during 7 flights 500–900 km off the east coast of Mainland China. In this study we report aerosol impacts on cloud microphysical properties by focusing on regional characteristics of two key parameters, namely updraft velocity and aerosol size distribution. First, we show that the cloud droplet number concentration (highest 5%, Nc_max) correlate well with the accumulation-mode aerosol number concentration (Na) below the clouds. We then show that Nc_max correlates partly with near-surface stratification evaluated as the difference between the sea surface temperature (SST) and 950-hPa temperature (SST – T950), likely due to the warm SST induced greater updraft velocity. Second, we show that not only higher aerosol loading in terms of total aerosol number concentrations (D > 10 nm) but also larger aerosol mode diameters are likely to contribute to high Nc during A-FORCE. A crude estimate of an aerosol induced cloud albedo radiative forcing is also given.

P-3-049 Chemical characterization of submicron aerosol with an Aerosol Chemical Speciation Monitor in Beijing, China

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Key words
ACSM particulate matter sources Beijing

Both satellite and ground measurement based studies indicate that Beijing is a city with significant particulate matter pollution. Since December 2011, an Aerodyne Aerosol Chemical Speciation Monitor (ACSM) was deployed in urban Beijing to characterize chemical compositions of non-refractory submicron particles

(NR-PM_1) at 15 min resolution. During four month measurement, the mean NR-PM_1 was found to be 36.7±47.4 μg/m^3, consisting of 39% organics, 22% ammonium, 19% nitrate, 15% sulfate, and 5% chloride. NR-PM_1 shows high concentration from evening to midnight and low concentration during daytime. Ammonium and nitrate have similar diurnal cycles. However, a continuous increase of ammonium and nitrate concentrations during the daytime was observed. This suggests that the formation of NH_4NO_3 overwhelms its evaporation into gaseous state and the dilution by the higher planetary boundary layer (PBL) during daytime. Multivariate analysis of the ACSM organic spectra with Positive Matrix Factorization (PMF) identified a hydrocarbon-like organic aerosol (HOA) and an oxygenated organic aerosol (OOA). The HOA component likely corresponds to primary OA material and shows a large increase during traffic and meal times, while the OOA component likely corresponds to secondary OA material and correlates well with the sum of sulfate and nitrate, two well-known secondary aerosol species. The NR-PM_1 concentration and composition observed during this study are highly variable. Its mass concentration increases with high humidity, low wind speed, and drops sharply with high wind speed or precipitant. Back trajectory analysis indicates that the variability also correlates with the change of air masses from different region, e.g., air masses from the south are associated with high PM_1 pollution. A close correlation between the NR-PM_1 measured by the ACSM and the $PM_{2.5}$ measured by a Tapered Element Oscillating Microbalance (TEOM) suggests that NP-PM_1 is the major source of $PM_{2.5}$ in Beijing.

P-3-050 Two-way coupling of global and regional chemical transport modeling

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Key words
Two-way Geos-Chem

Two-way coupling of global and regional chemical transport models (CTMs) is critical for analyzing the impacts of regional air pollution on the global environment, by exploiting results from high-resolution regional modeling to improve the coarse-resolution global simulation. Such coupling capability has not been well developed in current research due in part to the embedded complexity and computational cost. Here a novel approach is proposed to develop two-way coupling between the widely used global GEOS-Chem CTM (5° long×4° lat or 2.5° long×2° lat) and its nested version (0.667° long×0.5° lat). The approach allows for reduced complexity and computational cost while maintaining the accuracy of the coupling. Our preliminary analysis over Asia and North America suggests varying importance of two-way coupling for global transport of a number of air pollutants. Large differences are found also for regional modeling results between two-way and one-way coupling. For example, differences in black carbon (BC) concentrations between two-way and one-way regional simulations can reach 10% in some regions in the free troposphere. It is thus evident that two-way coupling is important for both global and regional simulations.

P-3-051 Chemical characteristics of PM_{10} over Delhi in the Indo-Gangetic Basin: Quantification of the impacts of anthropogenic aerosols

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Key words
PM_{10} Anthropogenic aerosols Indo-Gangetic Basin Radiative forcing

The present work is aimed to quantify optical and radiative impacts of anthropogenic aerosols over Delhi- one of the highly polluted urban megacities in the Indo-Gangetic Basin (IGB) region in India, using detailed chemical analysis of surface measured PM_{10} aerosol samples during the year 2007. The annual mean PM_{10} mass concentration was found to be about 219 (±84) μgm^{-3}, which is about twice to the prescribed Indian National Ambient Air Quality Standards values. The analyzed PM_{10} samples were found to be mainly composed of secondary inorganic aerosol species (~16 μgm^{-3}, 13%), mineral matters (~12 μgm^{-3}, 10%) and salt particles (~5 μgm^{-3}, 4%). However, the remaining part of PM_{10} is deemed to consist of carbonaceous aerosols such as black carbon and organic carbon, ammonium and other insoluble elements. Black carbon mass concentrations were measured concurrently from the Aethalometer, and the annual mean concentration was observed to be ~14 (±12) μgm^{-3} over the station during the study period, which is significantly large compared to those, observed at other locations in India.

An optically equivalent aerosol model was formulated based on the measured aerosol chemical compositions, black carbon and the ambient meteorological parameters to derive radiatively important aerosol optical parameters. The derived aerosol parameters were then used to estimate the aerosol direct radiative forcing over the station. The anthropogenic components were found to be contributing ~72% to the composite or total aerosol optical depth (AOD500 ~0.84) over Delhi. The estimated mean surface and atmospheric forcing for composite aerosols were found to be about −69, −85 and −78 Wm^{-2} and about +78, +98 and +79 Wm^{-2} during the winter, summer and post-monsoon periods, respectively. The

anthropogenic aerosols contribute ~90%, 53% and 84% to the total aerosol surface forcing and ~93%, 54% and 88% to the total aerosol atmospheric forcing during the respective periods. The annual mean surface and atmospheric forcing for composite aerosols were about −79 (±15) and +87 (±26) Wm^{-2} over Delhi, with respective anthropogenic contributions of ~71% and 75%. Aerosol induced large surface cooling, which was relatively higher during the summer as compared to the winter, suggesting an increase in dust loading over the station. The resultant atmospheric heating rate was also calculated over the station and found to be of the order of more than 2 $Kday^{-1}$ during all the seasons, which is quite significant and raised several climatic issues.

P-3-052 Intercontinental transport and climatic impact of saharan and sahelian dust

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Key words
Dust transport climate RegCM

Sahara and Sahel are important emission sources of dust particles in the atmosphere. Particles are transported through the Atlantic Ocean and even detected off the Eastern American Coasts. This transport varies temporally and spatially and often reaches its peak during the boreal summer (June-July-August). The Regional climate model RegCM 4.0 contains a dust module that takes into account their emission, transport and deposition processes. Saharan and sahelian dusts' emissions, transport and climatic impact on precipitations during the spring "March-April-May" and summer "June, July, August" were studied using this model. The results showed that simulated data were coherent with observations made by the MISR satellite, AERONET ground stations within the domain; Africa (Banizoumba, Cinzana and M'Bour) and Ragged-point (Barbados islands). The transport of dust particles occurred from North-East to South-West over the targeted period (2005-2010). The seasonality of dust plumes' trajectories was strongly linked to that of altitudes reached in the troposphere. The impact of dusts on climate consisted of a cooling effect both during the boreal summer and spring in West Africa (except South-Guinea and North-Liberia), Central Africa, South-America and Caribbean where increased precipitations were observed.

P-3-053 New particle formation in the Yangtze River Delta

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Key words
aerosols nucleation

Introduction. Aerosols are a growing public health concern, and they play a central part in global climate. It is important to point out the duality of high ambient aerosol concentration: they have adverse health effects (mainly lungs and heart), and they contribute to cool the atmosphere which in the light of global warming is rather positive. Simply removing aerosols from the atmosphere in the name of public health is thus a dangerous approach as long as the role of aerosols is not understood in full detail. One of the main sources of ambient aerosol particles is atmospheric new particle formation (nucleation) which has been observed all over the world but which remains incompletely understood. In order to better understand atmospheric nucleation, long-term aerosol measurements are needed, with as much supporting data as possible and from a wide range of representative environments.

Measurements. The Nanjing Xianlin measurement station is located in the Yangtze River Delta, probably the biggest agglomeration of megacities in the world and a global pollution hotspot. It is designed to measure aerosols, trace gases, radiation, and a variety of meteorological data continuously in order to understand atmospheric processes such as nucleation. The core of the aerosol measurements is formed by a DMPS (differential mobility particle sizer, built at Helsinki University) and an AIS (air ion spectrometer, Airel Ltd, Estonia) which measure aerosols from 6 to 800 nm and air ions from 0.8 to 48 nm respectively. Combining these devices, it is possible to determine nucleation frequency, annual cycle, particle growth rates, formation rates, vapor concentrations and vapor source rates. Besides DMPS and AIS, nucleation-relevant measurements include PM 2.5, ozone, radiation, and sulfur dioxide.

Results. Aerosol measurements began in November 2011. From November 17 to April 17 over 30 new particle formation events were observed. Growth rates of newly formed particles vary between 3 and 15 nm/h with a mean of about 6.5 nm/h. Single events have been observed continuously for up to nine hours, on average a formation event produces around 20000 new particles per ccm, with maximum values well over 100000. Based on wind speed, event duration, and homogeneity of the observed event, we can conclude that nucleation happens simultaneously in areas extending even beyond 100 km. As observed in other location as different as Finnish Hyytiälä or Paris, also Xianlin data suggests the typical "winter break" with almost no nucleation occurring in November and December and increasing activity from January into spring. A special if only temporary feature of the measuring site is the surrounding construction ground of the new university campus. Pollution caused by the site (mostly dust, but probably also a considerable contribution from an armada of ancient combustion engines) seems to suppress local nucleation almost completely. This can be concluded from a small gap between the ion cluster band at 1.5 nm and the observed onset of particle formation.

Outlook. The next steps include gathering data to establish an annual aerosol characteristics cycle and later on a comparison to the post-construction situation.

P-3-054 A tropospheric and stratospheric ozone climatology from global ozone

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Key words
ozone climatology stratospheric ozone tropospheric ozone

A three-dimensional (latitude, longitude, altitude) climatology of tropospheric and stratospheric ozone is derived from the global ozonesonde sounding record by a domain-filling trajectory mapping method. The trajectory technique provides a powerful tool to integrate sparse ozonesonde measurements, by filling in the spatial domain. This is possible because the lifetime of ozone throughout much of the atmoosphere is of the order of weeks. This physically-based trajectory mapping method offers evident advantages over typical statistical interpolation methods. The objectives are to create an ozone climatology for model and satellite retrieval a priori, trend analysis, tropospheric-stratospheric exchange research, and ozone-climate interaction studies.

We employ 51,898 ozone soundings at 116 stations over 44 years (1965-2008) from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC). Forward and back- trajectories are performed for 4 days each from each sounding, driven by NCEP reanalysis data using the HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory Model) from the NOAA Air Resources Laboratory. The resulting global ozone climatology is archived for five decades from the 1960s to 2000s with a grid size of 5 by 5 degrees and 1 kilometer vertically.

The trajectory-mapped ozone profiles show good agreement with independent profiles from SAGE and MOZAIC, and the patterns produced separately by forward and backward trajectory calculations are similar. This climatology dataset is further validated at selected stations by

comparing the actual ozone sounding profile with that found through the trajectory technique, using the ozone soundings at all the stations except one being tested. The two sets of profiles are in reasonable agreement with a correlation coefficient of 0.99 and mean differences less than 5% in the lower stratosphere between 20

and 25 km. Maximum differences are about 20%.

This ozone climatology is independent of any photochemical model. In the troposphere, features such as the continental outflow from North America, the summer ozone buildup over the Middle East, and biomass burning in the southern hemisphere, are clearly visible. In the stratosphere, it reveals strong longitudinal variation in ozone and covers higher latitudes than current satellite data. Variability in ozone on seasonal and decadal scales will be discussed.

P-3-055 BC in air and snow and the effect on snow albedo – measurements from three snow seasons in Changbai Mountain

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Key words
black carbon snow albedo

Black carbon (BC) atmospheric particles originate from incomplete combustion of fossil fuel and biomass. When deposited on the surface, even relatively small amounts of BC can reduce the snow albedo. However, the lack of observations and poor process understanding makes estimates of the climate impact highly uncertain.

We have conducted measurements of semi-continuous atmospheric BC concentrations, weekly snow BC concentrations, snow spectral albedo as well as snow physical observations at CAS Research Station of Changbai Mountain Forest Ecosystem in North East China during three snow seasons 2009/10-2011/12. The relation between BC concentration in air, snow and the meteorological variables gives us insight into the processes about how BC is embedded in the snow, while the concurrent spectral measurements of the snow albedo serves for finding a direct relation of the change in albedo to the levels of BC present in the snow.

Our measurements show elemental carbon (proxy for BC) concentrations for surface snow in Changbai station to be on average 464 ppbw (range 7-1512) ppbw during the three snow seasons. The concentrations in snow were highly correlated

with the amount of precipitation, and the different snow seasons show very different characteristics due to the different precipitation amount. For the snow season 2010/11, which was very dry, we see a steady increase in surface BC concentration during the months December-January with hardly any snowfall. From our measurements BC atmospheric deposition fluxes have been estimated. For our station, the wet and dry deposition fluxes are of equally importance, indication that also dry deposition plays an important role in this area.

We observe that the snow spectral albedo is strongly affected by the meteorological conditions through the snow physical properties, as well as the amount of BC in snow. A one-to-one relationship between visible albedo and the amount of BC in surface snow is difficult to excerpt due to the various additional factors influencing the relationship (mixing between BC and snow, light conditions, etc.) and the obtained accuracy of the optical instrumentation. However, through radiative transfer modeling with different type of snow (fresh, old, wet and melting snow) the understanding of the processes is improved.

P-3-056 Characteristics of carbonaceous aerosol measured at a background site in Jeju, Korea

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Key words
carbonaceous aerosol organic carbon and elemental carbon

In order to better understand the characteristics of carbonaceous aerosol in Northeast Asia, intensive measurements were conducted at Gosan superstation in Jeju island, Korea in summer and fall months of 2009, 2010, and 2011. During the intensive measurement periods, semi continuous organic carbon (OC), elemental carbon (EC) and highly time-resolved water soluble organic carbon (WSOC) were measured using a Sunset OC/EC analyzer and Pariticulate Into Liquid Sampler (PILS) coupled with Total Organic Carbon analyzer (TOC), respectively. Carbon monoxide and particulate matter (PM10 and PM2.5) concentrations were also

measured by a trace level carbon monoxide analyzer and a Grimm aerosol spectrometer. Air mass reaching the sampling site was classified into six types (Continental Southwest, Continental Northwest, Continental Clean, Marine, Ease Sea, and Stagnant) based on the NOAA Hysplit backward-trajectory analyses. Organic carbon and elemental carbon concentrations were measured in the ranges of 1.4 ~ 17.8 microgram carbon per cubic meters and 0.2 ~ 2.8 microgram carbon per cubic meters, respectively. High organic carbon concentration was observed occasionally due to local biomass burning and long range transport of aerosol. In September, relatively low organic carbon (2.5 microgram carbon per cubic meters) and elemental carbon (0.7 microgram carbon per cubic meters) concentrations were observed due to the increased influence of Marine air mass. The highest ratio of organic carbon to elemental carbon of 6.3 was observed during a Stagnant case in 2010 with the highest organic carbon concentration of 17.8 microgram carbon per cubic meters. Based on EC tracer method, higher contribution of secondary organic carbon to organic carbon was resulted during Continental Southwest (66.8 percent) and Stagnant (77.3 percent) cases.

P-3-057 Opportunities onboard Australia’s new research vessel, the RV Investigator

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Key words
marine science international collaboration

In May 2009 the Australian Government allocated $120 million for a new ocean-going research vessel to replace the current Marine National Facility Research Vessel, the 66-metre Southern Surveyor. The new vessel, the RV Investigator, will be owned and managed by CSIRO Australia. Its operation will be guided by an independent steering committee and funded by the Australian Government to support voyages mounted by Australian scientists and their overseas collaborators. Construction is underway with completion expected mid 2013.

The RV Investigator will operate from the tropical north to the Antarctic ice-edge and across the Indian, Southern, and Pacific oceans, greatly improving Australia’s capacity to investigate and understand marine geological processes, detect and predict changes in the ocean environment and their implications for weather and climate; and characterise and manage marine ecosystems, biodiversity and fisheries.

The RV Investigator will have a two dedicated laboratories for atmospheric research; an aerosol laboratory located at the bow of the ship and an air chemistry laboratory on the foredeck. Air will be drawn into the laboratories via an aerosol sampling mast with an inlet located 24 m above the sea surface, which point into the direction of the wind (Bates et al. 2002). A second mast will be available on the bow of the ship for the measurement of sea-air fluxes and a weather radar will compliment the range of detailed meteorological observations that will be routinely made. There will also be sites for two containers on the foredeck to provide laboratory space for intensive measurement campaigns. Atmospheric research on board the RV Investigator will take the form of both “routine” climate tracking observations and mission driven campaign experiments for which expressions of interest (EOI) and proposals will be sought.

The RV Investigator offers the scientific community opportunity to sample, observe and measure the atmosphere in one of the most under sampled areas of the world. The Marine National Facility Steering Committee calls for applications for sea time annually and encourages applications for national and international collaborative projects. Please visit the poster or the RV Investigator web site for more information.

P-3-058 Improving global aerosol forecasts by integrating Sun-synchronous and geo-stationary AOD observations

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Key words
aerosols, air quality forecasts data assimilation

The GEM-MACH Global is a unified global air quality forecasting model developed at Environment Canada by implementing tropospheric chemistry and aerosol processes on-line into the Global Environmental Multiscale weather prediction model – GEM. Due to the multi-scale features of the GEM, the integrated model, GEM-AQ/EC, is able to investigate chemical weather at scales from global to urban domains. However, because of the uncertainties in the global emission inventories, especially the timely emissions for the near real time forecasts, the model results are sometimes not agreed with surface observations very well. This research is to develop a data assimilation system that applies the satellite observation of aerosol optical depth (AOD) for the GEM-MACH to improve the forecast accuracy. The results will focus on the North America where both Sun-synchronous and geo-stationary AOD observations are available. Since the Sun-synchronous observation can only provide a data pass once a day at the same time, some systematic biases are retained in the modeling system. By using the geo-stationary AOD observations with the Sun-synchronous observations, such bias is reduced and the forecasting results are improved. This talk will present the development and preliminary results on the improving AQ forecasts in North America.

P-3-059 Sensitivity of Cloud Droplet Number to Uncertainties in Cloud Condensation Nuclei

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Key words

Global Model Cloud Droplet Cloud Condensation Nuclei Uncertainty Field Measurements

Aerosols, acting as cloud condensation nuclei (CCN) to form clouds, are known to be a significant but highly uncertain contributor to the Earth's climate. Current large-scale models almost exclusively use Kohler theory to predict CCN concentrations from aerosol size, chemical composition, and ambient water vapor supersaturation; however, given limited computational resources, models must make simplifying assumptions that introduce error into their application of Kohler theory. Consequently, a number of past field studies have attempted to quantify this error by comparing measured CCN concentrations to those predicted from simplified theory. Yet, this uncertainty represents only one link in our understanding of the aerosol-cloud climate impacts. The second link is the combination of CCN concentrations with cloud dynamics to determine the overall cloud droplet number concentration (CDNC). Limited attention to date has focused on extrapolating these CCN uncertainty studies to evaluate their impact on the overall CDNC uncertainty, and hence, climate.

We present results from a combined experimental and modeling study incorporating field data of aerosol size, chemical composition, and CCN activity measured in diverse environments throughout North America, including the Alaskan Arctic, agricultural areas, and urban centers in the southeastern and southwestern United States. First, the adjoint of a CDNC parameterization, driven by aerosol populations and updraft velocities obtained from a year-long execution of the NASA GMI Chemical Transport Model, is used to determine the regional sensitivity of CDNC to CCN concentration. Then, these sensitivities are used to extrapolate the field-measurement-derived CCN uncertainties to the overall CDNC uncertainty associated with simplified Kohler theory predictions. It is found that this uncertainty is much lower over polluted continental regions than in more pristine environments. Given the past focus on continental environments, this finding motivates future measurements in these pristine regions in order to quantify the impact of and uncertainty associated with transient processes in effecting changes in cloud properties.

P-3-060 Use the Aura TES and TCCON measurements to estimate the boundary layer CO_2 from free tropospheric and column CO_2

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Key words
boundary layer CO_2 TCCON TES

Our ability to infer surface carbon fluxes depends critically on interpreting spatial and temporal variations of atmospheric CO_2 and relating them back to surface fluxes. Although total column measurements will be useful for estimating large regional fluxes, the uncertainties in model transport remains a significant error source, particularly for local sources and sinks. To improve the capability of estimating regional fluxes, we estimate CO¯2 partial column averages within boundary layer by subtracting free tropospheric CO_2 by space based thermal IR measurements (TIR) from total column by ground-based near infrared (NIR) measurements. The NIR measurements are obtained from the Total Carbon Column Observing Network (TCCON) solar measurements, which provide an estimate of the total CO_2 atmospheric column amount. Estimates of tropospheric CO_2 that are co-located with TCCON are obtained by assimilating Tropospheric Emission Spectrometer (TES) free-tropospheric CO_2 estimates into the GEOS-Chem model.

We find that the calculated random uncertainties due to noise and temperature variability for the total column and boundary layer estimates are consistent with actual uncertainties as compared to aircraft data. For the total column estimates the random uncertainty is about 0.67 ppm with a bias of -5.71 ppm, consistent with previously published results. After accounting for the total column bias, the bias error of the boundary layer CO_2 estimates is 0.32 ppm with a precision of 1.44 ppm.

This work shows that total column from NIR measurements, e.g. TCCON, Greenhouse gases Observing SATellite "IBUKI" (GOSAT), and Orbiting Carbon Observatory (OCO-2), as well as the free troposphere measurement from TIR, e.g. TES and Atmospheric Infrared Sounder (AIRS) can be used to profile CO_2 and obtain boundary layer CO_2 with precision necessary to capture the atmospheric

CO_2 variability.

P-3-061 Methane and Carbon Monoxide Enhancements in Tropical Fire Smoke Plumes during the 2006 El Niño as observed by the Aura TES Satellite Instrument

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Key words
tropical fires methane

Tropical fires represent a significant source of methane, yet their contribution to the global methane budget is highly uncertain because fire emissions are highly variable from year-to-year and because the CH_4 emissions depend on the stage of the fire. In this paper we use new observations of CH_4 and CO in the free troposphere from the Aura Tropospheric Emission Sounder satellite instrument to place constraints on the role of fire emissions versus microbial production (e.g. wetlands and livestock) on the tropical atmospheric methane distribution during a time of significant peat fire emissions from Indonesia in the fall of 2006. We first evaluate the global CH_4 distributions from TES and GEOS-Chem and find that biases between observation and model are consistent with previous validation studies of TES data using aircraft measurements. Comparisons account for the vertical resolution and a priori constraints associated with the TES CH_4 and CO estimates. Tropospheric averages of CH_4 and CO are calculated and these observed distributions are compared to similar distributions from the GEOS-Chem model in order to quantify the total methane emissions and those from the fire.

For the Indonesia peat fires, we find that the TES and GEOS-Chem CH_4 methane distribution is consistent (within the TES observation error) and that the slope of the TES CH_4/CO distribution is consistent with the GEOS-Chem CH_4/CO distribution. This agreement between the TES observations and the GEOS-Chem model provides confidence in the total emissions used by GEOS-Chem of 4 Tg of methane during the time period of the Indonesian fires with 2.8 Tg of this total coming from the fire. The fractional error on the 2.8 Tg of methane fire emissions depends on the fractional error in the CO fire emissions. We also examine the CH_4 and CO distributions over burning regions in South America and Africa. We find that the methane and CO distributions observed by TES are inconsistent with those from GEOS-Chem. However, the slopes of the observed and modeled distributions are consistent. The values for the slopes, relative to the CH_4/CO emission factors expected for these fires, indicate that biotic emissions (e.g., wetlands and livestock) are the primary contributors to methane in these free-tropospheric smoke plumes. Sensitivity studies using the GEOS-Chem model suggest that the correlations between CH_4 and CO in these plumes are driven by near-colocation of burning with emissions from non-burning sources, followed by

transport of these emissions into the free troposphere.

P-3-062 Impact of cloud processes on aerosol particle properties: results from two ATR-42 flights in an extended stratocumulus cloud layer during the EUCAARI campaign (2008)

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Key words
aerosol-cloud interaction CCN EUCAARI

Within the European Aerosol Cloud Climate and Air Quality Interactions (EUCAARI) project, the Meteo France research aircraft ATR-42 was operated from Rotterdam airport during May 2008, to perform scientific flights dedicated to the investigation of aerosol-cloud interactions. Therein, the objective of this study is to illustrate the impact of cloud processing on the aerosol particles physical and chemical properties. The presented results are retrieved from measurements during a double-flight mission from Rotterdam (Netherlands) to Newcastle (UK) and back using data measured with compact Time of Flight Aerosol Mass Spectrometer (cToF-AMS) and Scanning Mobility Particle Sizer (SMPS). Cloud-related measurements during these flights were performed over the North Sea within as well as in close vicinity of a marine stratocumulus cloud layer. Particle physical and chemical properties observed in the close vicinity (V), below and above the stratocumulus cloud show strong differences. Firstly, measurements at constant altitude above the cloud layer show decreasing mass concentrations with decreasing horizontal distance (210–0 km) to the cloud layer by a factor up to 7, whereas below the cloud and by same means of distance, the mass concentrations merely decrease by a factor of 2 on average. Secondly, the averaged aerosol size distributions, observed above and below the cloud layer, are of bimodal character with pronounced minima between Aitken and accumulation mode which is potentially the consequence of cloud processing. Finally, the chemical composition of aerosol particles is strongly dependent on the location relative to the cloud layer (vicinity or below/above cloud). In general, the nitrate and organic fractions decrease with decreasing distance to the cloud, in the transit from cloud–free conditions towards the cloud boundaries. The decrease of nitrate

and organic compounds ranges at a factor of three to ten, affecting sulfate and ammonium compounds to be increasingly abundant in the aerosol chemical composition while approaching the cloud layer. Finally, the chemical composition of non-refractory evaporated cloud droplets measured within the cloud shows increased fractions of nitrate and organics (with respect to concentrations found below clouds), but also large amounts of sulfate, thus, related to activation of particles, made up of soluble compounds.

P-3-063 Impacts of climate change on surface ozone and intercontinental ozone pollution: A multi-model study

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Key words
surface ozone climate change emission reductions source-receptor relationships

Ozone recovery and climate change are both anticipated to influence the composition of the troposphere, particularly its ozone burden and the oxidizing capacity. Increasingly, links between tropospheric chemistry and stratospheric changes are being recognized; these include transport of ozone-rich air into the troposphere, attenuation of UV radiation driving tropospheric chemistry by stratospheric ozone, and tropospheric meteorology and circulation and its deep coupling with stratospheric dynamics. However, to date few studies have been published that adequately account for these links, by using a whole-atmosphere chemistry mechanism. We will present results from a study using the UKCA whole-atmosphere model, assessing the consequences of climate change and stratospheric ozone recovery on tropospheric chemistry.

P-3-064 On-line Analysis and Mass Concentration Characters of Ammonia in Aerosol PM_{10} in Beijing

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Key words
ammonia water-soluble diurnal variation PM_{10} Beijing

Ammonia is one of the most important components in water-soluble aerosols. The ammonia in PM_{10} measured from January 2004 to December 2005 in Beijing was determined using a system of combining rapid collection of fine particles and Ion Chromatography (RCFP-IC). The scientific research showed that the annual average concentration of water-soluble ammonia in PM_{10} in the urban area in Beijing was about 6.7（7.6±3.7）$\mu g \bullet m^{-3}$ in 2004 and 5.6（4.9±1.7）$\mu g \bullet m^{-3}$ in 2005 respectively. The concentration of ammonia in summer and autumn was higher than that in spring and winter. Trends of the statistics diurnal variation of ammonia showed different in different seasons. In summer, the higher concentration appeared at noon and the lower appeared in the afternoon. While in autumn (September), the higher concentration appeared at forenoon and the lower appeared in the afternoon. In summer and autumn, the diurnal variation of ammonia was different in 2004 and 2005. The typical diurnal variations of ammonia was analyzed in this paper, such as heating period and non-heating period, sunny day and rainy day, less polluted period and more polluted period. The relationship of ammonia and meteorological factors can be investigated using correlation analysis.

P-3-065 First results from the Airborne Tropical TRopopause Experiment (ATTREX)

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Key words
tropical tropopause layer water vapor Global Hawk platform

The NASA Airborne Tropical TRopopause EXperiment (ATTREX) is a series of airborne campaigns focused on understanding physical processes in the Tropical Tropopause Layer (TTL) and their role in atmospheric chemistry and climate. ATTREX is using the high-altitude, long-duration NASA Global Hawk Unmanned Air System to make in situ and remote-sensing measurements spanning the Pacific. A particular ATTREX emphasis is to better understand the dehydration of air as it passes through the cold tropical tropopause region. The ATTREX payload contains 12 in situ and remote sensing instruments that measure water vapor, clouds, radiation, multiple gaseous tracers (CO, CO_2, CH_4, NMHC, SF_6, CFCs, N_2O), reactive chemical compounds (O_3, BrO, NO_2), and meteorological parameters.

The first ATTREX flights were conducted out of Dryden Flight Research Center in southern California during the fall of 2011. Several future flight campaigns in 2013 and 2014 are planned. The fall ATTREX measurements provide clear evidence for two distinct classes of thin cirrus in the TTL: (1) layers with low ice concentrations and low extinctions that have little apparent effect on ice super-saturation, and (2) thin layers with much higher ice concentrations that effectively deplete any vapor in excess saturation. The ATTREX measurements, along with past high-altitude aircraft measurements, indicate that low-ice concentration TTL cirrus occur frequently in various tropical regions. These findings have implications for our understanding of the regulation of stratospheric humidity by TTL cirrus. Results from the fall 2011 campaign and plans for future ATTREX missions will be presented.

P-3-066 Characteristic of carbonaceous components in precipitation and aerosol at Japanese Monitoring Sites

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Key words
carbonaceous components precipitation

The methodology for collection and analysis of carbonaceous component in precipitation was established. By using standard black carbon particle, more than 90% of recovery efficiency was confirmed by a laboratory experiment. Field observations have been implemented at Niigata (rural), Sado (remote), and Tokyo (urban) sites in Japan since April 2011 to June 2012. The amounts of elemental carbon (EC) and organic carbon (OC) in precipitation and aerosol were measured by the IMPROVE protocol using a DRI model 2001 carbon analyzer. Seasonal variations, scavenging efficiencies, and transportation were discussed.

Concentrations of OC and EC in precipitation and aerosol at Tokyo were higher than those at Niigata and Sado. Both of the OC/EC ratios for precipitation and aerosol samples were highest at Sado and lowest at Tokyo. The importance of wet deposition of carbonaceous component was comparable to dry deposition. At all sites, the scavenging ratio of OC was higher than that of EC. The wash-out processes of OC and EC were more significant at an urban site because of high aerosol concentrations. The scavenging ratio of OC at Tokyo was two times higher than that at Sado. On the other hand, the scavenging ratio of EC at Tokyo was slightly higher than that at Sado. These observations infer that wash-out process of OC at an urban site is more important than that of EC. By back trajectories analysis, the events of high EC concentrations in precipitation during winter at

Sado were significantly affected by transportation from the Asian continent, whereas at Tokyo, it would be mainly derived from local emissions.

P-3-067 Characteristic of carbonaceous components in precipitation and aerosol at Japanese Monitoring Sites

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Key words:
carbonaceous components precipitation

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P-3-068 Retrieval of the global water vapor distribution from satellite observations in the blue spectral range

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Key words:
satellite observations water vapor

Water vapor is involved in many important chemical reactions in the atmosphere and contributes most to the natural greenhouse effect. Its atmospheric abundance is highly variable. Thus observations of the spatio-temporal variation on a global scale are of great importance. Water vapor observations are possible in different spectral ranges, e.g. in the microwave, thermal IR or near IR and visible spectral range. Satellite measurements in the red spectral range have the advantage that they are sensitive for the whole atmospheric column and that they provide global coverage. In this study we exploit the potential to retrieve the global water vapor distribution from satellite observations in the blue spectral range. Although the water vapor absorption in this spectral region is much weaker than in the red spectral range, such retrievals also have their advantages: First, because of the weak absorption, no corrections for spectral saturation effects (like in the red spetral region) have to be applied. Second, the surface albedo in the blue spectral region is very similar for land and ocean. Third, the water vapor distribution can be retrieved also from satellite instruments, which do not cover the red spectral range (like e.g. OMI or the future Sentinel missions).

Here we show first retrievals of the water vapor column in the blue spectral range from the GOME-2 instrument on METOP and OMI on AURA. Since GOME-2 also covers the red spectral region, a direct comparison with the results of the standard water vapor retrieval is possible. We characterise the accuracy and the detection limit of the new H2O analysis.

The OMI observations provide satellite DOAS results of H2O with unprecedented spatial resolution.

P-3-069 Polar stratospheric ozone: interactions with climate change, results from the ongoing project RECONCILE, and the 2010/11 Arctic ozone hole

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Key words: stratospheric ozone climate interactions

One of the most profound and well known examples of human impacts on atmospheric chemistry is the so called ozone hole. During the second half of the 20th century, anthropogenic emissions of chlorofluorocarbons (CFCs) led to a significant increase in stratospheric chlorine levels and hence the rate of ozone removal by catalytic cycles involving chlorine. While CFCs were essentially banned by the 1987 Montreal Protocol and its subsequent amendments, and stratospheric chlorine levels have recently started to decline again, another anthropogenic influence may at least delay the recovery of the stratospheric ozone layer: climate change, with little doubt a result of human emissions of carbon dioxide and other greenhouse gases, has led to changes in stratospheric temperature and circulation. The large ozone losses that typically occur in polar regions in spring are particularly affected by these changes. Here, we give an overview of the ozone-climate interactions affecting polar stratospheric ozone

loss, and present latest results from the international research project RECONCILE funded by the European Commission. Remaining open questions will be discussed including the possible impacts of recently suggested geoengineering concepts to artificially enhance the stratospheric aerosol loading. A special focus will also be put on the 2010/11 Arctic winter that saw the first Arctic Ozone hole, including an impact study on surface UV radiation in the densely populated northern mid-latitudes.

P-3-070 Particle formation events analysis in North Europe

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Key words:
particle formation and growth event Oxidant trajectory

The purpose of this study is to characterize the particle formation and growth events and find out the most favourable meteorological parameters, the most contribution air mass sources. The particle formation and growth events were analyzed from aerosol number size distribution data collected at a semi-rural background site in Demark during the period 2005-2007. The results showed that the events had clear seasonal variation occurred more frequently in spring and autumn, especially in May. The clearest events were observed occurring around 10% of all days, which was well associated with high solar radiation and photo chemical oxidant (Ox: O_3+NO_2) concentration. These evidences indicated that Ox played a crucial role for catalyzing the occurrence of the particle formation events. The main air mass sources were analyzed by 24 hours backward trajectories on the typical formation events days. The results showed that around 80% of the air masses came from north and northwest (NW) Nordic Sea passing through Denmark Jutland or Norway, about 10% of the air masses transporting form European mainland and a few coming from northeast. This analysis revealed that cleaner air masses were relatively more important for the contribution of Aitken mode particles than polluted ones and marine air masses from Nordic sea bring main sources for new particle formation and growth. NOx concentration was relative low when the air masses from NW and significant high when air masses from South, which indicates long-term pollution transportation from the mainland Europe. High number concentration of particles in diameter around 100 nm may be also transported from south. Further analysis in Denmark and south Sweden are planed to be conducted to proof this findings.

P-3-071 Complex vertical aerosol layering of natural andanthropogenic particles over the southeasternmediterranean: observations with earlinet lidar andaeronet photometer at lemesos, cyprus and comparisonwith transport modeling results.

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Key words:
natural particles anthropogenic particles polarization lidar sun photometer

In the south-eastern Mediterranean, complex aerosol profiles consisting of separate layers of different natural and anthropogenic particles or aerosol mixtures can be observed frequently. Consequently, these aerosols can have a complicated influence on climatic conditions, directly as well as indirectly via cloud processes. Observations of such complex aerosol layering within networks such as the European Aerosol Research Lidar Network (EARLINET) and the Aerosol Robotic Network (AERONET) and comparison of these observations with respective aerosol products of regional to global atmospheric transport modeling are required to improve our understanding of life cycles of aerosols and aerosol mixtures and their impact on climate. In this work is presented a case study of a desert dust outbreak from Syria and Saudi Arabia towards the eastern Mediterranean, occurred in September 2011. The observations were performed with a 532-nm polarization lidar (member of EARLINET) and a sun sky CIMEL sun-photometer (member of AERONET) operating at 8 channels from 340 to 1640 nm wavelength at Lemesos District, Cyprus (34°N, 33°E). The dust-laden air became mixed with air masses that crossed sources rich of biomass burning smoke and anthropogenic pollution (Ukraine, Russia, and Turkey). Over the Mediterranean Sea and more precisely over Cyprus, marine air and anthropogenic haze became mixed in the lower part of the plume by sea breeze circulations. This case study provides an ideal opportunity to demonstrate the potential of combined lidar-photometer observations to deliver detailed vertically resolved information of the aerosol characteristics in terms of particle backscatter and extinction coefficients, and the extinction-to-backscatter ratio, separately for the fine mode particle fraction (mostly smoke in the observed lofted layers) and the coarse mode fraction (mostly dust in the lofted layers), and volume and mass concentrations for fine and coarse mode particles. The retrieved aerosol profile data sets were compared with results of FLEXPART and the mineral dust regional BSC-DREAM8b model simulations. Good consistency of model and observational data were found and will be

presented at the conference.

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P-3-072 Attribution of direct ozone radiative forcing to spatially resolved emissions

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Tropospheric ozone has significant impacts on both air quality and climate. As a greenhouse gas, the global radiative forcing of tropospheric ozone since the preindustrial era is considered to be 350 [250 -- 650] mW/m^2 , the third largest behind CO_2 and CH_4, and it is projected to play an important role on both direct and indirect forcing. Consequently, there is considerable interest in controlling ozone precursors within the context of climate-air quality co-benefit strategies or as a means of achieving short-term climate forcing reduction to avoid ``dangerous" climate change. Using a combination of satellite observations of ozone and its radiative effect in conjunction with an adjoint chemical transport model, we quantify here the direct ozone radiative forcing due to NO_x, CO, and non-methane hydrocarbons (NMHC) emissions at an unprecedented spatial resolution that accounts for their dependence on latitude, altitude, chemical regime, and atmospheric circulation. We show that for equivalent radiative forcing, emission reductions vary intra-continentally by a factor of 2-3 and globally by an order of magnitude. The contribution of NO_x to the direct ozone radiative forcing varies from 50\% to 90\%, which must be offset by methane reductions for long-term climate mitigation.

P-3-073 Model-based evaluation of ozone climate penalty factor

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Poster Presentations, Session 4

P-4-001 Characteristics of Atmospheric Total Gaseous Mercury (TGM) observed in Urban Nanjing, China

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Key words
Total Gaseous Mercury Variation Meteorological parameter Nanjing

Mercury (Hg) is a well-known persistent and most toxic environmental pollutant. Gaseous elemental mercury (GEM) can be transported far beyond the regions where it is emitted and becomes well-mixed hemispherically due to its high volatility, low chemical reactivity and low solubility. Total gaseous mercury (TGM) was measured in Nanjing, the second largest city in Yangtze River Delta, for nearly one year (from Jan. 18 to Dec. 31, 2011). Meteorological parameters and other chemical compounds like CO, SO_2, NO_y and O_3 were measured at the same time. The annual average concentration of TGM is 7.26±4.26ng/m^3 with maximum of 179.76ng/m^3 and minimum of 0.83ng/m^3. TGM concentrations in Nanjing are significantly elevated compared to the continental global background values. A seasonal pattern of TGM with a descending order of summer, spring, fall, winter was observed. It's always opposite to most other measurements in China. The diurnal variation is conspicuous with daily maximum at 10 a.m, showing difference in each season. TGM concentrations are positively correlated with temperature and solar radiation with correlation coefficients of 0.432 and 0.534 respectively, suggesting the possible importance of natural source, which make comparable contribution to mercury contamination as anthropogenic source. TGM level increases when the wind is from south, southeast or southwest and decreases sharply as the wind from north and east. In addition, the concentration of mercury has significant correlation with CO and CO_2, but has negative correlation with O_3. The correlation with NO_y is suspicious.

P-4-002 The effect of Urban Air Pollutants on Pulmonary Inflammation and Systemic Oxidative Stress in Diabetic and Prediabetic Patients in Shanghai

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Key words
Air pollution Panel Study eNO EBC 8-OHdG

Background: Pulmonary and systemic oxidative stress and inflammation has been proposed as two plausible mechanisms linking air pollutants and corresponding health outcomes. People with diabetes may be especially vulnerable due to metabolic disorders. Objectives: 1) whether changes in pulmonary inflammatory biomarkers (eNO) are associated with acute changes in air pollutant levels in diabetic and prediabetic population. 2) if Diabetes are more vulnerable to the impact of air pollutant on pulmonary system. Methods: 60 elder people from communities managed by Luwan CDC in Shanghai were enrolled in this panel study, 34 of those were Diabetes and the rest are in prediabetic state. During 2010-2011 (Shanghai World Expo Period), each subjects has been followed two to four times repeated measurements of exhaled NO (eNO), Nitrite/Nitrate and IL-6 in Exhaled breath condensate (EBC) and 8-hydroxy-2'-deoxyguanosine (8-OHdG) in Urine. Meanwhile, regular gas pollutants (CO, SO_2, NOx, O_3), black carbon and number/surface/volume size distribution of particles with diameters between 5.6-560nm were monitored by a mobile platform. with hourly average record at a fixed site located less than 1km from communities where they live. Mixed linear effect model was used to identify associations between biomarkers and concentration of pollutants. Results: Significant change in eNO per interquatile range (IQR) increase of most pollutants by hourly average was found after controlling for 24hour and moving averages of temperature and RH, DOW, gender and Diabetic Type. This association showed a certain pattern in terms of lag hours with significant positive effect estimation before lag 8h (O_3 excluded), peak at lag 6-8h, and continuous trend towards no effect. Concentration of different particle size mode showed various pattern and influence on eNO. The largest positive effect was 15% change (CI: 7.99%-22.61%) in eNO which appeared in aitken mode on lag0. Different lag pattern was found after stratifying the association by diabetic type, BMI and waist-hip ratio(WHR). Group of diabetes, overweight (BMI＞25) and high obesity risk (WHR＞0.9) showed more significant and sustainable effect in SO_2, NO_2, BC and particle concentration compared with weak or even no positive effect in counter groups. Conclusions: These findings support that in this panel of diabetic and prediabetic subjects, changes of eNO, as the reflection of airway inflammatory status, are associated with increase in many gas pollutants as well as particle concentration. The association in terms of lag hours pattern may suggest a more acute effect of pollutants in airway inflammatory status and appeared stronger and more sustained effect in diabetic group.

P-4-003 Health impacts of ozone for present-day and under future scenarios over the UK and Western Europe.

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Key words
ozone health impacts future

Adverse effects of short-term exposure to ambient outdoor ozone concentrations have been well–documented. These effects range from symptom exacerbation and respiratory illness to increased hospital admissions and mortality. An important issue for policy-makers is how will air quality change in the future and what are the implications for human health. Here we quantify future changes in ozone levels across a range of future air quality emission scenarios as well as future changes in ozone-related health burdens for the UK and W. Europe. We also explore the interaction between elevated ozone levels and temperature that occur during heatwave episodes, which may become more frequent in the future.

We use the EMEP chemistry-transport model driven by meteorology from the Weather Research Forecasting (WRF) model to simulate ozone concentrations and temperature for present-day and under future emission scenarios. Three very different emissions scenarios were considered for 2030 ranging from high growth in future emissions (SRESA2), to following current legislation (CLE) and finally to implementing control measures that lead to “Maximum Feasible Reduction” (MFR). We perform simulations over W. Europe at 50 km by 50 km resolution and by use of nesting techniques at the higher resolution of 5 km by 5 km over the UK. We then perform regional health impact calculations based on these present-day and future ozone projections, accounting for future changes in population and thresholds below which no adverse ozone-related health impacts occur.

The simulated annual and summer-mean ozone response varies with emission scenario and region. However, the distinct difference is the projected change in ozone over highly populated urban vs. rural locations, as localised ozone changes (due to emission changes) are superimposed on background surface ozone changes. In the SRESA2 scenario, background ozone increases because of hemispheric increases in ozone precursors; consequently ozone increases in rural regions. However, the prominent effect over UK cities is reduced ozone since localised NO_x emission increases leads to increased loss of ozone by reaction with NO. Conversely, ozone increases over most UK cities but decreases over rural regions under the MFR scenario. The ozone increases are generally highest under the CLE scenario since both increases in background ozone concentration and decreases in UK NO_x emissions enhance ozone levels. In all three scenarios, annual deaths brought forward increase due to the greater projected populations in 2030 compared to present-day. However the annual number of deaths brought forward per 100,000 population reflects the differences in ozone changes between densely populated and rural regions described above. These changes are modified slightly and more substantially when a 35 ppbv and 50 ppbv respectively threshold for daily max 8 hour ozone, below which no harmful effects occur, is introduced. This highlights the importance of both of changes in average ozone concentrations as well as changes in the occurrence of ozone exceedence episodes. Over the UK, we found a significant interactive effect of ozone and heat on mortality in London. The issues of thresholds and modification by weather are clearly important for projections of future ozone-related health impacts

P-4-004 Aged sea salt aerosol in the urban environment: a cause for concern?

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Key words
sea salt aerosol health effects chemical ageing

The Sydney Particle Study was an investigation of the processes leading to particle formation in a major Australian city, Sydney with ~4.5 million inhabitants. A comprehensive observation program took place at the Westmead Air Quality Station between 5 February and 7 March 2011. Westmead is situated 36km inland from the entrance to Sydney Harbour. The study investigated the primary and secondary sources of aerosol and the conversion processes within the urban atmosphere, including new particle formation (see Keywood et al. IGAC2012 abstract). The largest source of primary aerosol was sea salt. Sea salt aerosol emissions are caused by wave-breaking in the open ocean and from coastal surf breaks. The magnitude of the open ocean sea salt fluxes is a strong function of the surface shear stress and hence the near-surface wind speed. Our chemical transport modelling (CTM) study shows that emissions from as far away as the Southern Ocean (50 degrees S) can influence sea salt levels in urban Sydney (33 degrees S). This is not entirely surprising given the distances other natural sources, i.e. soil dust is known to travel in Australia (1000's of km).

On average, sea salt composed 5 micrograms per cubic metre (33%) of the observed total mass of $PM_{2.5}$, with a peak of 19 micrograms per cubic metre (75%) during one event on 2nd March. Chemical analysis of the sea salt during the campaign showed it was a mixture of 65% fresh emissions and 35% aged particles. Of the aged particles, the sodium was bound with nitrate (60%) and non-sea salt sulphate (40%). The ISORROPIA code has been used to make an assessment of the equilibrium concentrations, given the observed concentrations of inorganic species, the temperature and relative humidity. This result was then compared to the CTM equilibrium predictions. The model also shows how the evolution of the chemical mass of Sydney particles occurs, including modification of sea salt particles via condensation of acidic and organic gases. Future work will include the microphysics model GLOMAP to elucidate how the number, size and composition of particles evolve in the Sydney air shed.

We have made an assessment of the contribution of the aged sea salt component to health effects in Sydney, and have compared this to an experimental run where the sea salt emissions were switched off. Given that sea salt contributes over 3000 Tg per year to total global aerosol, the chemical ageing of sea salt particles could result in significant human exposure to nitrate and sulphate particulate matter in coastal regions.

P-4-005 Improving Chemical Weather Forecasting Using Satellite Data

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The Met Office's operational regional Air Quality Unified Model (AQUM) contains a description of atmospheric chemistry/aerosols which allows for the short-term forecast of chemical weather (e.g. high concentrations of ozone or nitrogen dioxide, which can trigger poor air quality warnings). AQUM's performance has so far only been tested against a network of surface monitoring stations.

Therefore, with recent improvements in the quality and quantity of satellite measurements, data products (e.g. tropospheric columns, vertical profiles) from several satellite instruments will be used to test the performance of the model. First comparisons between an AQUM simulation for the UK heatwave event of July 2006 and data from OMI, TES (both on AURA) and MODIS (on AQUA) have identified a few model biases.

Model biases discovered between the AQUM and OMI July 2006 monthly mean NO_2 tropospheric columns highlight potential biases in the peak spatial NO_2 emissions over northern and southern England. In the OMI data, the peak NO_2 emissions are located over the the denser urban region in the south, whereas in the model the peak emissions, with similar magnitudes but higher in the satellite data, are located in northern England. Therefore, currently sensitivity tests are investigating the quality/accuracy of the emissions datasets used in the model to represent urban pollution.

Further more quantitative and statistical comparisons will hopefully identify further potential improvements to the model such as the chemistry/aerosol scheme, model resolution or initial and boundary condition datasets, and if time allows, implement improvements operationally.

P-4-006 Selectively Detecting Influenza Viruses in Exhaled Breath in Minutes Using Silicon Nanowire Sensor: A New Arena for Flu Diagnosis

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Key words
Influenza A H_3N_2 virus Exhaled Breath Condensate Silicon Nanowire Flu diagnosis RT-qPCR

Influenza epidemics worldwide result in substantial economic and human costs

annually. However, rapid and reliable flu diagnosis method is significantly lacking. Here we have demonstrated the selective detection of influenza A viruses down to 29/μL in clinical exhaled breath condensate (EBC) samples (diluted by 100-fold) within minutes using silicon nanowire (SiNW) sensor devices. In all cases, we have observed that EBC samples tested positive or negative by gold standard method RT-qPCR generated corresponding positive or negative SiNW sensor responses. High selectivity of SiNW sensing was also demonstrated using H1N1 viruses, 8 iso PGF 2a and inert nanoparticles. Finally, magnetic beads were shown capable of enhancing SiNW sensing directly for low level viruses and 8 iso PGF 2a. When calibrated by virus standards and EBC controls, our work suggests that SiNW sensor device can be reliably applied to the diagnosis of flu in a clinical setting with 2-order magnitude less time compared to gold standard method RT-qPCR.

P-4-007 Negatively and Positively Charged Bioaerosol Concentration and Diversity in Natural Environments

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Key words
Positive charge Negative charge Bacterial aerosol Culturable diversity PCR-DGGE

In this study, the positively and negatively charged culturable bacterial aerosol concentration and diversity were investigated in office, hotel and outdoor environments. For positively or negatively charged bioaerosols, the air samples were collected into different regions of two square agar plates using an electrostatic sampler (1.1 kv/cm) at a flow rate of 3 L/min for 40 min. Those bypassing the sampler were also collected using a filter at the downstream of the electrostatic sampler in the hotel room. The air samplings were independently conducted three times in each of the environments studied. The collected bacterial aerosols were cultured directly on agar plates at 26 °C, and the colony forming units (CFU) were manually counted for the culturable bioaerosol concentrations. In addition, the CFUs were washed off from the agar plates, and further subjected to polymerase chain reaction (PCR) and denaturing gradient gel electrophoresis (DGGE) for culturable diversity analysis.

P-4-008 Aerosol compositions during heat waves in Taipei city

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Key words
particulate pollutants urban air quality organic aerosols atmospheric chemistry and health

Introduction: Heat waves are expected to occur more frequently with record-breaking extremes under climate changes; and megacities are the most vulnerable areas. On top of the climate change, megacities are affected by both heat island effect and air pollution. During heat wave periods, the aerosol compositions may be changed due to higher photochemical reaction rates under higher temperatures. The levels of certain toxic compounds may be increased accordingly. The objective of this work is to assess the aerosol compositions, including toxic species, during heat-wave and non-heat-wave periods in Taipei metropolitan.

Methods: Aerosol sampling was conducted for two weeks in summer 2011 in the urban center of Taipei metropolitan and a downwind rural site located in mountainous area. One high-volume sampler and one RP2300 sampler with denuder were used for sampling. Heat wave periods were defined as dates with daily maximum temperature higher than 35°C. The concentrations of various pollutants were compared between urban and rural areas, and between heat-wave and non-heat-wave periods.

Results and Discussion: The concentrations of $PM_{2.5}$, sulfate, nitrate, ammonium, elemental carbon (EC), organic carbon (OC), water-soluble organic carbon (WSOC), and polycyclic aromatic hydrocarbons (PAHs) in the urban and rural sites are analyzed. The levels of EC, a primary pollutant, are mostly higher in the urban site. In the downwind rural site, the concentrations of $PM_{2.5}$, sulfate, EC and PAHs are sometimes higher at nighttime than at daytime. The patterns of OC and WSOC in the rural site are very close to those in the urban site, the levels are sometimes higher in the rural site. Two peak levels occurred in the daytime of August 18th and 20th of the heat wave period. The percentages of WSOC, which may affect cloud formation, in OC are slightly higher during heat wave periods compared to the non-heat-wave period. The levels of PAHs, a group of probable human carcinogens, show a significant peak in the urban site in the last day of the heat wave period, indicating potentially higher health risks. In addition, the levels of sulfate which is a respiratory-irritating indicator also show one peak occurred in the urban and rural sites on the same day. The mechanism of such peaks deserves further investigation. The current case study is a pilot work illustrated that the impacts of heat wave on the pollutant levels deserve more attention under climate change; the research outcomes could have important policy implications for pollution abatement during heat wave periods.

P-4-009 Three-dimensional variational assimilation of surface PM_{10} observations and its application over China

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Key words
data assimilation particulate matter

This study aims to reduce the uncertainty of initial conditions to improve the short-range aerosol forecasts. Based on the National Centers for Environmental Prediction (NCEP) Gridpoint Statistical Interpolation (GSI) three-dimensional variational (3DVAR) data assimilation system, ground-observed observations of particulate matter with diameters less than 10 μm (PM_{10}) are assimilated into the Goddard Chemistry Aerosol Radiation and Transport (GOCART) module within the Weather Research and Forecasting/Chemistry (WRF/Chem) model. The model state is then adjusted to improve the agreement between the model simulated PM_{10} and surface PM_{10} observations using GOCART aerosol species as analysis variables. The aerosol analysis is performed every 6 hour between UTC 01 and 28 June 2011 over China, using the previous cycle's 6-h forecast as the background. The impact of ground-based PM_{10} data assimilation was evaluated when compared to surface PM_{10} observations and independent AOD observations from the Moderate Resolution Imaging Spectroradiometer (MODIS) and the Aerosol Robotic Network (AERONET). Results show that assimilation of PM_{10} observations significantly improves aerosol analysis and subsequent one-day forecasts. However, fast model error growth occurs in the early forecast hours. Analysis indicates that this behavior is mainly induced by emission and advection, which points to the need for optimization of emission rate to improve more extended-range forecasts.

P-4-010 Health Risk Assessment of benzene series compounds in the Ambient Air of the Northeastern Urban Area of Beijing, China

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Key words
Ambient air BTEX Pollution status Health risk assessment Beijing

Benzene series compounds is the term that refers to such single-ring aromatic

hydrocarbons as BTEX (benzene, toluene, ethylbenzene, o-xylene, m-xylene and p-xylene), styrene and cumene. These compounds have harmful effect on the human respiratory system, hematopoietic system and the nervous system due to their strong toxicity and carcinogenicity. As a Class 1 human carcinogen identified by International Agency for Research on Cancer (IARC), long-term exposure to benzene can cause the human bone marrow and genetic damage, even may lead to leukemia and lymphatic diseases.

With the rapid development of the urbanization in China, human health, regional ambient air quality and ecological environment are negatively affected by the serious pollution of volatile organic compounds in ambient air, especially benzene series compounds. The hourly average concentrations of BTEX are higher than 20μg/m^3, even up to 90μg/m^3 with the highest concentration proportion of toluene followed by benzene and the highest concentrations observed in summer, lowest concentrations in winter in some large and medium-sized cities of China. Some monitoring data indicates that the annual average concentrations of benzene are higher than the values set in the Ambient Air Quality Standards for benzene in Japan (3.25μg/m^3), United Kingdom (5.0μg/m^3), Sweden (3.31μg/m^3) and New Zealand (10μg/m^3). Many studies showed that the concentration ratio benzene to toluene (B/T) in the urban ambient air is about 0.6, which indicated that the vehicular exhaust is the main source with and the use of paints and adhesives being the two important sources of benzene series compounds in the urban ambient air in China.

Health risk assessment of benzene series compounds in the ambient air is very necessary in the view point of the pollution status of benzene series compounds in the ambient air in China. Continuous on-line measurements of BTEX, styrene and cumene in the ambient air were made by an airmo VOCs instrument in the northeastern urban area of Beijing for two months in 2012 in this study. The monitoring site was located at the top of the building of the Atmospheric Photochemical Smog Simulation Laboratory of CRAES (40°02′N, 116°25′E), which is 6 m high from the ground with no obvious local air pollution sources nearby. The ambient level and the variation characteristics of BTEX, styrene and cumene were analyzed and the health risk assessment model developed by the United StatesEnvironmental Protection Agency (US.EPA) in 1989 was applied to make the preliminary health risk assessment of BTEX, styrene and cumene in the ambient air in the northeastern urban area of Beijing.

P-4-011 Health Risk Assessment of Volatile Halogenated Hydrocarbons in the Ambient Air in the Northeastern Urban Area of Beijing

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Key words
Ambient air Volatile Halogenated Hydrocarbons Pollution status Health risk assessment Beijing

Volatile Halogenated Hydrocarbons (VHHs) concerned most in the atmospheric environmental research field are Chlorofluorocarbons (CFCs) and the toxic and harmful VHHs. The toxic and harmful VHHs mainly include halogenated benzene and some halogenated hydrocarbons, such as trichloromethane, tetrachloromethane and chloroethylene. There are 32 kinds of VHHs among the list of “the 188 kinds of toxic gases (Hazardous Air Pollutants)” upgraded by United States Environmental Protection Agency (US.EPA) in 2012.

Since the toxic and harmful VHHs are irritating and corrosive, they may be detrimental to human skin, liver, heart, kidney, pancreas and central nervous system, resulting in discomforts, like fatigue, nausea and headache. In addition, some toxic and harmful VHHs such as vinyl chloride, trichloroethylene, trichloromethane have carcinogenicity. Vinyl chloride has been identified to be Class 1 carcinogens by the International Agency for Research on Cancer (IARC), long term exposure to vinyl chloride may cause malignant tumors such as liver angiosarcoma and biliary vascular sarcoma.

With the rapid development of the urbanization in China, human health, regional ambient air quality and ecological environment are negatively affected by the serious pollution of volatile organic compounds in ambient air. Though there are many researches about VOCs pollution in China, only a few studies concerns about toxic and harmful VHHs, which were mainly conducted in Hongkong, Guangzhou, Beijing, Shenyang and Changsha. These studies are mainly focused on the ambient level, composition, the variation characteristics and the source identification of the toxic and harmful VHHs, which showed that the anthropogenic sources including oil refining, degreasing agent, dry cleaning agent and maritime transportation are the main sources of the toxic and harmful VHHs in the ambient air in the urban areas in China, and the natural sources including marine and terrestrial biological activities, marine photochemistry and volcanic activities are also the important sources of the toxic and harmful VHHs in the ambient air in the urban areas in China.

Health risk assessment of toxic and harmful VHHs in the ambient air is very necessary the in the view point of the pollution status of the toxic and harmful VHHs in the ambient air in China. Continuous on-line measurements of 29 kinds of toxic and harmful VHHs including chlorobenzene and tetrachloride in the ambient air were made by an airmo VOCs instrument in the northeastern urban area of Beijing for two months in 2012 in this study. The monitoring site is located at the top of the building of the Atmospheric Photochemical Smog Simulation Laboratory of CRAES (40°02′N, 116°25′E), which is 6 m high from the ground with no obvious local air pollution sources nearby. The ambient level and the variation characteristics of toxic and harmful VHHs were analyzed and the health risk assessment model developed by US.EPA in 1989 was applied to make the preliminary health risk assessment of toxic and harmful VHHs in the ambient air in the northeastern urban area of Beijing.

P-4-012 Interaction between Pollutant Exposure and Human Metabolism: Case of PAHs

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Key words

Polycyclic aromatic hydrocarbons Metabolism Human exposure Hydroxyl PAHs One-compartment model

Our previous study suggested that maternal metabolism might play a key role in determining placental concentrations of polycyclic aromatic hydrocarbons (PAHs), which was significant risk factor for infants' neural tube defects. In this study, the goal was to explore the relationship between human's exposure and elimination to PAHs based on the biomarker analysis. Human biological samples were collected from population in rural region of Northern China, and PAHs in serum and their hydroxylated metabolites (hydroxyl PAHs) in both urine and serum were measured using electronic ionization gas chromatographic mass spectrometry (GC/MS). Human's exposure and elimination to PAHs were explored with one-compartment model based on the measured biomarkers.

In urine samples collected in the heating-season of winter, levels of 1-hydroxypyrene, hydroxyphenanthrenes, hydroxyfluorenes, and hydroxybiphenyls were 6.12, 3.98, 2.48, and 2.17-fold higher of those in non-heating season of spring, respectively, indicating that household heating was a major source of PAHs exposure. On the contrary, in heating season serum levels of precursor pollutants were 0.34, 0.13, 0.19, and 0.36 of those in non-heating season for pyrene, phenanthrene, fluorene, and biphenyl, respectively. The estimated constant of elimination to PAHs ranged from 19 to 54-fold faster in heating season than in non-heating season, and the step of hydroxylation metabolism accounted for most of the elevated constant of elimination. The results showed that enhanced disposal rate to PAHs might be a result of their elevated exposure. To reveal the underlying mechanism in this environment-gene interaction could be a key to evaluate health effects of exposure to these pollutants.

P-4-013 Climate, Air Quality, Health and Social Vulnerability in Asia: A New International Framework for Interdisciplinary Advances

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Key words
Climate Air Quality Social Vulnerability Health International Framework Interdisciplinary Advances

The paper presents a new program for studying the complex relations between air pollution (ozone, $PM_{2.5}$), climate change, and health impacts in Asia. The proposed interdisciplinary framework follows the recommendations of an international gathering of 60 scientists from Asian, American and European scientists held in Guangzhou, China, on 9-11 April 2012. The Project will focus specifically on the Asian regions affected by rapid economic and urban development and will provide science-based information to decision-makers to assess the vulnerability of the populations of the region, manage the health risks, and take the appropriate measures to optimally control the emissions of pollutants.

The key objectives of the project are:

1.To develop a framework to assess the various economic, social, ecological, meteorological, and climatic factors that contribute to air pollution in Asia and the related vulnerability regarding human health;

2.To determine the benefits and barriers for managing health risks and mitigating the emissions of pollutants;

3.To conduct integrated pilot study to compare three urban areas affected in different ways by air pollution problems and related health impacts;

4.To provide scientifically based information to help decision-makers to take appropriate actions to reduce emissions in the three cities and in other Asian regions under rapid development;

5.To compare results and strategies with those provided by other studies conducted elsewhere in the world.

A pilot study will be developed to test theframework to assess the various economic, social, ecological, meteorological, and climatic factors that contribute to air pollution in Asia and the related vulnerability regarding human health. The demonstration studywill focus on three major urban areas of Asia: Guangzhou, Taipei and Xi'An. These urbanized areas differ in several respects (population, economic development, climate conditions air pollution policies, etc.). A comparision of the results from these cities will enable a broad perspective tobe gained, and will hopefully guide the authorities of other Asian urban centers to rapidly take the actions that address air pollution and related health issues.

P-4-014 Atmospheric polycyclic aromatic hydrocarbons in north China during the non-heating season

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Key words
polycyclic aromatic hydrocarbons passive air samplers north China
non-heating season

Polycyclic aromatic hydrocarbons (PAHs) are one of the carcinogenic organic pollutants formed during incomplete combustion of organic materials. Moreover, inhalation of PAHs in atmosphere is the significant exposure pathway for human, and the subsequence health effects are also noteworthy. In this study, 89 air samples were collected using polyurethane foam (PUF) disks passive samplers at the Chinese Northern Plain, a region with a total area of 527 400 km^2 and a high density of population, from June 2011 to August 2011. The concentrations of PAHs including 25 kinds of PAHs and 13 Nitro-PAHs in ambient air were analyzed using electronic ionization gas chromatographic mass spectrometry (GC-MS). The passive air samplers were calibrated by one active high-volume air sampler and calculated by two-film model.

The average total PAH concentration was 350 ± 207 ng m^{-3}, and the level in Shanxi was 1.4 to 2 times significantly higher than in the other region. No significant difference was found in PAH concentrations among the urban, the suburban and the rural areas (337 ± 145 ng m^{-3}, 378 ± 226 ng m^{-3} and 348 ± 203 ng m^{-3}, respectively), while the concentrations of PAHs at control sites (55.3 ± 35.9 ng m^{-3}) were almost one order of magnitude lower than those of the other sites. Source of PAHs were identified by principle component analysis and contributions were further estimated by multivariate linear regression. The results showed that vehicle sources and coal combustion emission were identified as major sources in the non-heating seasons in these study areas. The varied spatial distribution of PAHs might confirmed that they have different sources to the environment, this high value of Shanxi might be caused by the plenty of works in recovering, processing and delivering of coal, for instance.

P-4-015 Meteorological factors modify the association of air pollution between mortality in Beijing

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Key words
meteorological factors air pollution modify mortality

Introduction: It is well known that high temperatures and air pollution are associated with short term increases in mortality; however, the combined effect of these exposures is less clear. The focus of this study was to increase understanding on the temperature-air pollutant-mortality relationship in Beijing, China.

Methods: Associations between daily mortality counts, daily average temperature, 8-hr daily maximum ozone (O_3) and 24-hr average PM_{10}, 24-hr average $PM_{2.5}$ were estimated using generalized additive Poisson regression (GAM). In order to emphasize the impact of weather conditions on air quality, a pollution linking meteorological index (Plam) was introduced to represent the air pollution meteorological condition, emphasizing the impact of meteorological factors on air pollution. The Plam index was derived from relationship of PM_{10} and key meteorological data in Beijing and its surrounding areas. A high Plam index at Beijing indicates the high degree of stable weather in Beijing. Thus, the Plam index represents one main sensitive dependent of air pollution impact, i.e., the combination of meteorological factors and air pollutants.

Moreover, we tried to calculate a closed form estimate of the asymptotically exact covariance matrix of the linear component of a GAM using the S package gam.exact, an extended version of gam that implements these estimates.

Results: During the year Jan. 2007-Dec., 2009, the mean daily death numbers for all nonaccidental causes, cardiovascular causes and respiratory causes – in respective order – were 118, 54 and 14 in Beijing.

Main effects estimates found that temperature and $PM_{2.5}$ were all associated with above three mortality outcomes. A 10-$\mu g/m^3$ increase in $PM_{2.5}$ (lag 1 day) was associated with a 0.37% (95% CI: 0.22 to 0.51) increase in total mortality, 0.49% (95% CI: 0.28 to 0.70) increase in cardiovascular mortality and 0.48% (95% CI: 0.07 to 0.89) increase in respiratory mortality. Joint effects demonstrated that the highest counts of mortality occurred when both temperature and air pollution were at their highest.

And, though PM_{10} showed no significant effect on mortality, which was in accordance with many studies; however, the Plam index showed suggestive correlation.

Conclusions: These findings suggest meteorological factors play an important role in the air pollution mortality relationship. The effects of air pollution differ by ranges of meteorological factors, that is, meteorological factors could “modify” the

effects of air pollution, and so could air pollution does that of meteorological factors.

Discussions: Analysis of meteorological factors was emphasized in our study. Meteorological conditions affect urban air quality trends, temperature and mixing height, strongly influencing ozone conditions, while moisture levels particularly relative humidity are the strongest predictors of PM concentrations. As a result, meteorological conditions affect human exposure to air pollution. Thus, it is reasonable to consider more meteorological data while studying the relationship between air pollution and health effects.

P-4-016 Top-Down Estimate of Dust Emissions through Integration of MODIS and MISR Aerosol Retrievals with the GEOS-Chem adjoint model

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Key words

GEOSchem adjoint MODIS, MISR

Predicting the influences of dust on atmospheric composition, climate, and human health requires accurate knowledge of dust emissions, but large uncertainties persist in quantifying mineral sources. This study presents a new method for combined use of satellite-measured radiances and inverse modeling to spatially constrain the amount and location of dust emissions. The technique is illustrated with a case study in May 2008; the dust emissions in Taklimakan and Gobi deserts are spatially optimized using the GEOS-Chem chemical transport model and its adjoint constrained by aerosol optical depth (AOD) that are derived over the downwind dark-surface region in China from MODIS reflectance with the aerosol single scattering properties consistent with GEOS-chem. The adjoint inverse modeling yields an overall 51% decrease in prior dust emissions estimated by GEOS-Chem over the Taklimakan-Gobi area, with more significant reductions south of the Gobi Desert. The model simulation with optimized dust emissions shows much better agreement with independent observations from MISR AOD and MODIS deep blue AOD over the dust source region and surface PM_{10} concentrations. The technique of this study can be applied to global multi-sensor remote sensing data for constraining dust emissions at various temporal and spatial scales, and hence improving the quantification of dust effects on climate, air quality, and human health.

P-4-017 “Taiwan Megacity Environmental Research” – Impacts of Secondary Organic Aerosols

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Key words
secondary organic aerosols aerosol and health impacts aerosols and cloud formation aerosols and climate change

This presentation introduces an integrated project “Taiwan Megacity Environmental Research”, focusing on the potential impacts of secondary organic aerosols (SOA) on air quality, regional climate, and human health in Taiwan megacity. This is an interdisciplinary research project with eleven sub-projects including two chamber studies, five field sampling/observation and chemical analysis works, two health-impact evaluation studies, and two modeling works. Multi-disciplinary researchers work together with complementary methods to study the following topics: the characteristics of emissions and emission factors of SOA precursors from important biogenic sources, the possible chemical reaction pathways and transformation rates of important SOA species, the concentrations of elemental carbon (EC), organic carbon (OC), total SOA, SOA markers, SOA precursors and other gaseous pollutants under different meteorological conditions and different land-use surfaces; the mass concentration, size distribution, vertical distribution, and optical property of aerosols and their relationships with SOA; the exposure level of Taiwan residents to aerosol and total SOA; and the relationship between temperature, aerosols, thermal stress, and health indicators. In addition, a detailed organic aerosol scheme will be established to accommodate the obtained emission factors, transformation rates, and reaction pathways. The effects of temperature increase caused by heat island effect and global/regional warming on aerosol characteristics and radiative forcing will also be evaluated with regional atmospheric chemistry models.

The detailed results of the chamber studies, field works, health-effect evaluation, and modeling works are presented in eight posters of this conference. In summary, we found that biogenic VOCs emissions from Areca catechu and its SOA formation in the smog chamber were affected by temperature and lighting conditions. In the field, a significant diurnal cycle for the ability of aerosols to form cloud condensation nuclei (CCN) was observed. The ratio of carbonyl-to-sulfate ratio during the daytime is usually at least twice more than that at nighttime. The contributed carbonyl related species might have lower hygroscopicity as compared to the sulfate. In addition, the contribution of biogenic vs. anthropogenic fraction of VOCs was evaluated; there may be a temperature threshold, beyond which the biogenic fraction of VOCs increases with ambient temperatures. Compositions of $PM_{2.5}$ including sulfate, nitrate, ammonium, EC, OC, water-soluble OC (WSOC), and polycyclic aromatic hydrocarbons (PAHs) showed different proportions during heat-wave and non-heat-wave periods. Besides, vertical profiles of aerosols and temperatures were obtained; the heights of the inversion layer were lower in the urban areas compared to the rural areas and these factors may affect the diurnal variation of SOA. CMAQ simulation considering photochemical processes and microphysical processes of SOA and inorganic aerosols was applied to help explaining the observations. Human exposure to PM and associated health impacts were assessed. Heart-rate variability (HRV) showed a significant change in young healthy adults while exposing to traffic emissions. A non-linear relationship was observed between HRV and environment factors (thermal conditions and aerosols) associated with time. The impacts on HRV could last for 60 minutes after exposure.

P-4-018 Exposure to Atmospheric Particulates in Some Area of Beijing Exacerbated Atherosclerosis in $ApoE^{-/-}$ mice Mouse Model

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Key words
ambient particulates atherosclerosis ApoE-/- mice systemic inflammation oxidative stress lipid metabolism

Air pollution is associated with significant adverse health effects, including increased cardiovascular morbidity and mortality. However researches on the cardiovascular effect of actual atmosphere are limited, particularly in terms of

susceptible animal models. In order to investigate the effects on atherosclerosis development of ApoE knockout mice (ApoE-/- mice) exposed to atmospheric particulates in some area of Beijing, we chose twenty 10-week-old healthy adult male ApoE-/- mice and randomly divided them into two groups (exposure group and control group). During Jan 18th, 2010 to Mar 18th, 2010, $ApoE^{-/-}$ mice of exposure group were exposed to actual atmosphere in some area of Beijing, while the control group exposed to atmosphere filtered without particulates. During the experiment period, PM_{10} and $PM_{2.5}$ in Beijing and the particulate concentrations in exposed chambers were monitored. Two months after exposure, the total cholesterol (TC), total triglycerides (TG), high-density lipoprotein (HDL), low-density lipoprotein (LDL), oxidized low-density lipoprotein (ox-LDL), malonaldehyde (MDA), super oxide dlsmutase (SOD), total antioxidant capacity (TOA), C-reactive protein (CRP), interleukin-6（IL-6） and tumor necrosis factor-α (TNF- α) in serum of $ApoE^{-/-}$ mice were determined. IL-6 and TNF-α in bronchoalveolar lavage fluid and the morphology of aortic arch pathology were also observed. Our study showed that TC, LDL, ox-LDL, TNF-αand Hs-CRP in serum and TNF-α and IL-6 in bronchoalveolar lavage fluid in the exposure group were significantly increased when compared with the control group. There was also significant difference between the two groups in terms of TOA and MDA in serum. For the pathology of aortic arch, it showed that plaques area and lipid center area in the exposure group increased significantly. Our study suggested that exposure to ambient atmospheric particulates in Beijing could induce considerable systemic inflammation and oxidative stress in $ApoE^{-/-}$ mice, which had effects on lipid metabolism, and progression of atherosclerosis.

P-4-019 Exposure levels of fine particle and carbon monoxide in transportation modes while commuting in Beijing, China

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Key words
personal exposure fine particle carbon monoxide transportation modes

Introduction The epidemiological association between exposure in traffic and increased health risk has been well documented in previous studies. It’s important to compare commuters’ exposure to traffic-related air pollutants in different transportation modes in order to provide some information which can help the public to avoid potential health risk related to traffic exposure.
Methods A field investigation measuring bus commuters, taxi commuters and cyclists’ exposure to fine particle ($PM_{2.5}$) and carbon monoxide (CO) were carried out in winter 2010 in Beijing. The commuters travelled along the same routes

during both traffic rush hours and non traffic rush hours by three transportation modes. In addition, simultaneous measurements of $PM_{2.5}$ and CO concentrations were conducted in a fixed monitoring site which located close to a busy road junction. Trip duration and inhalation rates were considered to calculate whole trip exposures and comparisons were made among the three transportation modes.
Results After confounding factors were controlled, we found that bus commuters (42.40 microgram per cubic meter) and cyclists (49.10 microgram per cubic meter were exposed to significantly higher concentrations of $PM_{2.5}$ compared with taxi commuters (31.64 microgram per cubic meter). By contrast, CO concentrations were the highest when commuting by taxi (5.21 ppm), in the middle level when by bus (2.41 ppm), and the lowest when by bicycle (1.90 ppm). There were significant differences between $PM_{2.5}$ and CO concentrations in transportation modes and at the fixed monitoring site ($p<0.05$), especially during traffic rush hours. Comparisons of whole trip exposures showed that cyclists experienced the highest whole trip exposures to both $PM_{2.5}$ and CO ($p<0.05$).
Discussion Ambient $PM_{2.5}$ level in our study was relatively higher compared with studies conducted in other places, such as London. Taxis with windows closed inclined to prevent infiltration of $PM_{2.5}$ into the compartment, thus taxi commuters were exposed to lowest concentration of $PM_{2.5}$. Meanwhile, as the main source of CO is in-vehicle pollution, semi impermeable characteristic of taxis lead to accumulation of CO. The complicated situations in traffic microenvironments can not be elucidate well by fixed site monitoring data. Cyclists' higher inhalation rate and longer trip duration across the same distance lead to their highest whole trip exposure to $PM_{2.5}$ and CO.
Conclusion $PM_{2.5}$ and CO concentrations are greatly influenced by transportation modes. Fixed site monitoring data can not present well the variations of pollutants' concentrations in traffic microenvironments. Furthermore, cyclists are possibly subject to higher health risk related to traffic exposure compared with bus and taxi commuters.

P-4-020 Degradation of Nitrogen Oxides on Photocatalytic Cements: Solution of the Urban NO_2-Problem?

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Key words
photocatalytic cements NO_2 air quality

Air pollution is known to directly affect human health by interaction with the respiratory and cardiovascular systems. One of the most harmful environmental pollutants are nitrogen oxides (NO_x) formed in automotive engines and industrial combustion systems when air is used as an oxidant agent. Therefore controlling air quality of urban areas appears to be an extremely challenging task. In the very recent years photo-catalytic self-cleaning and “de-polluting” materials have been suggested as a remediation technology mainly for NO_x and aromatic VOCs in the polluted urban environment. For this reason, new construction materials are developed based on a matrix with photocatalytic oxides (often titanium dioxide, TiO_2).

TiO_2 is quite well known for its photocatalysis properties. Accordingly it is suggested to react with atmospheric pollutants and induce a cleansing effect by reducing their concentrations. Therefore, the PhotoPAQ project (PHOTOcatalytic remediation Processes on Air Quality) was design to evaluate the feasibility of using such TiO_2 based material to alleviate the air pollution problem under real atmospheric conditions based on laboratory and field campaigns approaches.

Prior to a real test of such material coating in an urban busy tunnel in Brussels, experiments have been performed in the laboratory with a photocatalytic reactor. This presentation focuses on the characterization of the material for NO_2 removal as a function of various parameters i.e., irradiance, relative humidity. The depollution efficiency of such materials will be presented and discussed along with their potential applications to real case depollution in urban areas.

P-4-021 Combining CALIOP and MODIS to Estimate the ground extinction coefficient

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Key words
extinction coefficient remote sensing human health

Numerous studies have documented that aerosols have a large influence on human health, including oxidative damage and cell damage. According to the toxicology, the effects are from both the size and components of aerosols. Meanwhile, epidemiology uses $PM_{2.5}$ as the representation of aerosols, and finds that it has a statistics coefficient with human health, such as death rate or life expectancy. However, in China, the data of $PM_{2.5}$ covering large scale is hardly available. Considering that aerosol extinction coefficient can somehow reflect the size and components of aerosols. So, it can also act as an indicator of aerosols in the atmosphere pollution and human health study. In order to confirm this possibility, this research tries to combine the aerosol product of CALIOP and MODIS to estimate the ground extinction coefficient, preparing for the further research of the relationship between extinction coefficient and human health.

CALIOP and MODIS are both remote sensors fixed on NASA’s satellites, with the former on CALIPSO and the latter on TERRA and AQUA. MODIS provides the data of aerosol optical depth, which is the extinction coefficient of the total column atmosphere. And CALIOP provides the profile of extinction coefficient of column

atmosphere at the point under it. There are two steps of the work: first is checking the consistence of CALIOP's and MODIS's data, and second is estimating the ground extinction coefficient if the consistence is good enough.
From the whole year data of 2009 in China, the consistence varied severely, influenced by cloud and location. Without any screening, the result is very unsatisfying, with the coefficient (r) less than 0.1 (n=1012) for the whole year. However, after getting rid of cloudy points, r increases. For example, in Aug 1st 2009, before screening, r is 0.35 (n=358). While after screening, r increases to 0.55 (n=265). Furthermore, r increases more when only focus at the points under CALIOP (r=0.9, n=11, Aug 1st 2009). Therefore, it is available to estimate the ground extinction coefficient combining CALIOP and MODIS, but only in sunny day and in points under CALIOP, which is too narrow scale for aerosols and human health research.

P-4-022 High time-resolution chemical characterization of the water-soluble ions in PM with MAGAR

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Key words
biomass burning water soluble ions haze

The growing need for on-line high frequency aerosol measurement instruments, which prevent artifacts such as loss of semi-volatile compounds, has led to the development of the MARGA by the Enegy Research Centre of The Netherlands (Petten, The Netherlands). An online analyzer of Monitoring for AeRosols and Gases (MARGA) was employed to measure major water-soluble (WS) inorganic ions in aerosols in PM_{10} or $PM_{2.5}$ at 1-h time resolution in Shanghai, Hangzhou from 2009-2012. The 1 hourly data of the inorganic fraction of aerosol i.e. Na^+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, NO_3^-, SO_4^{2-} and related gas phase concentrations NH_3, HCl, HNO_2, HNO_3, SO_2 were measured using IC (Metrohm) systems. During the observation, hazy days were encountered over which atmospheric visibility was commonly less than 10 km, and the hourly concentration of PM_{10} reached peak values of more than 150 μg m^{-3}. Based on WS ions and pollution gases, the haze events were classified as biosmoke, complicated and secondary pollutions depending on their distinct formation schemes. During the biosmoke pollution aroused from biomass burning plume, the concentration of water-soluble K^+ was increasing up to a maximum about 19 times higher than the average of clear days, and K^+ behaved a strong positive linear correlation with Cl^-. Because of sulfate and nitrate significantly enhanced by secondary production, in which precursor gases of SO_2 and NO_2 were converted into SO_4^{2-} and NO_3^- on the surface of pre-existing KCl particles, the complicated pollution was responsible for a combining

contribution of biosmoke aerosols directly transported from biomass burning emission and known secondary aerosols linked to local emission. Under high atmospheric oxidation ability and steady atmosphere condition, the secondary pollution resulted from a significant increase of sulfate and nitrate aerosols which were oxidized from large amounts of anthropogenic pollution gases of SO_2 and NO_2 in the urban atmosphere.

P-4-023 The impact of atmospheric aerosol extinction coefficient on oxidative stress of school children in a panel study in Beijing

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Key words
air pollution intervention atmospheric aerosol extinction coefficient urine malondialdehyde oxidative stress panel study

Background: Atmospheric aerosol extinction coefficient is accepted as an indicator for air pollution. On the other hand, malondialdehyde (MDA) has been suggested to be a highly sensitive biomarker used to measure oxidative stress. Seldom epidemiologic research shows the association between extinction coefficient and health outcomes.

Objectives: Associations between oxdiative stress in schoolchildren and atmospheric aerosol extinction coefficient are estimated before and during the air pollution intervention for 2008 Beijing Olympic Games.

Methods: We measured the urine malondialdehyde (UMDA) in 36 school children ages 9-12 years during 5 visits in 2 years, and extinction coefficient is obtained during the same time. We used generalized estimating equations to assess the relations, controlling body mass index, asthma, temperature, and relative humidity.

Results: In air pollutants, atmospheric aerosol extinction coefficient can predict the concentrations of black carbon, $PM_{2.5}$, SO_2 and NO_x well. We found positive correlations of UMDA with atmospheric aerosol extinction coefficient.

Conclusions: Atmospheric aerosol extinction coefficient was significantly associated with oxidative stress in the panel study of school children in Beijing. Reduced air pollution levels during 2008 Olympics resulted less oxidative stress.

P-4-24 Development of a Mixtures Characterization Toolkit to estimate air pollution source impacts for time series epidemiologic analysis to investigate air quality and human health associations: Application to a High Biogenic and a High Industrial Emissions Areas

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Key words
Health Effects Exposure Characterization

Historically, epidemiologic analysis of the impact of air pollution on health has relied on measurements of pollutant concentrations, using data from regulatory monitoring networks maintained through government agencies. However, these data are limited spatially and temporally. Moreover, these data cannot directly identify the sources of the pollution, or other health-relevant pollutant mixtures. In-depth analysis of these data, including the use of models to characterize pollutant mixtures, sources and species impacting both individual locations and wider areas may be beneficial for health studies whose goal is to assess the health impacts of pollutant mixtures. As part of the Southeastern Center for Air Pollution and Epidemiology, a Mixtures Characterization (MC) Toolkit is being developed to effectively analyze air pollution and air quality modeling data to better understand how emission sources combine to impact air quality and to provide metrics for use in health assessments. In addition to source characterization, exposure estimates are a knowledge gap in health studies, thus the MC Toolkit includes spatial averaging and temporal interpolation components to improve the concentration distribution modeling.

Data analysis is done to calculate spatial air pollution metrics using both area and population weighted averages. Receptor oriented SA methods (CMB, PMF) are applied to the data to determine source impacts. Source oriented air quality models are also utilized in the MC Toolkit, including a chemical transport model (CMAQ). Additionally, emissions modeling (SMOKE) is used to generate a spatial and temporal allocation of the source emissions. Two novel SA techniques are applied, the first an ensemble approach that generates new source profiles for CMB based on an ensemble trained approach that includes results from CMB,

PMF and CMAQ. The second is a hybrid between CMAQ and observations in a CMB-approach to scale the original CMAQ source impacts.
This work will present results from St. Louis, Missouri and Atlanta, Georgia, where the MC Toolkit is being applied to data collected from regulatory and special monitoring networks to generate source impact estimates for acute health effects studies (e.g., time-series studies of emergency department visits). St. Louis is unique due to the varying emissions from point sources, e.g., chemicals manufacturing and metals processing. Atlanta is impacted by a very different set of sources, including large biogenic emissions that interact with mobile source and power plant emissions and lead to a large amount of secondary organic aerosol formation. Future work includes the application of the MC Toolkit to other cities with corresponding health data to investigate the extent of city-to-city variability in pollutant mixtures and associations with health outcomes. Tools and approaches to address the uncertainty due to the spatial variability of the source impacts are also being incorporated.

P-4-25 Contribution of water-soluble and insoluble species and their hydrophobic/hydrophilic sub-fractions in the toxicological properties of ambient atmospheric aerosols

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Key words
Hydrophobic organic aerosol DTT assay Ecotoxicity

The present study assesses the relative contribution of water soluble and insoluble compounds and their hydrophobic/hydrophilic sub-fractions in the toxicological properties of ambient fine aerosols (Dp<2.5 μm). The toxicity of the particles was measured by both chemical (dithiothreitol; DTT assay) and biological methods (acute toxicity test using aquatic organisms - Brachionus calyciflorus). The particles were collected at urban site near downtown Atlanta using a high-volume sampler and were extracted in both water and methanol. Hydrophobic and hydrophilic fractions were segregated by passing the extracts through a C-18 column. The responses of both toxicity assays (DTT and rotifers) were significantly higher for the methanol extract than water extract. A substantial fraction of toxicity was associated with the hydrophobic compounds as evident from a remarkable attenuation in DTT response (~60 % for water and ~75 % for methanol extract) after passing the PM extracts through C-18 column. The DTT

activity of water and methanol extracts were correlated with the WSOC (water soluble organic carbon; R = 0.86) and WIOC (water insoluble organic carbon; R = 0.94) content of the PM, respectively. Brown carbon (BrC) content of the PM, which predominantly represents the hydrophobic organic fraction, was also correlated with DTT activity of both water (R=0.78) and methanol extracts (R=0.83). On the contrary, regression results of the rotifer assay didn't yield any consistent association with the measured PM constituents; however, considerable reductions (>80%) were obtained in the rotifers mortality also by removing the hydrophobic PM species. These findings suggest that the insoluble organic aerosol, especially with high hydrophobicity, contribute substantially to the toxicity of aerosols and could have significant ecological impacts (e.g. on rotifers and other important animals in aquatic food webs). More research should be focused on this fraction than done in the toxicological studies conducted so far.

P-4-026 Estimation of evapotranspiration and soil moisture from LANDSAT products using a modified surface energy balance algorithm (SEBAL)

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Key words
Evapotranspiration Soil moisture SEBAL Estimation

Nitrogen oxides (NO_x) are key catalysts in the atmospheric oxidation of carbon monoxide, methane, and volatile organic compounds through OH and other radicals. While fossil-fuel combustion is still the most important (and best documented) global NO_x source, there is considerable uncertainty about the range of soil biogenic NO emissions, particularly from semi-arid, arid, and hyper-arid regions (which cover approx. 40% of planet Earth's total land surface). Because soil moisture is the most important (macroscopic) factor which controls soil biogenic NO emission, estimations (simulations) of the soil water content of arid (agriculturally managed) areas, preferably on local/regional scales are necessary to support the quantification of soil NO emission (e.g. by up-scaling). Furthermore, evapotranspiration (ET) in arid regions, strongly controlled by soil moisture, is of highest importance for understanding and eventually intervening in the water cycle of natural arid systems.

In this study, a surface energy balance method which combines meteorological observations with spectral data derived from remote sensing measurements, was used to estimate the ET on the scale of Milan oasis (Xinjiang, R.R. China). Based

on areal information derived from LANDSAT TM5 images, SEBAL (modified Surface Energy Balance Algorithm for Land) was used for the simulation of net radiation, soil heat flux, sensible heat flux and ET at Milan oasis, where the predominant land use is cotton and the Jujube fields.
For individual satellite overpasses, actual ET was computed on pixel-by-pixel basis resulting in diel variations of ET. From this, a site-specific empirical relationship between soil moisture and ET was established which has been used to calculate diel variation of soil moisture.
Simulation results by SEBAL (ET, soil moisture, soil temperatures, etc.) for Milan oasis have been compared to observational data, such as energy fluxes (net radiation, sensible and latent heat fluxes), surface temperatures, soil moistures, etc. which have been obtained during the 2011 DEQNO field campaign. Simulated and observed data are in pretty good agreement. Ongoing studies are using SEBAL to support the generation of seasonal data sets of biogenic NO soil emission for Milan oasis (and other oases in Xinjiang).

P-4-027 Personal exposure of carbonyl compounds and BTEXs and their source characteristics in residential dwellings of Beijing city, China

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Key words
Indoor air pollution Carbonyl compounds BTEXs Personal exposure Source characteristics

Exposure to indoor chemical air pollutants expected to increase potentially the chance of developing cancer risk. Ambient levels of carbonyl compounds (formaldehyde, acetaldehyde, acrolein and acetone) and BTEXs (benzene, toluene and xylenes) compounds were monitored at seven residential locations (210 residential dwellings) in Xicheng district, Beijing, during the period of November-December 2009. The concentrations of formaldehyde, acetaldehyde, acrolein, acetone, benzene, toluene and xylenes were in the rage of 0.2-213.4 μg m^{-3}, 0.7-140.9μg m^{-3}, 0.1-25.2μg m^{-3}, 0.3-102.1μg m^{-3}, 1.0-47.5μg m^{-3}, 1.3-552.6μg m^{-3} and 0.2-150.6μg m^{-3}, respectively. At the 210 residential dwellings, the average concentrations of formaldehyde, acetaldehyde, acrolein, acetone, benzene, toluene and xylenes were 21.8μg m^{-3}, 16.3μg m^{-3}, 2.8μg m^{-3}, 10.7μg m^{-3}, 9.0μg m^{-3}, 32.1μg m^{-3} and 12.3μg m^{-3}, respectively. The statistical analyses and I/O (indoor concentration/outdoor concentration) ratios results showed that the main sources of formaldehyde, acetaldehyde, benzene and toluene at residential dwellings (renovation age <5 years) in this investigation were organic solvents of adhesives

and the decoration materials. Meanwhile, outdoor emission factors might have impact on acetone, acrolein and xylenes concentrations of indoor air at different dwellings and formaldehyde, acetaldehyde, benzene and toluene concentrations of indoor air at residential dwellings (renovation age >5 years). Furthermore, the human exposure cancer risks of formaldehyde, acetaldehyde and benzene were also discussed.

P-4-028 Genotoxic and Inflammatory Effects of Organic Extracts of Traffic Related Particles in Human A549 Lung Cells: The Roles of Quinones

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Key words: traffic-related particles quinones DNA damage inflammation

Background: Traffic-related particles contribute a lot to the urban air pollution and are found to be associated with adverse health effects. Quinones, as the most important oxygenated polycyclic aromatic hydrocarbon (PAH) derivates in the ambient air, are frequently found in diesel exhaust and traffic-related particles, and are believed to contribute to the harmful effects induced by particles through reactive oxygen species (ROS) generation. However, their effects on pro-inflammatory processes and genotoxic damages are less known. Objectives: The aim of the present study is to investigate whether exposure to organic extracts of particles collected in a road tunnel in Shanghai, China can cause genotoxic and inflammatory effects in human alveolar type-Ⅱ like epithelial cells (A549 cells), and try to reveal the roles of quinones. Methods: The organic extracts of particles (oTRP) collected in the Dapu tunnel of Shanghai and three typical quinones (anthroquione (AQ), 1,4-nathroquione (NQ) and benzoquione (BQ)) were exposed to A549 cells with different concentrations. Cell viability and plasma membrane damage were measured by MTT and LDHrelease assay respectively. Comet assay (sensitive to DNA strand breaks, direct oxidative DNA lesions and alkali-labile sites) was applied to evaluate the genotoxic impacts in A549 cells. Expressions of five inflammatory cytokines/chemokines and two CYP related enzyme genes were measured by real time PCR. The ROS generated in cells was measured by fluorescent microscope. The different effects of quinones were compared. Results: Significant dose-dependent effects in cell viability, LDH-release, DNA damage were observed in A549 cells after treated with oTRP for 24h. Pro-inflammatory genes, interleukin-6 (IL-6), interleukin-8 (IL-8), tumor necrosis factor α (TNF- α) and monocyte chemotactic protein-1 (mcp-1) and the two Ahr-regulated genes, cyp1a1 and 1b1 were significantly induced by oTRP, but not for chemokine (C-C motif) ligand 5 (ccl5). Increase ROS was also observed in A549 cells, suggesting oTRP generated its genotoxic and inflammatory impacts through the ROS mechanism. The three types of quinones showed completely different gene expression patterns in A549 cells. NQ induced gentle cyp1a1 and 1b1 expressions, either for AQ or BQ. NQ was a more potent inducer of cytokine/chemokine than

AQ and BQ in A549 cells. At the concentration of 10 μM, NQ could up-regulate IL-6, IL-8, TNF- α, mcp-1 and ccl5,while, AQ could only induce ccl5 and BQ could induce mpc-1. These results indicated that NQ, BQ and AQ could participate in the pro-inflammatory responses through different cytokine/chemokine release. Conclusions: These results confirm and extend previous studies indicating that organic part of traffic related particles has a considerable pro-inflammatory and genotoxic potential, finally causing lung impairment, and reveal the considerable contributions from certain quinones inside the particles.

P-4-029 Genome-wide gene expression profiling reveals maldevelopment of placenta associated with ambient particle exposure in an animal model

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Key words: placenta Ambient particle Adverse pregnancy outcomes exposure

Recent studies have suggested a link between inhaled particles exposure in urban areas
and susceptibility to adverse pregnancy outcomes; however, the precise mechanisms remain to be determined. To test the hypothesis that subchronic exposure to environmentally relevant particulate matter, even at low concentrations, potentiates maldevelopment of placenta and alters gene expression profiling of placenta in the pregnant animal model. We carried out twice of the exposure experiments by using pregnant rats during 2009/12/28~2010/3/31. Ten /six pregnant Sprague Dawley (SD) rats were exposed in two parallel chambers based on randomized assignments as exposure group (EG) and control group (CG), 24 hours per day, 7 days per week for 20 days. RNA-Seq was used to analyze the genome wide gene expression profiling (GWGEP) of the placentae with abnormal and normal fetus. Adverse pregnancy outcomes were happened in both of the twice exposure experiments in EG, and the ratio was near 6%, while the CG was zero. The GWGEP results showed that compared to the genes of placentae of CG, 1352/ 509 and 1797/ 544 genes were downregulated/
up-regulated respectively, of the placentae with abnormal fetus in the twice exposure
experiments. By the gene ontology and function analysis, we found that 1) the angiogenesis, vascular development and labyrinth genes were significantly changed, resulting in dysplasia in placental blood circulation; 2) the nutrient metabolism and transport-related gene changed, resulting in poor nutrient supplement; 3) the hemoglobin genes significantly reduced, resulting in decreased oxygen transport capacity, lack of oxygen supply. Our results suggested that ambient particle exposure altered labyrinth development, nutrient metabolism and hemoglobin formation concerned genes, potentiated maldevelopment of placentae, and induced adverse pregnant outcomes.

Poster Presentations, Session 5

P-5-001 Dry deposition of nitrogen compounds (NO_2, HNO_3, NH_3), sulfur dioxide and ozone in West and Central African ecosystems.

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Key words
gaseous concentrations, passive samplers, dry deposition velocities dry deposition fluxes, West and Central Africa, IDAF stations.

Wet and dry deposition of chemical species to the earth's surface plays an essential role in controlling the concentration of gases and aerosols in the troposphere. The study of deposition thus allows for tracing the temporal and spatial evolution of atmospheric chemistry and is a pertinent indicator for evaluating natural and anthropogenic influences. International programmes such GAW (Global Atmospheric Watch), part of WMO, emphasize that dry deposition data are scarce and generated using various methodologies in different parts of the world. The international IGBP/IGAC/DEBITS (Deposition of Biogeochemically Important Trace Species) program was initiated in 1995 in Africa (IDAF program) with the view to study changes in deposition characteristics over a longer time period.

This work is part of the IDAF program (IGAC/DEBITS/AFrica*) and is based on the long term monitoring of gases concentrations (1998-2007) established on seven remote sites representative of major African ecosystems. The sites are located to represent a transect of African ecosystems, i.e., dry savannas - wet savannas - forests. This original study is a contribution to the estimation of dry deposition fluxes at the scale of the African ecosystems. Dry deposition fluxes were estimated by the inferential method using on one hand surface measurement of gases concentrations (NO_2, HNO_3, NH_3, SO_2, and O_3) and on the other hand simulated dry deposition velocities (Vd). Vd were calculated using the "big leaf" model of Zhang et al. (2003b). In the model of deposition, surface and meteorological conditions specific to IDAF sites have been adapted and validated in order to simulate Vd representative of major African ecosystems. The monthly, seasonal and annual mean variations of gaseous dry deposition fluxes (NO_2, HNO_3, NH_3, O_3, and SO_2) are analyzed.

Along the latitudinal transect of ecosystems, the annual mean dry deposition fluxes of nitrogen compounds range from 0,4±0,0 to 0,8±0,2 $kgN.ha^{-1}.yr^{-1}$ for NO_2, from 0,7±0,1 to 1,0±0,3 $kgN.ha^{-1}.yr^{-1}$ for HNO_3, and from 2,3±0,8 to 6,5±2,3

kgN.ha^{-1}.yr^{-1} for NH_3 over the studied period (1998-2007). The total nitrogen dry deposition flux ($N\text{-}NO_2+N\text{-}HNO_3+N\text{-}NH_3$) is more important in forests (8,0-8,5 kgN.ha^{-1}.yr^{-1}) than in wet and dry savannas (3,4-5,3 kgN.ha^{-1}.yr^{-1}). NH3 dominated nitrogen dry deposition, representing 67-80% of the total. The annual mean dry deposition fluxes of ozone range between 11,3±4,7 and 17,5±3,0 kg.ha^{-1}.yr^{-1} in dry savannas, 17,5±3,0 and 19,2±2,9 kg.ha^{-1}.yr^{-1} in wet savannas, and 8,2±0,6 and 10,2±2,1 kg.ha^{-1}.yr^{-1} in forests. Lowest O_3 dry deposition fluxes in forests are correlated to low measured O_3 concentrations, lower of a factor of 2-3, compared to others ecosystems. Along the ecosystem transect, annual mean of SO_2 dry deposition fluxes present low values and a small variability (0,5 to 1 kgS.ha^{-1}.yr^{-1}).

P-5-002 Dimethyl sulfide emissions from soils and vegetation in central Amazonia

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Key words
DMS Amazon

Surface emissions of reduced sulphur compounds like dimethyl sulphide (DMS) may significantly impact global climate through the formation of secondary sulphate aerosols which can act as potent cloud condensation nuclei and play a role in the Earth,s radiative balance. While oceans are generally considered the dominant source of DMS, terrestrial ecosystems are increasingly recognized as a potentially important source. However, great uncertainty remains regarding the strength of the terrestrial DMS emissions as well as the components within the ecosystem that are responsible for the emissions. While some plants have been shown to emit significant amounts of DMS into the atmosphere, others don,t. Although microbial processes in soils and litter have also been shown to be a source of DMS emissions, the lack of field observations precludes an understanding of terrestrial DSM sources, especially in the remote tropics where high temperatures, moisture availability, and productivity may encourage strong DMS emissions. In this study, we present new high vertically resolved ambient concentrations (and fluxes) of DMS in the central Amazon together with individual plant flux and mesocosm concentration in the Biosphere 2 tropical rainforest mesocosm. We found that at Biosphere 2, seven tropical plants emitted DMS at significant rates (from 5 up to 10 ρmolm^{-2}s^{-1}) with ambient concentrations at up to 1 ppbv. Both ambient concentrations and plant emissions at Biosphere 2 exhibited strong diurnal patterns that tracked light and temperature. In the central Amazon, vertical gradients showed higher concentrations of DMS (up to 0.71

ppbv) generally at night but also sometimes during the day. Unlike other volatile organic compounds, like isoprene and monoterpenes that are emitted exclusively from the vegetation and whose concentrations peaked within the canopy, DMS concentrations peaked near the ground. Preliminary studies revealed direct emissions of DMS from l eaf litter, but also from higher plants. Therefore, our results suggest that at the field site studied, DMS emissions are dominated by soil and plant emissions, rather than from long range transport of oceanic DMS. These findings highlight the potential importance for strong biosphere-atmosphere coupling via the emissions of DMS from the Amazon Basin, since it has been demonstrated the important role of DMS on Secondary Organic Aerosol (SOA) formation into sulphate aerosols and its consecuent great Cloud Condensation Nucleai (CCN) yield, possibly affecting precipitation dynamics in the Amazon.

P-5-003 Estimating the net ecosystem exchange for the major forests in the northern United States by integrating MODIS and AmeriFlux data

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Key words
NEE MODIS

The eddy covariance technique provides long-term continuous monitoring of site-specific net ecosystem exchange of CO_2 (NEE) across a large range of forest types. However, these NEE estimates only represent fluxes at the scale of the tower footprint and need to be scaled up to quantify NEE over regions or continents. In the present study, we expanded a method developed previously and generated a new NEE model exclusively based on the Moderate Resolution Imaging Spectroradiometer (MODIS) products, including enhanced vegetation index (EVI), land surface water index (LSWI), land surface temperature (LST) and Terra nighttime LST'. This method, in our previous research, provided substantially good predictions of NEE and well reflected the seasonal dynamics of the deciduous broadleaf forest at the Harvard forest site. Studying NEE of forests in the middle-latitude regions of the Northern Hemisphere is significant because it may help to understand the 'missing carbon sink' from terrestrial ecosystems.

In this study we selected eight eddy flux sites (conifer forest, 4; broadleaf forest, 4) to represent the major forest ecosystems in the northern United States. The sites are as follows: Blodgett Forest, Howland Forest, Niwot Ridge Forest, Morgan Monroe State Forest, Harvard Forest Site, Willow Creek, Metolius Intermediate Pine,

University of Michigan Biological Station. The last two sites are used for independent validation of the general models of evergreen needleleaf forest (ENF) and deciduous broadleaf forest (DBF). Both MODIS products and EC data are obtained at 8-day time scale. After analyzing the correlation between them, based on multiple linear regression method, the optimum model was selected with the maximum R2 and the minimum RMSE. Compared with the model based on a single flux site, we also established the general models that apply to ENF and DBF, respectively. The results showed that our simpler model based entirely on MODIS products promised well to estimate NEE and could capture the broad trends of seasonal dynamics of NEE. Moreover, the general models predicted NEE at the biome level well except large overestimation of annual mean carbon uptake and fairly constant underestimation of NEE during summer months for DBF, the assessment of ENF was rather coincident with measured NEE. The percentage deviations of the agreement between modeled and measured NEE for DBF and ENF were 44.4% and 5.5%, respectively. In the end, we also validated both general models for ENF and DBF using the independent flux sites. It demonstrated this method performed well for estimating NEE.

Although in this study we selected eight flux sites to represent the major forest ecosystems that included most of the available sites in the network, some geographical regions, community composition, and different forest ages (young, middle aged and mature) of the same forest ecosystem are still underrepresented, which may affect the universality of the general models. In future research, we could combine AmeriFlux, AsiaFlux, EuroFlux to explore the potential mechanism embedded under the environmental controls and the carbon, water fluxes of terrestrial ecosystem.

P-5-004 Mercury cycling in the Yellow Sea: the importance of atmospheric processes

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Key words
Mercury Yellow Sea

The Yellow Sea is located between East China and the Korean Peninsula, which is an important and growing emission source of anthropogenic mercury (Hg). This marine system is then a potentially important receptor for Hg emissions from East Asia. In this study, based on the carefully evaluated field data, modeling results and reasonable assumptions, we develop the first Hg mass balance for the Yellow Sea to obtain insights into the sources, sinks, pathways and processes regulating the Hg cycle in this marine system. The following terms are taken into account: Hg

pool in the water column, Hg exchange with the adjacent seas, Hg input via atmospheric deposition, rivers discharge and coastal erosion, and Hg output via elemental Hg (Hg(0)) evasion from sea surface, sedimentation and fish harvesting. The result suggests that the atmospheric processes play an important role in the Yellow Sea Hg cycle. The primary Hg input to the Yellow Sea comes from the atmospheric deposition (51.7 kmol yr^{-1} or 52% of input) and the primary Hg output is the Hg(0) evasion from sea surface (49.0 kmol yr^{-1} or 56% of output). Notably, the fluxes of atmospheric deposition (51.7 kmol yr^{-1}) and Hg(0) evasion (49.0 kmol yr^{-1}) are similar. This finding seems to support the hypothesis that in the aquatic Hg originated from atmospheric deposition is more available for biological and photochemical transformation to Hg(0). This pattern of mass balance for the Yellow Sea implies that a rapid reduction and reemission of deposited Hg from atmosphere may take place in the water column. However, this hypothesis needs to be confirmed by future research. The result of mass balance demonstrates that the input and output of Hg shows a relatively large imbalance with a net increase of 12.2 kmol yr^{-1}, indicating ~ 5.7% annual increase of Hg in the water column. Interestingly, this increase is very close to that of anthropogenic atmospheric Hg emission from China (3–6% per year), suggesting the trend of Hg burden in the water column of the Yellow Sea may reflect the growing trend of atmospheric Hg release induced by human activity in China/East Asia, although it is noted that the process from atmospheric emission to water accumulate in downwind and downstream is non-linear relationship. The mass balance and its uncertainties, the future change of seawater Hg levels and research priorities are discussed in detail in the presentation.

P-5-005 Atmospheric processing of dust and bioavailable iron deposition to the ocean

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Key words
biogeochemical cycle air-sea flux

Iron (Fe) and phosphorus (P) in the atmospheric dust have been shown to affect the primary productivity and ocean biogeochemistry. The impact of atmospheric supply of nutrient Fe and phosphorus on ocean biogeochemistry and thus the global climate change is dependent on the solubility of the nutrients in the dust. Fe solubility (dissolved to total Fe, FFS) in the atmospheric aerosols has been reported to range from 0.1% to 80%. However, FFS is usually less than 0.5% in the non-atmospherically-processed dust, suggesting that FFS can be enhanced by atmospheric processes. Apart from the process of mixing with combustion aerosols, which have higher Fe solubility, there are several other atmospheric processes which have been hypothesized to increase the Fe solubility during dust transport. We have investigated how different atmospheric processes may have increased the solubility and thus bioavailable deposition flux of Fe in dust during its transport by combining laboratory geochemical simulations and measurements

with global aerosol modelling. We demonstrated that size sorting, which was hypothesized to be an important process leading to observed increased Fe solubility during transport, is not important. We also showed that cloud processing itself is not important due to the high pH (usually >4) of the cloud water. Indeed, cloud processing could potentially decrease the Fe solubility in the dust as a result of the formation of Fe nanoparticles. On the other hand, acids in the aqueous water around aged dust particles have the potential to increase Fe solubility as a result of the high SO_4^+ and H^+ concentrations in the dust. However, acid processing of Fe in the dust is complex owing to the extremely high ionic strength in the aerosol water and extremely low amount of water per mass of dust. Further geochemical laboratory studies on the geochemical solubility and dissolution kinetics of Fe in the dust under realistic conditions, field measurements on the acidity of water around and Fe solubility in aged dust, and modelling works are needed to increase the ability of atmospheric models to quantify the atmospheric processing of Fe in the dust.

P-5-006 How much CO was emitted by the 2010 fires around Moscow?

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Key words
Biomass Burning Inverse modeling

The fires around Moscow in 2010 emitted a large amount of pollutants to the atmosphere. Here we estimate the CO source strength of the fires in July and August by using the TM5-4DVAR system in combination with CO column observations of the IASI instrument on the METOP satellite. We use a dedicated prior CO emission inventory that is based on burning scar observations from MERIS and MODIS. The vertical distribution of the emissions is derived from AATSR observations. It is shown that the IASI observations provide a strong constraint on the total emissions needed in the model. Irrespective of the prior emissions used, the total CO fire emission estimates in the study period amount to approximately 18 Tg CO, substantially higher than the bottom-up inventories. This is attributed to the widespread peat fires that are hard to detect using the burning scar methodology.

P-5-007 Evaluation of different ocean color products as predictive tools of the organic mass fraction in submicron sea spray

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Key words
ocean-atmosphere interaction sea sparay primary organic aerosol

Initial efforts towards developing a combined organic-inorganic sea-spray source function parameterization for large-scale models utilized chlorophyll-a (Chl-a) and wind speed as input parameters to combine oceanic biology and atmospheric dynamics. These first studies, performed using satellite derived Chl-a fields with low temporal resolution (monthly averages), reported only a modest correlation coefficient (at best 0.55) between Chl-a and organic matter enrichment in sea-spray, suggesting that Chl-a is only partially suitable for this aim.

In order to check if better biological activity surrogates are available from satellite measurements, the following research activity has been carried on. Submicron marine aerosol chemical composition data collected at Mace Head (Ireland) from 2002 to 2009, for a total of 52 samples, have been used to calculate the organic mass fraction of submicron sea spray. Daily satellite ocean color data, with 1 degree spatial resolution, were obtained from the GlobColour ESA project.

A reconstructed Chl-a field of the North-Atlantic Ocean, based on daily data, revealed an improved correlation of 0.73 between the fractional mass contribution of organics in sea-spray and Chl-a concentration. A similar analysis, using other satellite products as colored dissolved and detrital organic materials (CDM) absorption and particulate organic carbon (POC) concentration, revealed slightly lower correlation coefficients (0.70 and 0.69). These results indicate that, to date, Chl-a is the best biological surrogate for predicting sea-spray organic enrichment. In fact, considering the minimal difference between the correlation coefficients obtained with the three ocean color products, there is no reason to substitute Chl-a, which is the most accurate parameter obtained from ocean color data, with other biological surrogates which in general can be affected by larger and less known errors.

This work has evidenced a systematic delay of 7-8 days between Chl-a, CDM and

POC time series and the organic matter enrichment in sea spray that cannot be justified by the aerosol transport time and that, likely, is related to the timescale of the biological processes responsible for the production of transferable organic materials during the bloom evolution. This time delay should be taken into account when modeling the production of primary organic aerosols from the oceans based on satellite data, even though it is not clear if this result holds also for other oceanic regions.

A new relationship describing the organic enrichment of sea spray aerosol, as a function of both wind speed and sea surface Chl-a concentration, to be implemented in large scale models, is proposed as a conclusion of this study.

P-5-008 Ozone deposition to bare soil: analysis of the dependence of the soil resistance on the surface relative humidity for different soil types

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Key words
ozone soil resistance dry deposition

Tropospheric ozone (O_3) is mainly formed by photochemical reactions involving nitrogen oxides (NOx) and volatile organic compounds (VOCs). It is a well known greenhouse gas responsible for positive radiative forcing, a key compound of atmospheric chemistry and a harmful pollutant responsible for damages on plants, materials and human health.

Deposition to the Earth' surface represents the only net sink for O_3. Since O_3 is hardly soluble in water, it is deposited mainly through dry deposition to terrestrial ecosystems, and not to oceans or lakes. Despite this knowledge, the strength of this dry deposition sink is highly uncertain, with current estimates ranging between 530 and 1470 Tg O_3 y^{-1}. It is well established that deposition to terrestrial ecosystems occurs through three pathways: stomatal uptake by plants, cuticular deposition and soil deposition. The environmental parameters controlling stomatal and cuticular pathways are reasonably identified and parameterisations to model these pathways are available. However, the soil pathway is still not fully understood: only few studies dealt with O_3 deposition to soil and current parameterisations of soil resistance to O_3 assume a constant resistance at the weekly or monthly scale.

However, this pathway cannot be neglected, especially for agricultural ecosystems, for which we recently showed that soil deposition could accounts for 25% of the total O_3 deposition when crops were fully developed (data obtained from an agricultural field near Paris, France). In addition, we observed that O_3 deposition during bare soil periods was of the same magnitude as O_3 deposition over fully developed canopies.

In this study, we explore the parameters controlling the soil resistance to O_3 deposition. Ozone fluxes as well as heat fluxes, standard meteorological conditions (e.g. air temperature, air relative humidity) and soil parameters (e.g. texture, porosity) were measured at four different sites in France with contrasting soil types. The soil resistance to ozone, deduced fron measured deposition velocity and estimated aerodynamic and quasi-laminar boundary layer resistances, was expressed as a function of relative humidity at the soil surface (RHsurf) for each dataset as Rsoil = Rsoilmin x e(k x RHsurf), where Rsoilmin ($s\ m^{-1}$) is the soil resistance without water adsorbed at the surface (i.e. at RHsurf = 0%) and k is an empirical coefficient of the exponential function. The link between the soil properties and the two parameters Rsoilmin and k is then explored.

Our results show that (i) ozone deposition to soil decreases when relative humidity increases and (ii) the parameterisation of Rsoil depends on soil properties. A possible underlying process is the adsorption of water at the soil surface which decreases the surface available for ozone deposition. To establish a parameterisation of Rsoil that could be used for any site, it is necessary to take into account this phenomenon in a mechanistic way. This study is the first step to achieve this goal. We will also attempt to expand our analyses to other locations.

P-5-009 Long-term historical inventory of isoprene emissions over Asia : interannual variability, trends and evaluation against flux measurements

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Key words
isoprene emissions interannual variability Asia

We build a 32-year (1979-2010) inventory of isoprene emissions over east Asia at a resolution of 0.5 degree between 1979 and 2010 based on the Model of Emissions of Gases and Aerosols from Nature (MEGAN, Guenther et al., Atmos. Chem. Phys., 2006) version 2 and the Model for Hydrocarbon Emissions by the Canopy (MOHYCAN, Muller et al., Atmos. Chem. Phys., 2008) used to determine leaf temperature and the radiation fluxes dependence on the canopy height. MEGAN uses the latest compilation of basal emission factors for isoprene. The canopy model is driven by meteorological fields for air temperature, cloud cover, downward solar radiation, wind speed, and soil moisture provided by the ERA

Interim analyses of the ECMWF Data Center, and leaf area index is obtained from collection 5 8-day MODIS data. A positive long-term trend in isoprene fluxes, estimated at ca. 0.5% annually between 1979 and 2010, is estimated from our analysis. Although the calculated emission trend is mostly driven by the positive trend in air temperature observed throughout the 32-year period in Asia, the contribution of other environmental factors to the observed interannual variability and trend are investigated and analyzed. The potential impact of land use changes is not accounted in this study. Further, the comparison between the standard isoprene fluxes and those derived using the alternative simplified Parameterized Canopy Environment Emission Activity algorithm (PCEEA) reveals large differences, often by up to a factor of two, which are analysed in detail. The emission inventory is evaluated against flux measurement campaigns realized in southern Yunnan province in China (Baker et al., Atmos. Environ., 2005), as well as the OP3 campaign over a Malaysian tropical rainforest in 2008 (Langford et al., Atmos. Chem. Phys., 2010).

P-5-010 Stable carbon isotope ratios in atmospheric methanol

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Key words
stable carbon isotopes methanol

Methanol is the most abundant oxyganted VOC in the troposphere. Thus, methanol has a significant impact on tropospheric chemistry. It is a sink for hydroxyl radicals, but can also increase the concentration of oxidants in the atmosphere depending on the chemical situation. Despite its importance, sources, sinks and the atmospheric cycle of methanol are still not fully understood. Chemistry and transport models have to deal with large uncertainties in emission inventories and unresolved photochemical mechanisms.

Measurements of stable carbon isotope ratios (delta13C) have been successfully applied to other trace gases and improved our insights in their atmospheric budgets. In the same way we will use measurements of the stable carbon isotope ratios of methanol to investigate its role in atmospheric chemistry and its atmospheric budget. Knowing the delta 13C values of methanol allows to trace chemical pathways, determine the photochemical aging and help to understand the sources and sinks of methanol. The increasing number of laboratory investigations regarding the biogenic emissions of methanol will help to identify possible sources of methanol from measurements in ambient air.

Here we present delta 13C measurements of methanol from whole air samples obtained during airborne and ground based field campaigns in Spain and Germany. The samples have been collected with a custom made automated sampling unit aboard a Zeppelin NT over southern Germany and a CASA C-212-200 airplane over Spain. Included in this study are also whole air samples from the new sampling unit MIRAH (Measurements of Isotope Ratios in the Atmosphere on HALO), which was operated on the new German research aircraft during a technical mission in summer 2010 and during the ground based campaign PARADE (Particles And Radicals: Diel observations of the impact of urban and biogenic Emissions) at the Taunus observatory on the summit of Kleiner Feldberg, Germany, about 10 km north of Frankfurt in late summer 2011.

In general, the stable isotope ratios suggest a mainly biogenic origin of methanol. On average, the isotope ratios were around - 45 ‰. In contrast to the wide range of delta 13C values found in laboratory studies for different sources (- 80 ‰ to - 15 ‰), the isotope ratios measured in all field experiments varied significantly less. The largest variations were found during the airborne campaigns. During the aircraft campaign over Spain methanol isotope ratios ranged from - 50 ‰ to - 35 ‰ with no significant differences between urban and rural air masses. However, the variations decreased with increasing altitude in the planetary boundary layer. During the Zeppelin campaign methanol isotope ratios ranged from - 44 ‰ to - 22 ‰ and showed a pronounced bimodal distribution. This might be an indication that the observed methanol mixing ratios were either due to the contribution of completely different sources or to the mixing of different photochemically aged air masses.

P-5-011 Ship-borne measurements of greenhouse gases and ozone in the Southern hemisphere marine boundary layer

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Key words
Marine atmosphere trace gases

Climate change is one of the most pressing global environmental issues of our time. It is driven by atmospheric change, and in particular by the large growth in greenhouse gases. There have been a great number of measurement campaigns focused on the Northern Hemisphere, however the data coverage in the Australasian region is sparse. Here we present measurements of trace gas concentrations that will be made in the marine boundary layer by a continuous in-situ FTIR system and ozone monitor during two ship cruises. We will present the distribution of key greenhouse gases CH_4, CO_2, N_2O as well as CO and O_3 across the latitudinal and longitudinal transect made by the research vessel

“Southern Surveyor”.

During the first transect from Hobart (42.5 S, 147.2 E) to Brisbane (27.3S, 153.1 E) in April 2012 we hope to sample air influenced by terrestrial sources from Australia, such as pollution plumes originating from biomass burning or large metropolitan areas. These measurements will help to characterise the influence of such sources on greenhouse gas concentrations and ozone levels.

Southern Hemisphere marine background conditions over the Pacific ocean will be characterised during the second transect from Brisbane to Fiji (17.6 S, 177.5 E). The concentrations measured on both transects will be compared with chemistry transport model simulations to test the capacity of the models to describe adequately the sources, sinks and the chemistry affecting the marine Australasian Pacific boundary layer.

P-5-012 CO_2 effluxes from seasonal snow pack in a mid-latitude forest: profile gradient measurement through an automated tower

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Key words

CO_2 efflux Snowpack Profile gradient Wind pumping

As soil respiration plays an important role in the carbon cycle of the terrestrial biosphere, quantitative evaluation of CO_2 flux from soil is necessary for understanding the net flux of carbon between terrestrial ecosystems and the atmosphere. A bunch of researchers are focusing on soil respiration in the growing season as being dominated by the microorganism activities. It had been realized recently that soil CO_2 effluxes through the seasonal snowpack in the mid-to-high latitude winter contribute non-negligibly to the yearly biosphere-atmospheric carbon exchange. However, due to the difficult nature of accessing and reaching to the snow-covered field, researches on winter soil efflux under the snowpack were quite limited. In this study, we deployed an automated sampling-measurement system to estimate CO_2 efflux dynamics and quantity in a forest dominated by Todo-fir (Abies Sachalinensis) at Rishiri Island (45°07′N, 141°12′E), northernmost Japan.

Before the winter of 2010-2011, a profile gradient tower was constructed in the natural snow-falling field using stainless and aluminum frames. A series of gas samplers were set at 5 layers (10cm, 30cm, 60cm, 90cm, 120cm) of the snowpack spanning from the soil surface to the ambient air to investigate the snowpack CO_2 profile. Interstitial air was sampled at each layer every 2 hours and introduced to an NDIR analyzer that was placed in an indoor laboratory to determine the CO_2 concentrations. Snowpack temperature profile was simultaneously measured. Other environmental parameters such as soil temperature and soil water content; ambient temperature, pressure was also measured. A whole snow-cover season

CO_2 efflux was estimated based on the gas diffusion method.

An average apparent soil CO_2 flux of 1.03 g C m^{-2} d^{-1} (0.27 umol m^{-2} s^{-1}) was estimated for an 84-day period when the snow cover is deeper than 60cm, which resulted in a total amount of 83.41 g C m^{-2}. A general increasing trend, interrupted by several sharp decreasing events, characterized the seasonal dynamic of CO_2 flux. When soil temperature and moisture remained relatively steady, in which soil CO_2 flux was considered constant for the period of Day of Year 7-15; wind speed, which is associated with ambient pressure, had large influence on the apparent soil CO_2 flux estimation ($y = 0.005x2 - 0.0775x + 0.3424$, $R^2 = 0.74$, where y denoting soil CO_2 flux and x denoting wind speed) through pumping effect. The corrected soil CO_2 flux (1.30 g C m^{-2} d^{-1} , or 0.34 umol m^{-2} s^{-1}) when wind speed is zero, accounts to 273% of the apparent averages during this period. The results provide evidence for the quantitative estimate of winter carbon emissions in the annual carbon cycle in the mid-latitude forest.

P-5-013 Surface-atmosphere exchange of reactive nitrogen compounds

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Key words

exchange reactive nitrogen

We will present our recent results about the surface atmosphere-exchange fluxes of reactive nitrogen compounds (Nr). These include nitrogen monoxide (NO), nitrogen dioxide (NO_2), ammonia (NH_3), nitric acid (HNO_3), nitrous acid (HONO) and peroxyacetyl nitrate (PAN). Exchange fluxes of Nr are influenced by turbulence-chemistry interactions at the soil-vegetation-atmosphere interfaces. In and above tall canopies (forest) the magnitude and direction of Nr fluxes are controlled by physicochemical and/or surface characteristics as well as by the interaction of turbulent transport and chemical reactions of trace compounds. We have investigated these processes at a natural (nutrient-poor) grassland ecosystem using eddy covariance, relaxed eddy accumulation, aerodynamic gradient and chamber methods. The residence time of the air within the grass canopy was investigated using the Radon tracer technique.

We found that due to chemical divergence within and above the canopy exchange fluxes above the canopy may not be comparable to the fluxes at the surface also for low vegetation, which is in accordance with results for forests. Hence, the investigation of chemistry in combination with turbulence within low vegetation is crucial to study surface emissions and uptake. Based on rigorous error estimates we show that for natural grassland ecosystems and low ambient concentrations the capability of analytical techniques to accurately determine exchange fluxes and related parameters is limited. We will compare the results of the grassland ecosystem to results from a forest. We also found that HONO is released from soil as a function of temperature and soil water content, comparable to typically observed soil biogenic NO emissions. For natural ecosystems this implies that deposited Nr is re-cycled to the atmosphere.

P-5-014 An automated measurement system to investigate turbulence-chemistry interactions within a grassland canopy

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Key words
turbulence chemistry grassland canopy timescale

We developed an automated system for measuring a) simultaneous vertical profiles (three levels) of Radon (222Rn (here Rn)) and Thoron (220Rn (here Tn)), and b) soil fluxes of Rn and CO_2 with three static chambers. For the Rn/Tn profiles three Rn/Tn-discriminative RAD7 monitors (Durridge, USA) were used. The static chambers were sequentially closed, and the measurements of Rn and CO_2 were performed using an ALPHAGUARD (Saphymo, Germany) and a GMT222 (Vaisala, Finland), respectively. This system was complemented by simultaneous profile measurements of NO, NO_2, O_3, CO_2, and H_2O. Measurements were performed during two month (28th of July – 26th of September 2011) on the estate of the Mainz Finthen Airport (Mainz, Germany (49.969° N, 8.148° E)). The vegetation type at the site is classified as a nutrient-poor steppe-like grassland ecosystem. The inlets of the Rn/Tn and trace gas profiles were installed within the canopy at 0.04m, 0.20m and 0.80m above ground level.

We will present the setup of our Rn/Tn system and investigate the performance. The residence time of the air (τturb) within the grass canopy will be determined at different levels including rigorous error estimation. Additionally, we will present calculated bulk diffusion coefficients (Kbulk) within the canopy by using the Rn and CO_2 profiles in combination with the soil fluxes. We will compare τturb and the chemical conversion timescales (τchem) within the NO-NO_2-O_3-triad represented by the Damköhler numbers (DA = τturb / τchem). Using this method, the influence of chemical divergence on exchange fluxes within and above the canopy can be studied.

P-5-015 Laboratory based studies of primary sea-spray generation in plankton-enriched sea-water

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Key words
plankton sea spray organic matter

Marine aerosol enrichment by biogenic organic matter (OM) has been linked to phytoplankton activity (O’Dowd et al., 2004), thus having a strong seasonal impact on both the Earth’s albedo and climate. In addition to a seasonal cycle, sea-spray generation and its enrichment with OM is a very dynamic process producing regular OM plumes over N.E. Atlantic (Ovadnevaite et al., 2011). Plankton-enriched seawater contains a complex mixture of dissolved and particulate organic carbon components (POC and DOC) producing both water

soluble and insoluble organic aerosol species (Facchini et al., 2008; Russell et al., 2010); this warrants detailed laboratory studies aimed at establishing a link between observed ambient aerosol OM and its very primary form (e.g. Fuentes et al., 2010).

Laboratory studies using the microalgal species Emiliania huxleyi, Leptocylindrus danicus and Cylindrotheca closterium were performed by on-line and off-line analytical techniques using a sea spray production chamber and an ageing chamber with day-light and ozone. Under controlled conditions a sea spray highly enriched in OM was produced with levels similar to Facchini et al. (2008) over a period of 48 hours.

HR-ToF-AMS, 1HNMR and HTDMA techniques confirmed OM composition of highly hydrocarbon-like, water insoluble OM characteristic of unsaturated lipids exhibiting low hygroscopic growth factor. Freshly produced OM, while largely insoluble, was far less oxidised (less sugars) than the OM reported by Facchini et al. (2008). The HTDMA measurements revealed a gradual change in physical properties as contribution of OM increased along with a certain degree of external mixture.

Processing primary sea spray with light and/or ozone caused a gradual change in chemical composition (preserving low GF and slowly increasing O/C ratio) supporting primary origin and atmospheric evolution of observed ambient OM while not excluding trophic level interactions.

P-5-016 The effects of VSLS halocarbons and sea salt bromine on the stratospheric bromine and ozone losses -a study in UKCA

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Key words
halocarbons bromine

We have implemented an updated halogen chemistry scheme in UKCA model (a troposphere-stratosphere CCM). The tropospheric bromine chemistry was based on our previous work in the p-TOMCAT model (a global tropospheric CTM) with updated emission fluxes of the two key very short lived substances (VSLS), $CHBr_3$ and CH_2Br_2 based on our on-going continuous observations in South-East Asia. The stratospheric halogen chemistry in the UKCA model was also updated by introducing heterogeneous Br-Cl reactions on both polar stratospheric clouds (PSCs) and sulfate aerosols. The simulated concentrations of the VSLS halocarbons are compared with air-borne observations in order to validate our model. The modeled column BrO is compared with satellite observation to evaluate the model ability in reproducing the atmospheric background BrO as well as the springtime elevated-BrO over polar sea ice zones. We estimate the mount of VSLS halocarbons transported through the tropopause into the stratosphere as well as the amount of transported inorganic bromine (from degradation of halocarbons and sea salt bromine-depletion process). We then estimate their effects on the stratospheric ozone in both a present and future climate.

P-5-017 Investigating the effect of crop land expansion on tropospheric oxidising capacity with the UM-UKCA model

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Key words
Air quality Land use change/crop emissions isoprene chemistry

With the global population set to rise over the next 100 years, the fraction of land used for crop cultivation is likely to increase, the trend being most pronounced in developing regions such as Brazil and South East Asia. In these regions currently there stands natural rainforest, a high emitter of isoprene. As many staple crops, such as soy bean, are low emitters of isoprene, increasing the crop fraction in these regions will decrease isoprene emissions. Ozone over ~35 ppb has been shown to be damaging to plants, and as ground level ozone is sensitive to isoprene concentrations, altering isoprene emissions could increase ground level ozone, potentially resulting in crop damage. This mechanism was investigated by comparing two configurations of an atmospheric chemistry-climate model (UM-UKCA) under a 2100 climate following the A1B SRES scenario. The first run had a present day crop distribution but isoprene emissions concurrent with 2100 temperatures and general climate. The second run had isoprene emissions representative of both a 2100 climate and a 2100 crop distribution in accordance with the IMAGE model following the A2 SRES scenario. By comparing these runs it was established that ozone increased by up to 8 ppb (~30%) due to crop land expansion. Over the Amazon (the most affected region) it was found that crops were exposed to a daily maximum 8-hour (DM8H) ozone above the 35 ppb threshold for up to 65 days more per year than in the base case. These conclusions suggest that increasing the crop fraction in current areas of natural rainforest could increase regional ground level ozone, having a significant negative effect on crop yield and air quality.

The sensitivity of the conclusions to isoprene chemistry was examined by varying the isoprene chemistry scheme within the model. The CheT isoprene scheme used here (50 reactions) was compared with the AQUM (23 reactions) and CESM Superfast (2 reactions) isoprene schemes, all of which are currently used in

Earth-system models. It was found that the effect of transplanting these isoprene schemes into the CheT model, lead in both cases to higher ozone over isoprene rich regions by up to ~40 ppb (50%). Furthermore, upon repeating the land use change experiment with these other isoprene schemes, it was found that the AQUM scheme produced more ozone (up to ~20 ppb more) in isoprene rich regions due to crop expansion than CheT. However the CESM scheme showed the opposite effect, producing less ozone than the CheT scheme in isoprene-rich regions. These varied responses highlight the sensitivity of conclusions about future trends in surface ozone with respect to isoprene chemistry within the range of some currently used chemical schemes, and suggest that further research is needed into effective parameterisation of this complex chemistry.

P-5-018 Thailand Emissions Inventory from 2005 to 2010: A Major Update on Fugitive Dust and VOCs

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Key words
Thailand Emissions Fugitive VOCs Dust

In this study, we updated Thailand Emissions Inventory from the year 2005 to 2010. Rather than anthropogenic and natural (biogenic) emission sources, we considered about Fugitive Emission Source (FES) as one of the major concern and separated into another key source. FES can be classified by the contribution of Fugitive (Non Methane) Volatile Organic Compounds (FVOCs) and Fugitive Dust (FD).

For FVOCs, we reclassified fuel sale in gas station, printing businesses, and oil storage tanks out of the original anthropogenic sources that they contributed a large amount of annual FVOCs. Thus, FVOCs was comparable to anthropogenic VOCs from this context. New types of fuels were introduced in service and distribute on public (i.e., gasohol mixture, liquid petrochemical gas, and compressed natural gas) as well as their storages. Registered vehicle garages and car maintenance services from Original Equipment Manufacturer (OEM) were only included; therefore, FVOCs leakage was carefully in control. Petrochemical transport at the seaport was out of scope and considered as near boundary out of the mainland or moving sources.

For FD, the combination of Meso-Micro Scale of Meteorological Model (MM5-CALMET) was configured to simulate the wind field near ground. Firstly, a year 2010 simulation of MM5 will be initiated for the mother domain by using the optimal settings and parameters from the literature. CALMET, a preprocessor meteorology of CALPUFF, was then run for the child domain in order to relief a huge computational cost with the acceptable solution of the wind field parameter in a complex terrain. The output of friction velocity and roughness length played an important role on wind brow erosion over vegetation and soil. In addition, dust from paved and unpaved road over Thailand Road Network (TRN) can be further estimated.

In summary, we classified FES as the new emission sources consisting of VOCs and Dust. In year 2010, VOCs emissions from FES were approximately 1.3 x 106 Gg, comparable to those from natural sources. FVOCs will be in concern in the future and tend to rapidly increase based on the current situation of fuel and energy consumption of Thailand. A good practice to prevent FVOCs by storage and usage shall be dicussed.

For FD, understanding FD enlightened us to clearly illustrate size and its distribution of Particulate Matter (PM). It is clearly seen that not only urban but also rural areas can generate FD due to the unpaved road. In addition, other FD sources such as construction/demolition or agricultural operation should be included. At this end, FD can be useful for the further study of atmospheric aerosols in chemical transport model, which will be subjected to begin for the upcoming work.

P-5-019 Biogenic VOC emission modeling: Progress and Challenges

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Key words
biogenic VOC

The initial attempts to estimate emissions of biogenic volatile organic compounds (BVOCs) were made almost 50 years ago. These pioneering studies demonstrated the importance of BVOC and it is now widely recognized that these chemicals emitted from terrestrial ecosystems can influence atmospheric composition and quantitative estimates are needed for numerical assessments of past, present and future air quality and climate. The development and evaluation of the earliest BVOC emission models relied on branch enclosure measurements to identify and quantify the controlling processes and used inverse modeling of ambient concentrations to evaluate the emission estimates. Later biogenic VOC emission models have added complexity and are based on many more observations, including tower and airborne direct flux measurements, but are still associated with very high uncertainties. Model sensitivity studies demonstrate that these uncertainties limit our ability to characterize the earth system and develop air quality and climate management strategies. This presentation will provide an overview of the major advances, current status, and future needs for biogenic VOC emission modeling.

P-5-020 Enhanced profiling of tropospheric and stratospheric ozone, with near surface sensitivity, using simultaneously measured radiances from TES and OMI

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Key words

Atmospheric Ozone Satellite Remote Sensing Troposphere Planetary boundary layer thermal infrared ultraviolet-visible

Ozone, a radiativelly and chemically important trace gas, plays various roles in different altitude range in the atmosphere. In the stratosphere, it absorbs the solar UV radiations from the Sun and protects us from the sunburns and skin cancers. In the upper troposphere, ozone acts as greenhouse gases and relates to the global warming. In the middle troposphere, ozone reacts with many anthropogenic pollutants and cleans up the atmosphere. Near surface, ozone is being harmful to human health and plant life. Accurate monitoring of ozone vertical distributions is crucial for a better understanding of air quality and climate change.

The Tropospheric Emission Spectrometer (TES) and the Ozone Monitoring Instrument (OMI) are both onboard the Earth Observing System Aura satellite in orbit and have been providing ozone concentration profile measurements separately. TES is a Fourier transform spectrometer covering the 650 – 3050 cm^{-1} spectral range and measuring the thermal infrared (TIR) light radiances emitted by Earth's surface and by gases and particles. OMI is a nadir-viewing pushbroom ultraviolet-visible (UV-VIS) imaging spectrograph covering the 270–500 nm wavelength range and measuring backscattered radiances. We present an approach to combine simultaneously measured TES TIR and OMI UV-VIS radiances to improve the tropospheric ozone sounding. The tropospheric ozone profiles from the joint retrievals are characterized using the ozonesonde measurements and are consistent with co-located sonde measurements (differences are generally within 10%). The joint retrievals of TES and OMI measurements show the capability of capturing the variations of ozone volume mixing from surface to 700 hPa, while separate retrievals of TES measurements and OMI measurements are not sensitive in this altitude range. This work demonstrates that ozone profiles can be estimated from combined TES and OMI radiances and enhanced profiling of tropospheric and stratospheric ozone can be obtained with near surface sensitivity. To the best of our knowledge, our work is the first one that combines simultaneously measured radiances from two different instruments to jointly retrieve ozone profiles.

P-5-021 New constraints on North American acetone sources from tall tower measurements and inverse modelling

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Key words
acetone PAN source and emission

Acetone ($(CH_3)_2CO$) plays an important role in the atmosphere as a source of peroxyacetylnitrate (PAN) and hydrogen oxide radicals (HO_x). Here we apply a full year of continuous atmospheric acetone measurements from the University of Minnesota tall tower Trace Gas Observatory (KCMP tall tower; 244m a.g.l.), with a 1/2° x 2/3° GEOS-Chem nested grid simulation to develop quantitative constraints on seasonal acetone sources over North America, and assess the corresponding impacts on atmospheric chemistry. An inverse model analysis of the tall tower observations implies a 30% model underestimate of acetone emissions from broadleaf trees, shrubs, and herbaceous plants, and an offsetting 30% overestimate of emissions from needleleaf trees and secondary production from biogenic precursors. Our analysis shows that primary + secondary anthropogenic acetone sources in the model (based on EPA's NEI 2005 inventory) are accurate to within the constraints provided by the KCMP tall tower observations. The resulting optimized simulation captures 65% of the variance ($R = 0.8$) in the hourly measurements at KCMP tall tower, with minimal bias. We find during winter that acetone in the US Upper Midwest arises mainly from sources outside North America (50%), with primary (15%) and secondary (29%) anthropogenic sources within North America also important. During summer, North American biogenic sources predominate (47% primary; 14% secondary), with anthropogenic sources (20%) and long-range transport (19%) playing more modest roles. Initial results suggest that acetone can contribute up to 25 pptv (~10%) of PAN during summer over the US Midwest, but has only a minor impact during winter.

P-5-022 Diurnal variations of organic molecular tracers and stable carbon isotopic composition in atmospheric aerosols over Mt. Tai in North China Plain: an influence of biomass burning

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Key words

organic aerosol Mt. Tai Stable carbon isotope ratios

Organic tracer compounds, as well as organic carbon (OC), elemental carbon (EC), water-soluble organic carbon (WSOC), and stable carbon isotope ratios (δ13C) of total carbon (TC), have been investigated in aerosol samples collected during early and late periods of Mount Tai eXperiment 2006 (MTX2006) field campaign. Total solvent extracts were investigated by gas chromatography/mass spectrometry. More than 130 organic compounds were detected in the samples. They were grouped into twelve organic compound classes. In early June when the field burning activities of wheat straws were very active in North China Plain, the total identified organics (2090 ± 1170 ng m^{-3}) doubled those in late June (926 ± 574 ng m^{-3}). All the compound classes were more abundant in early June than in late June, except for phthalate esters, which were higher in late June. Levoglucosan (88-1210 ng m^{-3}, 403 ng m^{-3}) was found as the most abundant single compound in early June, while diisobutyl phthalate was predominant in late June. During the biomass-burning period in early June, we found a diurnal trend for most primary and secondary organic aerosol tracers, whose concentrations peaked in mid-night or early morning. On the other hand, in late June most organic species peaked in the late afternoon. This diurnal pattern suggests that smoke plumes derived from biomass burning can uplift the aerosol particles to a certain levels of altitude, which are then transported to and encountered the summit of Mt. Tai during nighttime. Based on the tracer-based method that can evaluate biomass-burning OC, fungal-spore OC and biogenic secondary organic carbon (SOC), we estimated that on average 24% of the OC was due to biomass burning in early June, followed by the contribution of isoprene SOC (4.3%). In contrast, isoprene SOC was the main contributor (6.6%) to OC, and only 3.0% of the OC was due to biomass burning in late June. In early June, δ13C of TC (−26.6‰ to −23.2‰, mean −25.0‰) were lower than those (−23.9‰ to −21.9‰, −22.9‰) in late June. In addition, a strong anti-correlation was found between levoglucosan and δ13C values. This study demonstrates that burning activities of crop-residues can significantly enhance the organic aerosol loading, alter the organic molecular compositions and control the stable carbon isotopic compositions of aerosol particles in the troposphere over North China Plain.

P-5-023 Atmospheric deposition of phosphorous over the east mediterranean

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Key words
Atmospheric deposition P speciation Eastern Mediterranean

With humans having an increasing impact on Earth, the perturbation of biogeochemical cycles of nutrients (phosphorus, nitrogen and carbon) was inevitable. The interactions between phosphorus-carbon cycles and climate are expected to become an increasingly important determinant of the Earth biogeochemical cycles. Since P is the limiting factor for many oligotrophic marine environments, such as the East Mediterranean Sea (EMS), valuable information could be arisen by defining the role of organic and inorganic forms of P. Moreover, since the oceans act as an important sink of atmospheric CO_2, P limitation of marine primary productivity could play a key-role to this natural process, affecting indirectly the global warming.

This study aims to investigate the sources, the forms and the biogeochemical significance of soluble atmospheric P over the EMS. Wet (n=28) and bulk deposition samples (n=28) have been collected during two year period (2008-2009) and analyzed for P speciation. Following the analytical protocol referred in Standard Methods for the Examination of Water and Wastewater (20th Edition), Dissolved Reactive Phosphorous (DRP) was determined colorimetrically as PO_4^{3-} with stannous chloride method, while Total Dissolved acid hydrolized Inorganic Phosphorous (TDIP) was determined after mild oxidation of sample. Total Dissolved Phosphorus (TDP) was measured after the acid digestion of samples according to Persulfate Digestion Method. Dissolved organically bound phosphates (DOP) was determined by subtracting TDIP from TDP, while pyro-, meta- and other polyphosphates (condensed phosphates, CP) was determined by subtracting SRP from TDIP. In selected samples P speciation was also conducted using 31P-NMR spectroscopy.

Total of 28 rain events were collected over two years (2008-2009) at marine background area of the EMS. Volume weighted mean (VWM) concentration of TDP was estimated at 0.8 μM, while the percentage contribution of DRP, DOP and CP to TDP was estimated 22%, 58% and 20%, respectively. Investigating the role of air mass origin on the P speciation in rainwater, rain samples have been classified in two classes (N/NW and S/SW) corresponding to the main wind sectors influencing the area. Maximum VWM of DOP (0.7 μM), is associated with S/SW winds, which enrich the atmosphere over the EMS with African dust, while N/NW winds, which transport the anthropogenic pollution from N/NW Europe, is associated mainly with the TDIP (0.5 μM).

Significant is the dry deposition of phosphorus since the average percentage contribution of dry deposited TDP during both years was estimated 72% of total deposition. Dry deposition has been calculated as the difference between bulk and wet deposition. Furthermore the average percentage dry deposition of DOP, CP and DRP to total deposition was estimated at 57%, 65% and 86%, respectively. Since DRP is considered as the most bioavailable form of phosphorus, important role plays the dry deposition of P in oligotrophic marine environment.

To enable source apportionment analysis, chemical speciation for the major anions, cations and trace metals has been performed. The results are going to be presented and the factors controlling the P concentration levels in deposition samples over the Eastern Mediterranean are going to be thoroughly discussed.

P-5-024 Measurements of biogenic and anthropogenic volatile organic compounds by PTR-ToF-MS in a semi-rural area in Germany

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Key words
volatile organic compounds (VOC) PTR-ToF-MS

Volatile organic compounds (VOC) play an important role in the atmosphere since they are the “fuel” that keeps atmospheric chemistry running. Despite their low concentrations they have great effects on atmospheric chemistry like the formation of ozone and secondary organic aerosol. Due to various measures aiming at a reduction of anthropogenic emissions the pattern of VOC in the atmosphere has significantly changed in the last decade. Especially in rural areas, the amount of biogenic emissions is exceeding the anthropogenic emissions. The emission rates of biogenic VOC are expected to depend on parameters like air temperature, humidity and photosynthetic active radiation and might show diurnal cycles and seasonal variations.

In order to examine the effects of both biogenic and anthropogenic emissions on radical chemistry, in August and September 2011 the field campaign PARADE (PArticles and Radicals: Diel observations of the impact of urban and biogenic Emissions) was conducted at the Taunus observatory on the summit of Kleiner Feldberg, Germany, about 10 km north of Frankfurt. The measurement site is surrounded by mainly coniferous forest, but also heavily impacted by anthropogenic emissions from the large urban agglomerations of the Rhein-Main area with large cities, a dense motorway system, and Germany’s largest airport.

We applied a PTR-ToF-MS (Proton Transfer Reaction – Time of Flight – Mass Spectrometer) for online detection of VOC in the atmosphere. The system is able to measure organic compounds with proton affinities higher than that of water with comparably high time resolution. The measurements were carried out continuously, day and night, with only a few data gaps. Full mass spectra were recorded up to 450 Th with a time resolution of one minute covering a measurement period of four weeks. Regular blank measurements were performed during the campaign. Calibration measurements were performed before and after the campaign with a multi component gas standard.

The results show that methanol seems to be the most abundant biogenic compound with volume mixing ratios ranging from 1 to 10 ppb depending on the environmental conditions. Toluene as a tracer of anthropogenic emissions showed relatively low mixing ratios between 50 and 150 ppt with pronounced spikes up to 600 ppt indicating epsiodes of air masses advected from the Rhein-Main area.

The mixing ratios of selected biogenic and anthropogenic VOC are used to study the impact of these emissions on atmospheric chemistry at different meteorological conditions.

P-5-025 15N tracer study of N_2O emissions from soil using FTIR spectroscopy

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Key words
Nitrous oxide isotopic tracers 15N FTIR spectroscopy soil fluxes

Estimates of greenhouse gas emissions can be improved by understanding how nitrogen transformations in the soil affect fluxes of nitrous oxide (N_2O). Soil nitrogen transformations and their impact on N_2O emission are frequently studied using 15N labelling techniques, but continuous measurements of the total amount of added 15N transformed to N_2O are rarely achieved in the field. Here we study the short-term (2-4 week) fate of applied nitrogen using a mobile Fourier Transform Infrared (FTIR) spectrometer to measure 15N isotopic signatures of N_2O emitted from an agricultural soil after 15N addition as nitrate or urea. The experiment site was on a coastal floodplain site managed for silage production near Nowra, New South Wales, in SE Australia. We deployed five 0.25 m^2 automated chambers connected to a multi-pass cell and low resolution FTIR spectrometer to measure mole fractions and fluxes of all N_2O isotopologues every three hours. Root mean square uncertainties for all isotopologue mole fractions were less than 0.3 nmol mol^{-1} for 1 minute average measurements, and minimum detectable fluxes for each isotopologue were <0.1 ng N m^{-2} s^{-1}. We applied a light dose of 15N-labeled substrate (0.4-0.8 g 15N m^{-2}) as potassium nitrate (KNO_3) or urea [$CO(NH_2)_2$]. Flux measurements were made for 1-2 weeks before and after application of the labelled nitrogen.

Emissions of all isotopologues were evident within three hours following 15N addition. 15N15NO emission subsided in less than 7 days, and emissions of

14N15NO and 15N14NO isotopomers after 10 days. The chamber soils were destructively sampled after 2 weeks, so that 15N could be analysed in soils and plant material using isotope ratio mass spectrometry. Emissions of 14N14NO were evident throughout the entire experiment. Cumulative 15N-N_2O fluxes (sum of the three isotopologues) per chamber for the 14 days following 15N addition ranged from 8 to 20 mg 15N-N_2O m^{-2}, representing 1 to 3% of the applied N. The results show that FTIR spectroscopy provides a powerful isotopic labelling technique for study of N_2O emissions under field conditions.

P-5-026 Formation of secondary organic aerosol from biogenic emissions

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Key words
biogenic VOCs secondary organic aerosol

Atmospheric oxidation of volatile organic compounds (VOCs) contributes to new particle formation and atmospheric organic aerosol mass. The formation of aerosols from biogenic VOC emissions constitutes a possible feedback element in biosphere-atmosphere-climate interactions due to the overall cooling effect of aerosols. This assumes increasing VOC emission strengths with increasing temperature and emission patterns being invariant to temperature changes. Recently evidence emerges that temperature induced changes in VOC emission patterns may alter the picture. It has been shown that increased isoprene emissions may suppress atmospheric new particle formation thus dampening the cooling effect of aerosols formed from biogenic VOCs. In addition the use of direct emissions of VOCs from plants in experiments studying secondary organic aerosol (SOA) formation shows that, beyond the so far considered main compound classes isoprene and its derivatives monoterpenes and sesquiterpenes, other VOC classes significantly impact SOA formation. Many of these VOCs are emitted under plant stress conditions. An overview of the state of the art knowledge of SOA formation from biogenic VOCs with respect to different VOC classes will be given.

P-5-027 Comparison of model-simulated atmospheric CO_2 with satellite retrievals over East Asia

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Key words
Carbon Dioxide East Asia Modeling Remote Sensing

Global atmospheric CO_2 distributions were simulated with a chemical transport model (GEOS-Chem) and compared with space-borne observations of CO_2 column density by GOSAT from April 2009 to January 2010. The GEOS-Chem model simulated 3-D global atmospheric CO_2 at 2°×2.5° horizontal resolution using global CO_2 surface sources/sinks as well as 3-D emissions from aviation and the atmospheric oxidation of other carbon species. The seasonal cycle and spatial distribution of GEOS-Chem CO_2 columns were generally comparable with GOSAT columns over each continent with a systematic positive bias of ~1.0%. Data from the World Data Center for Greenhouse Gases (WDCGG) from twelve ground stations spanning 90°S-82°N were also compared with the modeled data for the period of 2004-2009 inclusive. The ground-based data show high correlations with the GEOS-Chem simulation ($0.66 \leq R2 \leq 0.99$) but the model data have a negative bias of ~1.0%, which is primarily due to the model initial conditions. Together these two comparisons can be used to infer that GOSAT CO_2 retrievals underestimate CO_2 column concentration by ~2.0%, as demonstrated in recent validation work using other methods. We further estimated individual source/sink contributions to the global atmospheric CO_2 budget and trends through 7 tagged CO_2 tracers (fossil fuels, ocean exchanges, biomass burning, biofuel burning, net terrestrial exchange, shipping, aviation, and CO oxidation) over 2004-2009. We downscaled modeled CO_2 data with regional-scale model and investigated the seasonal changes of atmospheric CO_2 distribution over East Asia.

P-5-028 Investigating the effect of the seasonal variation of emissions from Arctic wetlands on the atmospheric methane budget using 4D-Var inverse modelling

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Key words
Methane Inverse Modelling 4D-Var Wetlands Arctic

Methane (CH_4) is a greenhouse gas which is emitted from a variety of anthropogenic and natural sources. CH_4 is the second most important greenhouse gas in terms of radiative forcing, behind only carbon dioxide (CO_2) in its influence on the climate, and the atmospheric concentration of CH_4 is currently growing at a unprecedented rate. Surface emissions of CH_4 originate from a range of contributing biogenic sources, including wetlands, rice agriculture and livestock, and non-biogenic sources such as the fossil fuel industry and biomass burning. It is important that the nature, location and magnitude of these emissions are fully understood in order for policy-makers to be able to reduce anthropogenic contribution to climate change, but also so that atmospheric models may use such

inventories as boundary conditions in order to accurately reproduce atmospheric concentrations of the species.

It has previously been suggested that natural variation in the rate of CH_4 emissions from wetland regions are responsible for the majority of the inter-annual variation of the atmospheric CH_4 budget. However, there are currently large uncertainties in our emission estimates, especially from natural processes such as biogenesis from wetlands or those with varying geographical distributions such as biomass burning. Two methods of reducing these uncertainties which have become prevalent in recent years are synthesis inversions and four-dimensional variational (4D-Var) inverse transport modelling, both of which are Bayesian data assimilation techniques which seek to constrain emission rates by minimising the difference between the observed atmospheric concentrations and the modelled estimate of an atmospheric species.

We have used the TOMCAT 3D Chemical Transport Model (CTM) to assess the ability of current emission inventories to capture the observed inter-annual variation of atmospheric CH_4. In order to contribute to the improvement of these inventories, we have also developed both a synthesis inversion system and an inverse 4D-Var version of the TOMCAT model and used these new systems to examine the relationship between the atmospheric concentration of CH_4 and its surface emissions. Particular emphasis has been placed upon investigating the magnitude and seasonality of wetland emissions in the high-latitude Northern Hemisphere, such as those from Scandinavia, Siberia and North America.

P-5-029 Regional Differences in the Organic Composition and FTIR Spectra of Marine Aerosol Particles

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Key words
Marine Aerosol Organic Composition

Marine aerosol particles play an important role in the earth’s radiative balance, yet the sources and composition of the organic fraction remain largely unconstrained. Recent measurements have been made in order to characterize the sources, composition, and concentration of particles in the marine boundary layer. The organic compositions of submicron particles derived from multiple seawater regions have been measured using Fourier Transform Infrared (FTIR) spectroscopy. Cluster analysis of FTIR organic spectra suggest different spectral signatures based on collection location, seawater composition, and ambient conditions. Measurements including aerosol composition from a high resolution time of flight aerosol mass spectrometer (HR-ToF-AMS), seawater composition,

and wind speed were used to interpret the cluster results, depending on the availability from each campaign. Recent measurements used in the comparison include ambient and generated marine aerosol particles measured on board the R/V Atlantis during CalNex in May and June 2010. The ambient clean marine particles had an average organic mass (OM) of 0.80 μg/m^3 and average chlorophyll and wind speed values of 6 m/s and 7 μg/L, respectively. Additional remote ambient marine aerosol particles were collected 100 miles off the coast of Monterey in the eastern Pacific on board the R/V Point Sur during the EPEACE experiment in July 2011 and had an average OM of 0.64 μg/m^3 and an average wind speeds of 7 m/s. These particles are compared with those measured in the southeastern Pacific during VOCALS and the north Atlantic during ICEALOT. One spectral cluster from the eastern Pacific showed the highest fraction of hydroxyl functional groups (77%), which corresponds to periods of high relative chlorophyll concentrations and high wind speeds (>10 m/s).

P-5-030 Evaluation and Improvements of Two Community Models in Simulating Dry Deposition Velocities for Reactive Nitrogen Oxides over Forests

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Key words

dry deposition velocity reactive nitrogen oxides WRF-Chem dry deposition module Noah-GEM 1-D model

Dry deposition velocities (Vd) for peroxyacetyl nitrate (PAN) and total reactive nitrogen oxides (NOy) calculated using two community dry deposition models with different treatments of both stomatal and nonstomatal uptakes were evaluated using measurements of PAN eddy covariance fluxes over a Loblolly pine forest in July 2003 and measurements of NOy eddy covariance fluxes and NO, NO_2, PAN, HNO_3 and NOy concentrations over a temperate deciduous forest in October 2000. The observed daytime maximum of Vd(PAN) was about 1.0 cm s-1 on average, while the estimates by the WRF-Chem dry deposition module (WDDM) and the Noah land surface model coupled with a photosynthesis-based Gas Exchange Model (Noah-GEM) were only 0.2 cm s^{-1} and 0.6 cm s^{-1}, respectively. The observations also showed considerable PAN deposition at night with typical Vd values of 0.2-0.6 cm s^{-1}, while the estimated values from both models were less

than 0.1 cm s^{-1}. Noah-GEM modeled more realistic stomatal resistance (Rs) than WDDM, as compared with observations of water vapor exchange fluxes. The poor performance of WDDM for stomatal uptake is mainly due to its lack of dependence on leaf area index. Thermal decomposition was found to be relatively unimportant for measured PAN fluxes as shown by the lack of a relationship between measured total surface conductance and temperature. Thus, a large part of the underprediction in Vd from both models should be caused by the underestimation of nonstomatal uptake, in particular, the cuticle uptake. The NO_y species have various physical and chemical natures, leading to very different behaviors in the deposition process. The Vd(HNO_3) estimated by Noah-GEM is one order of magnitude larger than Vd(NO_2) and Vd(PAN), whereas Vd(NO) is close to zero. HNO_3 was found to dominate the NOy flux during the measurement period at the temperate deciduous forest site; thus the modeled Vd(NO_y) was mainly controlled by the aerodynamic (Ra) and quasi-laminar sublayer resistances (Rb), both being sensitive to the surface roughness length (z0). Using an appropriate value for z0 (10% of canopy height), WDDM and Noah-GEM agreed well with the observed daytime Vd(NO_y). The differences in Vd(HNO_3) between WDDM and Noah-GEM were small due to the small differences in the calculated Ra and Rb between the two models which both parameterizes the surface exchange coefficient based on the Monin-Obukhov Similarity Theory (MOST); however, the differences in Rc of NO_2 and PAN between the two models reached a factor of 1.1-1.5, which in turn caused a factor of 1.1-1.3 differences for Vd. Combining the measured concentrations and modeled Vd, NOx, PAN and HNO_3 accounted for 19%, 4%, and 70% of the measured NOy fluxes, respectively. Our future efforts will involve 1) investigating the impact of different Vd(PAN) on atmospheric chemistry and nitrogen deposition budget by using a 3D atmospheric chemical models (e.g., WRF-Chem); and 2) utilizing vertically-varying profiles of mean scalar concentration or a multi-layer canopy model that explicitly resolves the radiative, dynamical, and thermal transfer within vegetation canopies in order to improve the parameterization of the surface exchange coefficient within the models.

P-5-031 Nitric oxide over Milan oasis (Xinjiang, PR China) due to biogenic emissions from soil – Results of soil sample laboratory incubations vs. 3D dispersion modeling vs. ground and satellite based MAXDOAS measurements

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Today's knowledge of soil biogenic NO emission rates from arid and hyper-arid land is based on a total of about 20 experimental studies. Nevertheless, biogenic NO emissions even from non-managed arid and hyper-arid soils are significant and may range between 1 10 ng m^{-2} s^{-1} (in terms of nitrogen, if conditions for soil NO production are favourable (optimum soil moisture, high soil temperatures). Irrigated and fertilized oases, ranging about 3000 km long around the great Central Asian Taklimakan desert form the backbone of the agricultural output (80% of the Chinese cotton production) of the Xinjiang Uygur Auto¬no¬mous Region (NW-China). Recent and future development of farmland and intensification of agriculture will definitely impact the regional soil NO emission and consequently the budget of nitrogen oxides and ozone. Up to today, only a few studies have preliminarily addressed soil biogenic NO emissions from the Taklimakan desert.

In our contribution, we will focus on the quantification of the area integrated NO emission from the Milan oasis located on the most southern fringe of the Takalimkan desert (39.26°N, 88.91°E). At a first step, soil samples have been taken from individual fields, representative for the land use classes of Milan oasis (cotton fields, Jujube orchards, natural oasis forest, desert). Next, field specific potential net NO emission has been parameterized in terms of soil temperature, soil water content, and soil nutrient content by laboratory incubation. Actual dis¬tri¬bution of Milan fields has been determined by advanced GIS techniques. Consequently, the strength of biogenic NO emissions from Milan fields could be calculated, since their geographical location as well as their areal extent, their stage of vegetation growth as well as irrigation and fertilization events and amounts, soil temperatures and soil water contents are known. Next, the 3D distribution of ambient NO concentration is calculated using a state-of-the-art commercially avai¬l¬able dispersion model (LASAT 3.2, Lagrange Simulation of Aerosol-Transport). Meteorological input for LASAT (wind speed and wind direction, atmospheric stability, roughness length, radiation intensity) is provided by the results of an automatic weather station network, consisting of six individual stations which have been distributed over the entire oasis.

Since ambient ozone concentrations and NO_2 photolysis frequencies have been measured in-situ, 3D distribution of ambient NO_2 concentrations can also be determined. This is necessary for comparison of the laboratory incubation/3D-LASAT simulation data with height-integrated NO_2 concentrations provided by ground and satellite based multi-axis differential optical spectroscopy (MAXDOAS) measurements which have been performed at and over Milan oasis. Vertical cross-sections of NO_2 concentrations along the “beam” of ground-based MAXDOAS measurements have been constructed, vertical integration and horizontal averaging of corresponding NO_2 concentrations simulated the results of MAXDOAS measurements (in terms of vertical NO_2 column densities, VCDs).

Results of a three week period (June 2011) are presented. Comparison of VCDs derived from (a) laboratory incubation-3D LASAT simulation, (b) ground-based, and (c) satellite-based MAXDOAS agreed astonishingly well. Consequences of the relationship between NO_2-VCDs and NO biogenic emissions from (arid) soils are discussed as well as the impact on up-scaling on oasis-, regional- and province wide scales.

P-5-032 Biogenic volatile organic compound emission from a temperate forest in Changbai Moutain

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Key words

Biogenic volatile organic compound monoterpene relaxed eddy accumulation (REA) Changbai Mountain

Biogenic volatile organic compound (BVOC) play important roles in atmospheric chemistry and photochemistry, BVOCs’ oxidation influences OH radical and O_3 concentrations, regulate the earth’s climate directly and indirectly. BVOC emissions were measured in a temperate forest, Changbai Moutain, China during the growing season in 2010 along with meteorological parameters including global radiation and PAR. BVOC flux measurements were made by relaxed eddy accumulation (REA) on an above canopy flux tower. The monoterpene emission at this site was dominated by α-pinene, terpinolene, o+p-cymene, d-limonen, camphene, myrcene with averaged emission fluxes of 0.068, 0.053, 0.045, 0.037, 0.025, 0.025 mg m^{-2} h^{-1}, respectively during the growing season. The highest emission fluxes of the above monoterpenes were 0.21, 0.32, 0.28, 0.20, 0.36, 0.43

mg m^{-2} h^{-1}, respectively. For the identified 12 monoterpene species, the mean total flux was 0.22 mg m^{-2} h^{-1} and ranged from 0.01 to 1.67 mg m^{-2} h^{-1}.

Measurements were conducted from 6:30 to 18:30 with 3-h interval to investigate the diurnal variations of BVOC. The highest monoterpene emissions occurred in the afternoon with lower emissions in the morning and late evening. In addition, the highest monoterpene emission appeared 3 hours later than that of PAR on most days and appeared simultaneously with the peak of air temperature on some days. An empirical model was developed for calculating monoterpene emission flux based on PAR energy balance, the calculated flux was in agreement with the observed.

P-5-033 On the history, background and recent findings of biogenic emission of nitric oxide (NO) from soils as a strong chemical sink for near-surface ozone

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Key words

surface-atmosphere exchange of ozone ozone, nitric oxide dry deposition biogenic soil emission

In the dry season of 1992, measurements in the Kalahari region (southern Africa, SAFARI'92), occasionally revealed extremely low surface concentrations of ozone (O_3) in a fully developed, very strong and shallow nocturnal boundary layer. That and other observations have raised reasonable suspicion that extremely low O_3 concentrations are due to more than dry deposition of ozone to vegetation and/or bare soil surfaces. Quite similar results from a long series of similar field measurements in Africa, Brazil, Germany, and China followed (1993-2011), which led to the hypothesis, that the necessary sink of O_3 is just the near-surface titration of ozone by nitric oxide (NO), which is biogenic emitted from microbes in the top soil layer.

Meanwhile, there is much more knowledge about the nature and amount of biogenic NO soil emissions − from mechanistic, laboratory, and field experiments, as well as from mechanistic, local, regional and global modelling. Furthermore, about a decade, concurrent measurements of NO-NO_2-O_3 concentrations and fluxes in the field (Africa (savannah), Brazil (primary rainforest), Germany (agricultural plots, forests), and China (oasis agriculture)) are available to study the strong interaction between turbulent transport, biological uptake, and biogenic soil emissions within the surface–atmosphere exchange of the NO-NO_2-O_3 triad.

We will report on experimental findings of field measurements with respect to the suspected near-surface chemical O_3 sink, as well as on the mechanistic background of the ubiquitous NO emission from soils. Attempts to model the turbulence-chemistry driven surface exchange of the NO-NO_2-O_3 triad will also be discussed.

P-5-034 Emissions of Air Pollutants and Greenhouse Gases from Xi'an, China: Measurements by Eddy Covariance

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Key words

Eddy Covariance Xi'an China Green House Gases

Xi'an, the capital of Shaanxi Provence, is one of the fastest growing cities in inland China in terms of population and economic output. Coupled to this growth is an increasing demand for energy and transportation, leading to rises in emissions of both greenhouse gases (GHGs) and air pollutants. The increased emissions of GHGs from Xi'an and similar rapidly developing urban areas are expected to have significant negative impacts on the regional and global climate. In addition, the increased emissions of air pollutants will have negative impacts on the health of the human population and on the surrounding ecosystems. For these impacts to be mitigated, we first need to understand how these rapidly growing cities contribute as both a source and a sink of air pollutants and GHG emissions. In August 2011, we conducted an intensive field campaign in the city of Xi'an to begin to investigate these questions.

During the Xi'an campaign, we monitored the major boundary-layer meteorological variables as well as the high frequency fluctuations of carbon dioxide (CO_2), nitrous oxide (N_2O), methane (CH_4), oxides of nitrogen (NOx), carbon monoxide (CO), selected volatile organic compounds (VOCs), and particle number concentration. These data was analyzed using the eddy covariance method, a statistical approach that directly measures the net ecosystem exchange of a scalar quantity of interest by its concentration within turbulent eddies as they pass through a point in space. Our analysis indicates that on average the city of Xi'an acted as a source of GHGs as well as the other air pollutants monitored during the study. However, the GHG fluxes observed were significantly lower than those observed in other cities around the world. This was attributed to the presence of a highly vegetated recreational park in the prevailing wind direction, which acted as a sink for CO_2 and N_2O during daytime hours. For CO_2 this uptake was attributed to increase photosynthesis and for N_2O it was attributed to soil uptake. In addition, ambient concentration of the GHG monitored exhibited a diurnal trend that deviated from those observed in other major cities around the world, where correlations with vehicular activity are predominant. Instead, in Xi'an, the diurnal trend for GHG and especially CO_2 emissions exhibited a pattern that is usually associated mixing layer dynamics. Where concentration increases during nighttime hours when the mixing layer is shallow, followed by a gradual dilution of the concentration as the mixing layer height starts to increase due to surface heating at sunrise. This suggested that ambient concentrations of the GHG were mostly driven by emission further from our sampling footprint.

P-5-035 Correlations between Ozone and Its Precursors in Peninsular Malaysia

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Key words
Ozone Precursors Correlation Exceedance Peninsular Malaysia

Ozone (O_3) is one of the most influencing factors affecting human health, animals and vegetation ecosystems. That is why, nowadays, ozone is a widespread photochemical oxidant phenomenon in many monitoring stations in the world. This study aims to identify and describe the correlations between ozone and its precursor emissions, the number of total hours, days and the maximum concentration of ozone at three selected monitoring stations in the urban, sub-urban and rural areas of the Malaysian peninsular over a five-year period (2005-2009). Analysis of ozone was done using the SPSS (Statistical Package for the Social Science) to obtain the correlation of ozone and also to clarify the relationship between stations. The comparison of the results showed that among the three monitoring stations, semi-urban area (Putrajaya) had the highest concentration of O_3 with 148 ppbv. Besides, the highest exceedance during the period 2006 in this area, and the lowest which considered between early 2008 to 2009 annual exceedance was also investigated. The data analysis also revealed that the correlation between O_3 and NO was high at urban area (Petaling Jaya) with significant value (ρ = -0.678), whilst the relationship between O_3 and NO_2 was high (ρ = -0.499) at Putrajaya (S1). It was concluded that in urban and rural areas of Malaysian peninsular, the concentration of ozone depends on the concentration of NOx and seasonal meteorological factors.

P-5-036 Degradation of Nitrogen Oxides on Photocatalytic TiO_2 Cement: Kinetic and Product Study

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Key words
Nitrogen oxides Photocatalytic degradation

Photocatalytic degradation of pollutants is known since 1921 (Renz) and was later established as the “Honda-Fujishima-Effekt” (1967). Photocatalytic activated TiO_2-containing materials are used to decrease the concentration of air pollutions since more than 20 years and have applications in different materials, like paints, concrete, self cleaning window glass, etc.. Especially, the degradation of nitrogen oxides (NO_x= NO+NO_2) by photocatalytic materials is of high interest, to meet the EU threshold limit values introduced in 2010, which are typically exceeded under urban conditions. First pilot studies (e.g. PICADA) have shown high reduction of NO_x, however, extrapolation to atmospheric relevant conditions and identification of potentially harmful products is still open issue.

In the present study, TiO_2 containing photocatalytic cement, which was recently used in the Brussels tunnel study of the PhotoPaq project (Photocatalytic remediation Process on the air Quality), was investigated in the laboratory. Both, the kinetics and the products were studied for the photocatalytic reactions of nitric oxide (NO), nitrogen dioxide (NO_2) and nitrous acid (HONO) on the cement surfaces in a photo-flow reactor (ISO). A reaction mechanism and the possible atmospheric implication will be discussed.

P-5-037 Fluxes of Nitrous Acid (HONO) above an Agricultural Field Side near Paris

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Key words
Nitrous acid Flux measurement

HONO is an important precursor of the OH radical, the detergent of the atmosphere. Field measurements show high diurnal HONO mixing ratios that cannot be explained by chemical models with known gas phase chemistry. Therefore, daytime sources of HONO are still under discussion. During the last decade many experimental investigation were performed to study heterogeneous production of HONO like the photo enhanced reduction of NO_2 on humic acids or photolysis of HNO_3 on surfaces. Recently, nitrite produced by bacteria, present in soil, was discussed as a source of HONO as well. In addition gas phase sources like the photolysis of nitrophenols, or the reaction of excited NO_2 are discussed. Gradient measurements show high mixing ratios of HONO even above the boundary layer. However, beside intensive investigations on the sources of HONO, it is still an open question whether heterogeneous or gas phase sources are more important in the atmosphere. Flux measurements could represent a method to find the origin of missing sources of HONO.

Until now instruments are not sensitive and fast enough to do Eddy correlation measurements for HONO. Alternatively, HONO fluxes are estimated by the Aerodynamic Gradient (AGM), or Relaxed Eddy Accumulation (REA) methods.

Here we present HONO fluxes estimated by AGM and the LOPAP technique (Long Path Absorption Photometer) above an agricultural field in Grignon, Paris (48°51'N, 1°58'E). Fluxes during different seasons and different types of vegetations including bare soil will be presented and compared with different parameters like chemically corrected NO_2 fluxes.

P-5-038 Modeling Monoterpene Emissions from Boreal Pine Forest in Southern Finland

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Key words
Monoterpene SOSA Chemotype

As one of the important biogenic volatile organic compounds (BVOCs) groups, monoterpene has been drawing more and more scientific attention in atmospheric research because of their chemical reactions to produce and destroy tropospheric ozone, their effects on aerosol growth and formation and their potential influence on global warming. Regional measurements and estimates are urgently needed to study carbon budgets and global climate. However, since various factors such as genetic variation, temperature and light, water availability, seasonal changes, and

environmental stresses have complicated impacts on monoterpene emissions, comprehensive inventories are not so often reliably defined. To further track monoterpene concentrations and their chemical transformations, the model SOSA (model to Simulate the concentrations of Organic vapours and Sulphuric Acid) is applied to investigate Scots pine (Pinus sylvestris) tree emissions in a boreal coniferous forest at SMEAR II (Station for Measuring forest Ecosystem-Atmosphere Relations) in Hyytiälä, Finland. SOSA combines meteorological transport, emissions and chemistry. To test the reliability of SOSA, simulation outputs are compared with measurement data collected from on-line chambers analysed by Proton-transfer-reaction mass spectrometry (PTR-MS) and Gas chromatography-mass spectrometry (GC-MS) analyzer. Results indicate that modeling and observations of monoterpene concentrations and fluxes reasonably agreed both day and night. However, the correlation coefficients still reveal some additional parameters like environmental stress, plant development and leaf maturation, nutrient and injury status also influencing monoterpene emissions, but were not taken into account in the model. The dominant monoterpenes species emitted from these coniferous trees are alpha–pinene and delta3–carene with the proportion of 48% and 23% respectively. Monoterpene emissions did continue with low rates during the night. Diurnal and seasonal variations are demonstrated both quantity and quality. Summer is the most active season and emission rate increases to the peak around 3pm during daytime. According to vertical profile, monoterpene concentrations are highest at lower high level during night and well mixed during day. All the results are generally supported by measurement. Another significant phenomenon for monoterpene emitters is the discrepancy between branch scale emissions and above-canopy concentrations. SOSA is also used to estimate the contribution of different monoterpene distributions to the total OH reactivity. Results show that by selecting alpha–pinene and sabinene as the chemotype-species, the OH reactivity differs by a factor of almost 3. In order to reduce uncertainty in measuring and modeling, a more detailed chemotypic characterization blends needs to be constructed.

P-5-039 The impact of deep convection on the contribution of very-short-lived bromocarbons to stratospheric bromine

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Key words
bromocarbons convection

Recent observations and modeling studies suggest that oceanic emission of very short-lived substances (VSLS) contribute 3-8 pptv to reactive bromine in the stratosphere in addition to long-lived halons and methyl bromide. Inorganic bromine produced from VSLS (BryVSLS), when lofted via tropical deep convection, can catalytically destroy ozone in the stratosphere. VSLS contribution may vary significantly in a warmer climate as convection is likely to increase with warmer sea surface temperatures. Our recent model analysis using a 50-year simulation from 1960-2010 with the GEOS Chemistry Climate Model (GEOS CCM) with coupled bromoform (CHBr3) and dibromomethane (CH2Br2) chemistry suggestion the inclusion of CHBr3 and CH2Br2 adds ~7.5 pptv to inorganic bromine in the stratosphere. Due to the substantial differences exist between different convective schemes and their implementation in GCMs, significant uncertainty can be introduced into simulated BryVSLS. We conduct two sensitivities runs with maximum and minimum convective strength, respectively, to quantity the uncertainty in the troposphere to stratosphere transport (TST) of VSLS source gases (SG) and product gases (PG) due to variations in deep convection. We will also examine how PG and SG TST vary between El Nino and La Nina years. Through the above, we aim to improve our understanding of how convective transport and the associated scavenging affects TST of VSLS and how this may change in a future climate.

P-5-040 Eddy covariance flux measurement for biogenic volatile organic compounds using proton transfer reaction – time of flight – mass spectrometry (PTR-TOF-MS) over an orange orchard in the Central Valley of California

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Key words
PTR-TOF-MS Eddy Covariance BVOCs Flux

During summer 2010, a proton transfer reaction – time of flight – mass spectrometer

(PTR-TOF-MS) and a standard proton transfer reaction mass spectrometer (PTR-MS) were deployed simultaneously for one month in an orange orchard in the Central Valley of California to collect continuous data suitable for eddy covariance (EC) flux calculations. The high time resolution (5 Hz) and high mass resolution (up to 5000 m/Δm) data from the PTR-TOF-MS provided the basis for calculating the concentration and flux for the full range of biogenic volatile organic compounds (BVOCs). Throughout the campaign 664 mass peaks were detected in mass to charge ratios between 0 and 1278. We proved 493 ion species had fluxes above signal to noise of 3, and 82 masses identified as specific molecular formulae made significant contributions to BVOC fluxes at this site. The EC flux data from PTR-TOF-MS measurement were validated through spectral analysis (i.e. cospectrum, normalized co-spectrum, and ogive). Based on inter-comparison of the two PTR instruments, no significant instrumental biases were found in either mixing ratios or fluxes, and the data showed agreement within 5% on average for methanol and acetone. For commonly measured BVOCs, the EC fluxes from PTR-TOF-MS were in agreement with the qualitatively inferred flux directions

from vertical gradient measurements by PTR-MS. The PTR-TOF-MS measured total mean net flux of ~397 µg C m^{-2} h^{-1} (or ~3.38 nmol m^{-2} s^{-1} in mole unit). The dominant BVOC emissions from this site were terpenes (~26%, ~102 µg C m^{-2} h^{-1}) and methanol (~18 %, ~72 µg C m^{-2} h^{-1}), followed by acetone (~9.2 %, 36.5 µg C m^{-2} h^{-1}), acetic acid (~9.0%, 35.7 µg C m^{-2} h^{-1}), and acetaldehyde (~2.9 %, 11.5 µg C m^{-2} h^{-1}). Low level of emissions/deposition (less than 1% of the total) was observed for 72 distinct masses. We classified these 72 masses by the number of oxygen in the molecule. The pure hydrocarbon emission was largest, followed by one-, two-, and three- oxygen containing compounds. However, the total flux of oxygen containing compounds (55%) was greater than the flux of pure hydrocarbons (45%) which probably results from both significant photooxidation within the canopy and direct emissions of oxygenated species. Oxygen containing species (67%) dominated the deposition at this site which suggests that photooxidation leads to significant dry deposition of volatile organic carbon that may have originated as either biogenic or anthropogenic emissions. Total net BVOC fluxes measured by PTR-TOF-MS were at least twice larger than the total flux of the species measured by PTR-MS. Overall, our results show that eddy covariance flux measurements using PTR-TOF-MS is a powerful new tool for characterizing the biosphere- atmosphere exchange for a large range of BVOCs and their oxidation products, and that a very broad array of VOCs are significant in determining the total biosphere- atmosphere flux.

P-5-041 Measurements of carbon monoxide and its isotopes covering the past two millennia from Antarctica

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P-5-042 Stable isotopic compositions of atmospheric CO at high northern latitude during 2004-2009: observations and model simulations

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Key words:
carbon monoxide isotopic composition high northern latitude MOZART-4

Carbon monoxide (CO) plays a key role in the chemistry of the troposphere, largely determining the oxidation potential of the atmosphere through its interaction with hydroxyl radical (OH). The global CO cycle and its change are closely related to both the change of CO mixing ratio and the change of source strength. In this study, we present a six-year record of atmospheric CO concentration and stable isotopic ratios ($\delta^{13}C$ and $\delta^{18}O$) at a high northern latitude site (Westmann Islands, Iceland) between January 2004 and December 2009. A slight decreasing trend is observed for CO concentrations over time. No significant interannual variability of $\delta^{13}C$ occurred, however the interannual variability of $\delta^{18}O$ is considerable. The interannual variations of isotopic ratios reflect the variations of source contributions from the individual CO sources and the removal of CO by OH. MOZART-4, a 3-D global chemical transport model, is used to simulate CO concentration and its isotopic signature at the observation site. A tracer version of MOZART-4 which tagged for $C^{16}O$ and $C^{18}O$ from each emission region and each source was developed to analyze their contributions.

Based on the observations and model simulations, we analyzed the influences of individual CO source to the CO concentration and isotopic ratios at high northern latitudes in 2004-2009.

P-5-043 The isotopic record of Northern Hemisphere atmospheric carbon monoxide since 1950

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Key words:
carbon monoxide isotopic ratios NEEM firn

The importance and interest for measuring atmospheric CO arises from its significant role on the chemistry of the troposphere, since CO is a major sink for hydroxyl radical (OH). Hydroxyl radical is the most important oxidant in Earth's atmosphere, thus its abundance affects the lifetimes of reactive greenhouse gases and ozone depleting gases. In this study, we present a 60-year record of the stable isotopes of atmospheric carbon monoxide (CO) from firn air samples collected under the framework of the North Greenland Eemian Ice Drilling (NEEM) project. This is the first firn air CO isotope study in Northern Hemisphere. CO concentration, delta13C, and delta18O of CO were measured by gas chromatography/ isotope ratio mass spectrometry (gc-IRMS) from trapped gases in the firn. We applied LGGE-GIPSA firn air models to correlate gas age with firn air depth and then reconstructed the trend of atmospheric CO and its stable isotopic composition at high northern latitudes since 1950. The most probable firn air model scenarios show that delta13C decreased slightly from -25.8 per mil in 1950 to -26.4 per mil in 2000, then decreased more significantly to -27.2 per mil in 2008. delta18O decreased more regularly from 9.8 per mil in 1950 to 7.1 per mil in 2008. Those same scenarios show CO concentration increased gradually from 1950 and peaked in the late-1970s, followed by a gradual decrease to present day values. Results from an isotope mass balance model indicate that a slight increase, followed by a large reduction, in CO derived from fossil fuel combustion occurred since 1950. The reduction of CO emission from fossil fuel combustion after the mid-1970s is the most plausible mechanism for the drop of CO concentration

during this time. Fossil fuel CO emissions decreased as a result of the implementation of catalytic converters and the relative growth of diesel engines, in spite of the global vehicle fleet size having grown several fold over the same time period.

P-5-044 Impacts of East Asian summer monsoon circulation on aerosol distribution and its direct radiative effect

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Key words:
monsoon circulation aerosol optical depth shortwave radiative forcing

The effect of East Asian summer monsoon circulation on aerosol distribution is analyzed based on twelve years (2000-2011) daily data which contains NCEP reanalyses and aerosol optical depth (AOD) at the 550nm wave length that derived from MODIS-TERRA data set. Fu-Liou radiative transfer model is also used to calculate the shortwave radiative forcing due to the ADO anomaly at surface level and the corresponding radiative temperature-change. Four principal conclusions can be achieved and listed as follows. First, an intimate relationship is existed between the aerosol distribution and monsoon circulation, especially in areas with high concentration of aerosol, such as the lower-middle reaches of the Yangtze River and the coastal area in east China. However, the aerosol distribution in some areas is controlled by the system which is located in westerlies. Second, characteristics of the evolving of summer monsoon circulation can be properly reflected by the spatial patterns of the first MV-EOF modes of 850hPa horizontal winds, and whose effect is much greater than those of the second MV-EOF modes which can be explained as the main systems that perturb the strength of monsoon circulation. Third, although the concentration of aerosol over the ocean is much lower than the adjacent continent, the anomaly of AOD could be extreme high and this phenomenon substantiates the transfer function of monsoon circulation. Finally, the radiative effect due to the AOD anomaly , the consequence of East Asian summer monsoon circulation, is remarkable: the shortwave radiative forcing varies from -18 W m^{-2} to 15 W m^{-2} and the corresponding radiative temperature-change is approximate from -1K to 1K under the simple radiation equilibrium at surface level. The monsoon-aerosol-monsoon feedback is formed exactly because the above-mentioned radiative effect may have a further influence on East Asian summer monsoon.

Poster Presentations, Session 6

P-6-001 Atmospheric Heterogeneous Reactions of Mineral Dust Aerosols with N_2O_5 and NO_3 radicals

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Key words
Atmospheric heterogeneous reactions Mineral dust N_2O_5 NO_3

Heterogeneous reactions of N_2O_5 and NO_3 radicals with mineral dust particles have been investigated at room temperature and different relative humidities. These studies are important for assessing the importance of these reactions in the removal of NO_x from the troposphere and the chemical aging of dust aerosols during transport.

The heterogeneous interaction of N_2O_5 with Saharan dust particles was investigated using aerosol flow tubes with detection of N_2O_5 by cavity ring-down spectroscopy. The uptake coefficient was determined to be 0.02 ± 0.01 on airborne Saharan dust particles, independent of relative humidity (RH, 0-67%) and initial N_2O_5 concentration (~20-1000 ppby). Analysis of gas and particulate phase products suggests that N_2O_5 undergoes heterogeneous hydrolysis forming particulate nitrate. The independence of the uptake coefficient on the initial N_2O_5 concentration indicates that, on the seconds time scale of these experiments, the heterogeneous reaction of N_2O_5 with dust particles is not restricted to the external particle surface but internal reactive sites are also available for the N_2O_5 uptake. The particles could be deactivated with respect to N_2O_5 uptake only when pre-treated with very high levels of HNO_3.

The uptake of NO_3 radicals to Saharan dust particles was investigated using a novel relative rate method. The use of cavity ring-down spectroscopy to detect both NO_3 and N_2O_5 enabled the measurements to be carried out at low mixing ratios (< 500 pptv). The uptake coefficient ratio, (NO_3 divided by N_2O_5) was determined to be 0.9 ± 0.3 for Saharan dust, independent of relative humidity and exposure time, though surface deactivation was observed for both species.

Our studies suggest that the uptake of N_2O_5 to dust particles can influence the NO_x budget and also the formation of particulate nitrate in dust-impacted regions, while the heterogeneous reaction of NO_3 radicals with dust aerosol will generally not contribute significantly to the loss of NO_3 in the troposphere or to the nitrate content of dust particles.

P-6-002 Seasonal and inter-annual changes in the computation of aura mls hcl depletion and psc-induced areas in the antarctic polar stratosphere: 2005-2010 climate-chemistry assessment. the role of clouds in the antarctic middle atmosphere

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Key words

PSC-induced Antarctic lower-middle stratosphere Antarctic Main chlorine reservoir Hanson and Mauersberger [1988]' TNAT threshold Drdla and Mueller [2010]'s TACL threshold HCl deficits vs Temperature synoptic fluctuations satellite observations sulfate aerosol

An examination of the seasonal and spatial distribution of Polar Stratospheric Clouds (PSCs) inferred from standard temperature profiles in the lower-middle atmosphere above Antarctica, as derived from the Earth Observing System (EOS) Aura Microwave Limb Sounder (MLS) satellite observations and NCEP/NCAR assimilations, is provided. Chemical volume mixing ratio (VMR) observations of EOS Aura MLS v2.2 hydrogen chloride (HCl) were used to show the interannual variability of PSC formation with respect to stratospheric chlorine partitioning during five Southern Hemisphere Antarctic seasons from 2005 to 2009. A remarkable first set of results, obtained from an algorithm developed for modelling HCl depletion areas in the Antarctic polar vortex region, and based on satellite observations, is presented. In particular, the analysis of HCl concentration data obtained from 2006 indicated that the area processed for HCl was larger than the area of PSC during some periods of Antarctic winter, and that this result was robust with respect to the various PSC formation and HCl depletion thresholds utilized. The results suggest that an underestimation in chlorine activation area can occur when temperature thresholds for PSC formation thresholds are employed. The work presented here also evaluated chlorine activation via sulfate aerosol (SA) in the Southern Hemisphere 2006 stratosphere, based on satellite measurements of water vapor (H_2O) and constant values of SA, by implementing the TACL formula of Drdla and Müller [2010] in contrast to the TNAT formula of Hanson and Mauersberger [1988]. The results indicated that the former formula was not completely sufficient for accurately modeling areas of depleted HCl and chlorine deactivation for all pressure surfaces in the Antarctic stratosphere.

Based on the results of this study, the role of SA in chlorine activation appears to be more important at lower altitudes than for areas higher in the stratosphere.

P-6-003 Optical property analysis of diesel exhaust particles using a three wavelength photoacoustic soot spectrometer

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Key words
optical properties of earosol diesel exhausts partials

[Introduction] Diesel exhausts partials (DEP) are known as one of main anthropogenic source of element carbon (EC), and organic carbon (OC). The light absorption enhancement of black carbon (BC) is generally considered to be increased by coating with OC, which is called lensing effect. Recently, light-absorbing OC, “brown carbon”, involving humic-like substance (HULIS), organonitrate, and nitro-aromatics etc., has been proposed as a source of significant absorption, particularly in the near-UV. However, contributions of lensing effect and brown carbon for DEP have not been well known. In this study, a photoacoustic soot spectrometer three wavelength (PASS-3) has been applied to research optical properties of DEP.

[Methods] A diesel engine was operated in a car driven on a chassis dynamometer with an urban driving mode (JE05) and constant speed mode (0 km/h or 70 km/h). The diesel exhaust was diluted and then sent through a heater before the measurement of optical properties. Absorption and scattering coefficients at 405, 532, and 781 nm of the DEP are measured by the PASS-3. Size distributions of DEP before and after heating are also measured by two scanning mobility particles sizers (SMPS) during constant mode experiments.

[Results and Discussion] For the JE05 mode, optical properties were measured with inlet temperatures of 20, 100, and 300 degC. Enhancement of scattering coefficient was observed during acceleration and deceleration patterns before and after high speed driving (~80 km/h) at 20 and 100 degC. This enhancement was not detected at 300 degC indicates that emission of volatile OC compositions increased during the patterns. The Angstrom exponent (AAE) values between 405 and 532 nm are found to increase during the high speed period, while the AAE values are almost constant at 300 degC. The contributions of light absorption by OC to total light absorption at 405 and 532 nm are estimated to be ~15% and <5%, respectively.

For the constant mode, optical properties of DEP were measured with variety of inlet temperatures between 20 and 400 degC. No significant temperature dependence of AAE between 405 and 532 nm was found in both case of idling and 70 km/h patterns. This indicates that the contribution of light absorption by OC is small (<6%). Light absorption increasing by coating during idling and 70 km/h patterns are estimated to be 20 and 15%, respectively.

Recently, Dr. Inomata and co-workers (NIES, Japan) detected nitro-aromatics in gas and particle phases of diesel exhausts from the same car. The concentrations of nitro-aromatics in gas phase are significantly high during the same periods, when the light absorption by OC was found in the present study. Therefore, nitro-aromatics in DEP can be considered as plausible sources of light absorbing OC, observed in this study.

P-6-004 Ammonia uptake by pure and secondary organic aerosol particles:

dependence on particle phase

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Key words
organic aerosol laboratory experiments

Chemical aging of organic aerosol particles, which is induced by uptake of reactive molecules, is known to alter their important characteristics, including hygroscopicity and optical properties. Recent studies demonstrated that secondary organic materials (SOM) particles are likely semisolid, raising a question if the uptake process is restricted by slow bulk diffusion. In this study, phase dependent ammonia uptake by pure and secondary organic aerosol particles was investigated. Crystalline pure organic particles were generated using a newly-developed particle generator, and its phase was regulated by controlling water exposure history. Particles were exposed different concentration levels of ammonia in a mixing reactor, and the particle phase ammonium was subsequently quantified using a high-resolution time-of-flight aerosol mass spectrometer (AMS). In addition, comparison of the AMS data with mobility size distribution data provides qualitative information on particle phase and morphology. No measurable ammonia uptake was observed for crystalline particles of the pure organic test compounds, while ammonia uptake was observed for aqueous and semisolid particles. Ammonia uptake by semisolid particles was significantly smaller than that observed for aqueous particles, likely due to slow mass transfer processes. In addition, ammonia uptake by SOM particles originated from biogenic precursors, those are known for their solid-like mechanical properties, were generated using the Harvard Environmental Chamber (HEC). Ammonia uptake by biogenic SOM particles was not a fast process at low relative humidity (RH) condition, consistent with the result for solid and semisolid pure organic particles. Ammonia uptake was enhanced at high RH, likely due to decreased viscosity. The results presented here demonstrate that uptake of reactive species by semisolid SOM particles is limited by slow bulk diffusion.

P-6-005 Spatial and seasonal variability of aerosol organic matter-to-organic carbon mass ratios in Chinese cities

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Key words

organic aerosol organic carbon oxalic acid

We calculated the organic matter to organic carbon mass ratios (OM/OC mass ratios) in $PM_{2.5}$ collected from 14 Chinese cities during summer and winter of 2003 and examined their variability. The OM/OC mass ratios were calculated two ways. Based on a mass balance method, the calculated OM/OC mass ratios averaged 1.90 0.53 in summer and 1.83 0.25 in winter, with no significant spatial differences. A second set of OM/OC mass ratios was calculated from identified organic compounds extracted from $PM_{2.5}$ using dichloromethane/methanol and water. The calculated OM/OC mass ratios averaged 1.75 0.13 in summer and 1.59 0.18 in winter, indicating that Chinese urban aerosols are more oxidized in summer. We found that oxalic acid is the most important driver to increased OM/OC mass ratios in both summer and winter, due to its high oxidation state and high concentrations in aerosols. In winter, levoglucosan from biomass burning also contributed significantly to enhanced OM/OC mass ratios in northern Chinese cities due to its high molecule/carbon mass ratio and relatively high concentrations. We reported, for the first time, a high correlation between measured aerosol oxalic acid and zinc concentrations in summer (R=0.90). This suggests that (1) the sources of major oxalic acid precursors may coincide with zinc emissions, or (2) that a large fraction of aerosol oxalate exists in the form of zinc oxalate. On average, organic aerosol constituted 36% and 34% of Chinese urban $PM_{2.5}$ mass in summer and winter, respectively.

P-6-006 Photosensitized radical formation at the air/aerosol interface: impact on aerosol formation and growth.

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Key words

dust photochemistry SOA growth

Aerosols are still considered as being one of the less understood atmospheric objects and are therefore the focus of many studies. Currently, it is believed that aerosols are being formed and growth subsequently to a series of gas phase processes leading to the formation of products having low (or even very low vapour pressures). For example, the oxidation of SO_2 by gaseous OH to H_2SO_4 leads to nucleation events, while the oxidation by the same radical of VOCs leads to a full series of products able to change the organic content of aerosols and their ageing.

In this presentation, we are going to revisit this concept and suggest a few other possible pathways that do not involve gas phase chemistry but still lead to particle formation and growth.

These processes will exemplified in two cases considering the long wavelength chemistry of mineral dust as a pathway leading to nucleation events and SOA photochemistry inducing particle growth.

P-6-007 Radical loss in the atmosphere from Cu-Fe redox coupling in aerosols

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Key words
free radical chemistry aerosol uptake aerosol chemistry

The hydroperoxyl radical (HO_2) is a major precursor of OH and tropospheric ozone. OH is the main atmospheric oxidant, while tropospheric ozone is an important surface pollutant and greenhouse gas. Standard gas-phase models for atmospheric chemistry tend to overestimate observed HO_2 concentrations, and this has been tentatively attributed to heterogeneous uptake by aerosol particles. It is generally assumed that HO_2 uptake by aerosol involve conversion to H_2O_2, but this is of limited efficacy as an HO_2 sink because H_2O_2 can photolyze to regenerate OH and from there HO_2. Joint atmospheric observations of HO_2 and H_2O_2 suggest that HO_2 uptake by aerosols may in fact not produce H_2O_2. Here we propose a catalytic mechanism involving coupling of the transition metal ions (TMI) Cu(I)/Cu(II) and Fe(II)/Fe(III) to rapidly convert HO_2 to H_2O in aerosols. We implement this mechanism in two global chemistry models (GEOS-Chem and GFDL AM3). Our results show significant improvements in several aspects: (1) this mechanism helps to correct the long-standing model underestimates of CO in the extratropical northern hemisphere (by 20-30 ppb); (2) it helps to correct general model overestimates of summer surface ozone over the eastern US (by 3-5 ppb) and over East Asia (up to 20 ppb); (3) it improves the global mean OH (decrease by 12% from 1.24×10^6 to 1.10×10^6 molecules cm^{-3}) as well as the northern-to-southern hemisphere ratio of model OH. Our work thus suggests that this mechanism may have a major and previously unrecognized impact on atmospheric oxidant chemistry, with profound implications for global air quality and climate radiative forcing.

P-6-008 Identification and characterization of atmospheric organic aerosol sources by parallel nuclear magnetic resonance (NMR) spectroscopy and aerosol mass spectroscopy (AMS) measurements

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Key words
organic aerosol Nuclear Magnetic Resonance Aerosol Mass Spectrometer

Organic compounds constitute a large fraction of submicrometer atmospheric aerosol particles mass at the global scale (up to 90%). Therefore, their accurate quantification and source apportionment are necessary in order to determine their role in the environment and to conceive efficient abatement strategies. Outside urban areas, oxidized organic aerosols (OOA) constitute the dominant fraction of the organic aerosol. Within the EUCAARI (European integrated project on Aerosol Cloud Climate Air Quality Interactions) project, the OOA composition within submicrometric aerosol particles was investigated during nine intensive observation periods (IOPs) in nine different field stations over Europe, by means of proton-Nuclear Magnetic Resonance spectroscopy (1H-NMR) and aerosol mass spectrometry (AMS) with the aim of characterise the organic aerosol and its sources.

Factor analysis of spectra for the identification of a small number of recurrent chemical classes was performed. Some recurrent profiles were identified during the different IOPs and their contribution to the total mass of organic aerosol were quantified.

Comparison of NMR results with those of Positive Matrix Factorization (PMF) applied to a parallel AMS dataset showed that the NMR factors during specific IOPs broadly match the chemical classes already identified by AMS. In particular, there is an overall good overlap between the two techniques in tracing wood burning products and atmospheric humic-like (HULIS) compounds.

Our findings indicate that factor analysis applied to NMR atmospheric datasets can efficiently complement AMS in lumping the complex oxidized organic mixtures into chemical classes characterized by specific sources or ageing states. More specifically, NMR spectroscopy provides a discrimination between aromatic and aliphatic structures, which is critical for the quantification of biomass burning products, and for discriminating biogenic from anthropogenic SOA.

P-6-009 Heterogeneous and Multiphase Formation Pathways for Gypsum in the Atmosphere

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Key words
gypsum heterogeneous and multiphase reaction

Sulfate aerosols are significant components of the tropospheric aerosol mass from various environmental viewpoints. Anthropogenic sulfate is estimated to be a major contributor to the earth's radiative forcing by scattering and reflecting the solar radiation. Its cooling effect is comparable in magnitude to anthropogenic greenhouse gas forcing. Field measurement results showed that gypsum ($CaSO_4$•$2H_2O$) is a major sulfur-containing component of atmospheric particulate matter. To date, however, mechanism for its formation pathways in the atmosphere is still not well known.

In this study, we used in situ Raman spectroscopy to the heterogeneous reaction on calcite and the humidifying process of mixtures containing Ca^{2+} and SO_4^{2-} at room temperature. Several potentially important formation pathways for gypsum in atmospheric aerosols are proposed. In particular, the presence of hygroscopic components, such as $(NH_4)2SO_4$ and $Ca(NO_3)_2$, are necessary for the conversion of calcium carbonate ($CaCO_3$) upon heterogeneous reaction of either SO_2 + O_3 or SO_2 + NO_2 as well as anhydrous calcium sulfate ($CaSO_4$) to gypsum ($CaSO_4$•$2H_2O$) under ambient condition. We found that a deliquescent layer is crucial for the formation of gypsum from Ca^{2+} and SO_4^{2-} ions. Besides the need for a deliquescent layer, this study also provides definitive evidence that synergistic effects in the physical and chemical processing of aerosol particles have a significant effect on their final chemical composition, mixing state and hygroscopic behavior which dictates the environmental and climate impacts of the resulting aerosol. Interestingly, the atmospheric aging processes identified here are analogous to those used in the industrial flue gas desulfurization (FGD) process and thus can be considered nature's way of removing gas-phase pollutants from air in this desulfurization pathway.

P-6-010 The role of organic carbon in the sunlight enhanced atmospheric aging of soot by O_2

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Key words
organic carbon soot photochemistry

Soot originated from incomplete combustion is a mixture of elemental carbon (EC) and organic carbon (OC). Soot particles are ubiquitous in the atmosphere and have important climatic and health effects. Once emitted into the atmosphere, soot undergoes aging processes through the uptake of reactive gases and photochemical reactions. Aging processes of soot during long range transport result in modification of composition, morphology, hygroscopic and optical properties of soot, subsequently its climatic effects. Compared to the uptake of gas-phase species by soot, the modification of soot during aging processes is not so clear. Particularly, previous works were only focused on the interactions between soot and gaseous pollutants in the dark.

Here we report the enhanced aging process of soot by molecular O_2 under simulated sunlight irradiation using in situ attenuated total internal reflection infrared (ATR-IR) spectra. Soot residues after heated at 300 °C or extracted with n-hexane shows very low photo-reactivity towards O_2, suggesting that the active components toward photo-oxidation should be OC rather than the carbon skeleton in soot. The fuel/oxygen ratio can significantly modify the reactivity of soot by influencing the degree of soot surface oxidation and content of OC on soot. Due to the formation of highly polar species containing oxygen, heterogeneous photochemistry can promote interactions between soot and water. Thus, it will enhance a series of changes in the properties of soot such as CCN and IN, transport of soot in the atmosphere, light scattering and absorption, and precipitation. The photochemical oxidation of OC on soot by 20% O_2 is faster than that by 100 ppb O_3. Considering the abundance of O_2 in the troposphere and its higher photo-reactivity, the photochemical oxidation by O_2 under sunlight irradiation should be a very important aging process of soot.

P-6-011 Alpha-oxoketenes from isoprene efficiently convert HO_2 into OH radicals

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Key words
isoprene OH radical HO_2 radical

4-Hydroperoxy-2/3-methyl-but-2-enals (HPALDs) have been predicted to be produced by 1,6-H shift isomerisation of Z-delta-hydroxy-isoprenylperoxy radicals in the atmospheric oxidation of isoprene by OH at low NO (Peeters et al., Phys. Chem. Chem. Phys. 11, 5935, 2009). They have recently been observed and recognized as important intermediates in isoprene oxidation (Crounse et al., Phys. Chem. Chem. Phys. 13, 13607, 2011) although the peroxy isomerisation rate and HPALD yield as well as the contribution to OH recycling still remain very uncertain. The HPALD formation and subsequent photochemistry involving very fast photolysis processes have been proposed to result overall in 2 HO_2 and up to 3 OH radicals, with alpha-oxoketenes as main products (Peeters and Muller, Phys.

Chem. Chem. Phys. 12, 14227, 2011). On the other hand, recent modeling studies suggest that the very high OH observed in isoprene-rich areas could be explained (partly) by unknown compounds reacting fast with HO_2 and converting it into OH (Lu et al., Atmos. Chem. Phys. Discus. 11, 11311, 2011; Whalley et al., Atmos. Chem. Phys. 11, 7223, 2011). However, at present no VOCs or OVOCs are known to react fast with HO_2.

In this work, the reactions of HO_2 with the alpha-oxo-ketenes from the HPALDs have been investigated yheoretically. The potential energy surfaces, established using suitable DFT/ab initio methods, reveal that HO_2 adds to the alpha-oxo-ketenes in a three-step process over submerged transition states, forming a peroxy radical. The theoretically estimated rate constants are sufficiently high that reaction with HO_2 should be the predominant removal route of the oxo-ketenes in the conditions of interest. The subsequent chemistry of the peroxy product is expected to result in additional removal of HO_2, and to yield overall up to 3 OH radicals. Modeling results, using the IMAGES CTM, are presented on the impact of these reactions on OH recycling and HO_2 levels in isoprene-rich areas for various scenarios regarding the isoprene-peroxy isomerisation rates and the yields of the HPALDs and their alpha-oxoketene products.

P-6-012 The Effects of Environmental Factors on Secondary Organic aerosol (SOA) formed from atmospheric photo-oxidation reactions

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Key words
Environmental factor secondary organic aerosol size distribution

To understand the effects of environmental factors, such as the light intensity, reaction time, the concentration of reactant on the growth of secondary organic aerosol is very important. In this work, the effect of environmental factors on SOA formed from hydroxyl radical (OH•) initiated photooxidation of biogenic volatile organic compounds (BVOCs) was investigated by using a home-made smog chamber. The size distribution of SOA particles from the above reaction was measured using aerodynamic particle sizer spectrometer (APS). The effect of radiation time, reactant concentration, and CH_3ONO concentration on SOA formation from biogenic volatile organic compounds is evaluated by performing photooxidation experiments under varying conditions. Experimental results show that SOA formed from OH-initiated photooxidation of BVOCs is predominantly in the form of fine particles, which have diameters less than 2.5 μm. These fine particulate matters are more easily deposit in the lung of the human being, and do great harm to the health. This paper also highlighted that both the number and mass concentration of SOA particles were increasing with the prolonging of the radiation time, the increasing of light intensity, and the increasing of the reactant concentrations. The results could provide useful information to infer possible emission sources of atmospheric particles in future field measurement.

P-6-013 Study optical properties of secondary organic aerosol using broadband cavity spectroscopy

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Key words
IBBCEAS SOA Optical properties

Near-ultraviolet optical properties of anthropogenic and biogenic secondary organic aerosol (SOA) are poorly characterised but could significantly influence local atmospheric photochemistry and the earth's radiative balance. This work describes a novel and sensitive spectrometer based on incoherent broadband cavity-enhanced absorption spectroscopy (IBBCEAS) and its application to investigate the optical properties of SOA. The system was used in a simulation chamber to study the early stage photochemical oxidation of isoprene by OH under high NO_x conditions. The aerosol extinction, which was calculated by subtracting the absorption of gas phase absorbers from the total extinction, was strongly correlated with the aerosol particle diameter. This work extends the IBBCEAS technique to the shortest wavelengths yet reported while retaining high sensitivity across a wide spectral window (320 to 450 nm). We evaluate the usefulness of the chamber approach and discuss the potential of the IBBCEAS method for field observations to characterise the radiative properties of aerosols. The approach is most likely to be valuable in megacities with high aerosol loadings, such as Beijing and Shanghai.

P-6-014 Photo-enhanced HONO production from the heterogeneous hydrolysis reaction of NO_2 on the ammonium sulfate surface

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Key words
photo enhanced HONO production

The photo-enhanced HONO formation is a major concern of the HONO source. As one of the widely discussed photo-enhanced HONO formation route, photo-enhancd NO_2 heterogeneous hydrolysis reaction has been in dispute. To exam this route on the surface of real atmospheric particulate matter components is with the major scientific and practical significance. This study observed in a flow tube reaction system on ammonium sulfate coating a photo-enhanced HONO formation and the photo-enhanced HONO is mainly, as suggested, generated from photo-enhancd NO_2 heterogeneous hydrolysis reaction. Since, on one hand, experiments ruled out other routes, such as adsorption state nitrate photolysis reaction and photo-enhanced reduction reaction on the surface of organic matter. On the other hand photo-enhance NO_2 uptake happened to be twice the photo-enhanced HONO formation. This result suggests that of NO_2 photo-enhanced heterogeneous hydrolysis reaction is potentially important daytime HONO source in atmospheric conditions.

P-6-015 Uptake of glyoxal by organic and inorganic seed aerosols: Optical, physical and chemical properties of the product aerosols

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Key words
glyoxal heterogenous reactions

Constraining the optical properties of secondary organic aerosol (SOA) is a major challenge since they often consist of a mixture of several components with various internal structures, including homogeneous mixtures and core/shell structures. The importance in understanding the optical properties of aerosols is their effect on the magnitude and uncertainty of Earth's total radiative forcing and climate change. SOA form by the condensation of less-volatile gaseous compounds onto pre-existing aerosols, and by heterogeneous and multi-phase reactions.

The heterogeneous reaction between glyoxal gas and three atmospheric aerosol types; ammonium sulfate (AS), glycine and glycine-AS 1:100 was studied. The optical extinction cross section at λ=355nm and mobility size increased following the reaction under a broad range of RH values (30-90%), indicating that the reaction is relevant for a wide range of atmospheric conditions.

We propose that at low RH values, below the deliquescence point, the reactions occur in interfacial monolayers of water, supporting previous findings about the importance of interfacial water in heterogeneous.

The reactions exhibit a trend of increasing growth in physical and optical cross sections with decreasing seed aerosol size, as well as a clear dependence on ambient RH values (Figure 1). For small particles with near-zero extinction efficiency (Qext) values, the reaction induces the greatest increase in optical extinction cross section (up to 2 orders of magnitude enhancement) due to a combined effect of changes in optical properties and in size, resulting in a change in the location on the Mie curve.

AMS analyses of the reactions show that the main reaction products are glyoxal oligomers, and a small contribution from the formation of various C-N compounds, identified as imidazoles.

Our results suggest that unlike previously postulated, the reactions at RH values below deliquescence occur on interfacial water layers and their optical enhancement is mainly due to enhanced scattering and geometric cross section growth.

This study contributes to the understanding of the effect of the reactions on the optical properties of the aerosols in the atmosphere by demonstrating that the heterogeneous reactions between abundant atmospheric components may alter the aerosols' optical, physical and chemical properties on short timescales (~ 1 hour) and may have substantial implications on the radiative effects of these aerosols.

P-6-016 Isoprene chemical mechanism evaluation using ensemble observations of ambient HO_x concentrations during the HO_xComp field campaign

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Key words
isoprene chemistry isomerization OH radical regeneration ensemble observations

Daytime OH concentrations in ambient air were observed by multiple instruments for three days of the HO_xComp field campaign held in Jülich, Germany, in July 2005. The concentrations were compared with box-model simulations using different assumptions for isoprene chemistry and for additional hydrocarbons to explain the observed OH reactivity. The agreement in OH concentrations with the Base run was good (within 33%), where the isomerization of isoprene peroxy radicals was not taken into account, suggesting that strong radical production to explain OH in the presence of isoprene at low NO, as proposed for the cases for the measurements in Suriname and Pearl River Delta (PRD) was not required for HOxComp. Introducing isomerization of isoprene peroxy radicals incurred model overprediction of radical concentrations with respect to OH observed by a CIMS instrument and two LIF instruments. The degree of overestimation in OH could be diminished only when reactive alkanes (HC8) were solely introduced to the model to explain the missing fraction of observed OH reactivity. An important difference in the chemical conditions at HOxComp is the fact that the measurement site experienced fresh isoprene emissions that were only a little photochemically aged. This may be an indication that the unexplained large OH concentrations in Amazonia and PRD were caused by second- or third-generation products from VOC oxidations.

The HO_2 levels measured with an LIF instrument, for which the degrees of interferences from various RO_2 radicals were known, were compared with the modeled $HO_2(*)$, taking into account the sensitivity toward RO_2. The modeled $HO_2(*)$ levels in the Base run were higher than the observations by a factor 1.9, indicating an overprediction of the HO_2 to OH recycling in the base case model. This and the good model-measurement agreement for OH imply a missing OH source. Modeled OH and $HO_2(*)$ could only be matched to the observations by addition of a strong unknown loss process for $HO_2(*)$ that recycles OH at a high yield. The $HO_2(*)$ discrepancy was even enhanced by the introduction of the isoprene isomerization mechanism. Therefore the isomerization of isoprene peroxy radicals at the rates proposed by the Leuven group was therefore regarded as unlikely.

P-6-017 Light-activated chemical reactions trigger atmospheric particle growth

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Key words
SOA growth aerosol photochemistry

Atmospheric aerosols impact global climate, influence the hydrological cycle, and adversely affect human health. However, the processes that drive aerosol growth are among the most poorly understood topics in atmospheric science. The traditional view of aerosol growth holds that certain gases in the atmosphere react and produce semi-volatile organic compounds that can, under the right conditions, condense onto existing surface airborne particles.

Here, we show experimental evidence of a new photoinduced pathway that leads to particles growth, using a flow tube reactor, of non condensable gases without any requirements of gas phase oxidation.

We investigated the effect of near-UV irradiation on the change in size, mass and chemical composition of organic aerosol particles containing photoactive compounds when they are exposed to volatile organic compounds (VOCs), such as terpenes. Fast heterogeneous chemical reactions occur at the surface of secondary organic aerosols (SOA) that are activated by UV-A light, and lead to the uptake of non condensable VOCs. Experimental growth rate values are in agreement with atmospheric observations; suggesting that this photochemical process provides a new pathway to explain particle growth and should be considered by models.

P-6-018 Investigation of isoprene photooxidation in the HO_2-controlled regime using PTR-TOF-MS

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Key words
Isoprene photooxidation low NO_x methyl vinyl ketone methacrolein

Isoprene is the dominant biogenic non-methane hydrocarbon in the atmosphere. The atmospheric oxidation of isoprene affects the oxidation capacity of the atmosphere and secondary organic aerosol formation. Photo-oxidation of isoprene was investigated in the Harvard Environmental Chamber at steady-state mode using H_2O_2 as the primary HO_x source. The concentrations of isoprene, H_2O_2 and NO were precisely controlled such that the isoprene peroxy radical

chemistry was dominated by the HO_2 pathway. There are few other laboratory studies that specifically probe this reaction channel, which is important in remote regions of the atmosphere with low NO_x levels. A proton-transfer-reaction time-of-flight mass spectrometer (PTR-TOF-MS) equipped with switchable reagent ion capability (SRI, including H_3O^+, NO^+ and O_2^+) was used to measure isoprene and its oxidation products. Isomeric ketones and aldehydes, such as the two major isoprene oxidation products methyl vinyl ketone (MVK, C_4H_6O) and methacrolein (MACR, C_4H_6O), which cannot be separated using the normal H_3O^+ reagent ion, were successfully differentiated using NO^+ at optimized drift tube conditions. In addition, a novel method was developed to separate the contributions of multiple different chemical species detected at the same mass-to-charge ratios as the measured MVK and MACR product ions. The MVK and MACR yields from HO_2 pathway were hence separately quantified. Since the production of MVK and MACR via the HO_2 pathway may be accompanied by HO_x recycling, the quantified yields of MVK and MACR are valuable to understand possible HO_x recycling mechanisms in remote regions of the atmosphere with low NO_x level.

P-6-019 Mechanistic Insights into Tropospheric Ozonolysis of Short Chain Alkenes: Radical and Carbonyl Product Yields

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Key words
alkene ozonolysis OH

Gas-phase alkene ozonolysis has been the subject of considerable research interest over several decades owing to its key role in the Earth's tropospheric chemistry. Alkene ozonolysis leads to the formation of a wide range of functionalised oxygenated products, including carbonyls, organic acids and hydroperoxides. Larger biogenic alkenes, notably terpenes (C_{10}) and sesquiterpenes (C_{15}), are known to lead to the production of semi volatile oxygenated compounds, that have been detected in secondary organic aerosol. Alkene-ozone reactions are widely recognised as a non-photolytic source of OH, HO_2 and RO_2 radical products, initiating further oxidation in the troposphere and contributing to the overall tropospheric HO_x budget.

In this study we report the radical (OH, HO_2 and RO_2) and stable co-product yields for ethene, propene, 1-butene, 2-methylpropene, cis-2-butene, trans¬-2-butene and 2,3-dimethyl-2-butene and isoprene ozonolysis. The experiments were performed under tropospherically relevant conditions and as a function of humidity, in the European Photoreactor (EUPHORE) chamber, using a suite of instrumentation including Chemical Ionisation Reaction Time-of-Flight Mass Spectrometry (CIR-TOF-MS) and laser induced fluorescence (LIF) to measure VOC/OVOCs and radical products respectively. Experiments were performed in the absence / presence of radical scavengers, in order to suppress side reactions. The measurements were used to derive stable product yields using a detailed chemical mechanism based upon the Master Chemical Mechanism (MCMv3.2) with mechanistic updates from the recent literature.

Reaction rate coefficients, radical yields and stable product yields are compared with those simulated, and the implications for atmospheric oxidant levels are presented. Directly measured OH yields are found to be in good agreement with existing literature, supporting the widely accepted hydroperoxide mechanism. In the presence of OH scavenger, the measured HO_2 yields are lower than those inferred in recent studies, while yields in the absence of OH scavenger are biased high owing to the interference from the peroxy radicals formed from OH + alkene reactions within the LIF instrument. Mechanistic insights of alkene ozone reactions and their atmospheric implications are presented.

P-6-020 Investigation of heterogeneous reactions between SO_2/O_3 mixing gas and individual $CaCO_3$, Ca $(NO_3)_2$ particles by Micro-Raman Spectrometry

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Key words
heterogeneous reaction SO_2 O_3 $CaCO_3$ $Ca(NO_3)_2$ individual particle Micro-Raman Spectrometry

$CaCO_3$ is one of the most important components of the atmospheric mineral aerosol. $CaCO_3$ particles can undergo heterogeneous reactions with nitrogen-containing gases (e.g., NO_2, gaseous HNO_3 and N_2O_5) in the atmosphere during long-range transport, producing $Ca(NO_3)_2$. $Ca(NO_3)_2$ is much more hygroscopic than $CaCO_3$, and may have a reactive activity different from $CaCO_3$ in the atmosphere.

Heterogeneous reactions between SO_2(50PPM)/O_3(50PPM) mixing gas and individual $CaCO_3$, $Ca(NO_3)_2$ particles (with the geometric diameter of about 6 to 7 micrometers) under various RH conditions were investigated using Micro-Raman Spectrometry and flow reaction system. Chemical composition and microscopic morphology of the individual particles were determined in situ with Micro-Raman spectrometer during the reactions. Individual $CaCO_3$ particles started reaction with SO_2/O_3 mixing gas at RH=55%, producing $CaSO_4$. The reactive uptake coefficient (γSO_2) was in the order of 10-7 to 10-8 using the geometric surface area of the individual particles. Individual $Ca(NO_3)_2$ particles could react completely with SO_2/O_3 mixing gas at RH values of $\geq$ 15%. The product was $CaSO_4$. The reactive uptake coefficient (γSO_2) was in the order of 10-6 to 10-7 using the geometric surface area of the individual particles. The reaction rate increased with the RH. At RH=80%, individual $Ca(NO_3)_2$ particles with the geometric diameter of 6 micrometers could react completely with SO_2/O_3 mixing gas in 480 minutes to produce $CaSO_4$. Under the same RH and concentration of SO_2 and O_3, the reaction rate of individual $Ca(NO_3)_2$ particles with SO_2/O_3 mixing gas was much higher than that of individual $CaCO_3$ particles with SO_2/O_3 mixing gas. Thus the formation of $Ca(NO_3)_2$ from $CaCO_3$ particles could accelerate the transform of S(IV) to S(VI) in the atmosphere. The possible gaseous product of the reaction between SO_2/O_3 mixing gas and $Ca(NO_3)_2$ particles was HNO_3, indicating the heterogeneous reactions between SO_2/O_3 mixing gas and $Ca(NO_3)_2$ particles are sinks of SO_2 and O_3 and a potential source of gaseous HNO_3, and might have important impacts on the formation of acid rain and atmospheric oxidizability.

P-6-021 Heterogeneous reactions of gaseous hydrogen peroxide on acidic gas-aged calcium carbonate particles: effects of relative humidity and surface coverage of coating

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Key words

Heterogeneous reactions Atmospheric hydrogen peroxide Calcium carbonate Mineral dust Atmospheric aging

Atmospheric aging and processing appears to alter physical and chemical properties of mineral dust aerosol and thus its role as reactive surface in the troposphere. Yet, previous studies in the atmosphere have mainly focused on the clean surfaces of mineral dust aerosol, and the reactivity of aged mineral dust toward atmospheric trace gases is poorly recognized. This work presents the first laboratory investigation of heterogeneous reactions of gaseous hydrogen peroxide (H_2O_2), an important atmospheric oxidant, on the surfaces of HNO_3 and SO_2-aged calcium carbonate ($CaCO_3$) particles as surrogates of atmospheric mineral dust aged by acidic trace gases. It is found that the aging of the calcium carbonate particles with HNO_3 and SO_2 has a strong impact on their reactivity toward H_2O_2. On HNO_3-aged particles, the presence of nitrate acts to either decrease or increase H_2O_2 uptake, greatly depending on RH and surface coverage of nitrate. For example, as surface coverage of nitrate increases from 1 to 10 monolayers, the

uptake of H_2O_2 on the aged particles will decrease by 30–85% at 3%RH whereas increase by 20–60% at 25% RH, a factor of 1–3 at 45% RH, and a factor of 3–8 at 75% RH, compared to the clean particles. On SO_2-aged particles, the presence of adsorbed sulfite appears to enhance the intrinsic reactivity of the mineral particles due to its affinity for H_2O_2, and the uptake of H_2O_2 increases significantly relative to the clean particles, in particular at high RH. The results of our study suggest that the heterogeneous reactivity of mineral dust aerosol toward H_2O_2 and maybe other trace gases is markedly dependent on the chemical composition and coverage of the surface coatings as well as ambient RH, and thus will vary considerably in different polluted air masses.

P-6-022 The Microstructure of Ice and effective Diffusion of VOCs in snow: A laboratory study

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Key words
adsorption structure

Trace-gas emissions from the snow-pack in polar regions can have a significant impact on the chemistry and thus composition of the overlaying air. Here we describe well-controlled laboratory experiments that investigate the fundamental basis of the relevant ice-gas interaction processes on the snow surface. These findings help to improve our understanding and the ability to model atmospheric chemistry at Antarctica, in the Arctic and in the upper troposphere, where ice clouds are present. Results from these experiments further highlight the role of the microstructure of ice and compare its behavior as uptake reservoir for atmospheric gases to that of water.

We describe new results from laboratory experiments that focused on the role of grain boundaries on the transport of volatile organics and nitrous acid through a snow-pack.

Two different types of snow pack where used for the experiment:

Water was frozen in the gas phase at moderate temperatures leading to artificial snow where each snow grain is a single ice grain.Water is rapidly frozen from the liquid at harsh liquid nitrogen conditions. This produces artificial snow where 60% - 70% of the snow grains are formed from several ice grains.

For both snow samples the grain-boundary density was determined using a recently developed stereology method. This method also allowed to quantify other parameters that determine the transport, or the effective diffusion, such as the total surface area, porosity and tortuosity of the samples.

The observed effective diffusion of acetone, methanol, and nitrous acid revealed that the transport of both species was governed by gas-phase diffusion and adsorption to the snow surface. These results are discussed taking into account recent results from studies of our group on the uptake of trace gases on ice surfaces using synchrotron based, surface sensitive spectroscopic tools.

P-6-023 A High-Sensitivity Low-Cost Particle Counter

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Key words
Aerosol Sizing

We report the design of a lightweight particle counter with high sensitivity for atmospheric aerosol measurements. Principal objectives of the design were simplicity and low cost, thereby making the instrument suitable for balloon-borne applications in the troposphere and stratosphere. Particle detection and sizing are based on the measurement of the forward scattering of laser light. The separation of the laser beam and scattered light is achieved with a novel yet simple optical scheme. A laboratory prototype system using a 405-nm laser and photomultiplier tube detector has successfully detected polystyrene latex particles as small as 125 nm in diameter. Theoretical calculations suggest that a lower detectable size limit of 100 nm can be achieved with the current optical arrangement by a reasonable reduction of background scattered light. The new counter will be useful in a variety of ground-based as well as small balloon-borne applications such as profiling the Asian Monsoon and the proposed Asian tropopause aerosol layer, and sampling particles from explosive volcanic eruptions.

P-6-024 Effects of Branching on the Heterogeneous OH Oxidation of Motor Oil Particles: Impacts on Ambient Aerosol and Reaction Pathways

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Key words
aerosol chemistry heterogeneous oxidation

Motor oil serves as a useful surrogate for anthropogenic particulate emissions due to its compositional similarity to both diesel exhaust and the complex mixture of hydrocarbons observed in typical ambient aerosol samples. However, because of its complexity, detailed analysis of the composition and atmospheric degradation of complex hydrocarbon mixtures is difficult using traditional techniques. In order to improve this understanding, we subjected homogenously nucleated motor oil (15W-40) particles to heterogeneous hydroxyl radical oxidation. Samples were collected and analyzed by twodimensional gas chromatography using vacuum-ultraviolet ionization and high resolution time-of-flight mass spectrometry (GCxGC/VUV-HRTOFMS). This “soft” ionization technique allows us to classify compounds by carbon number, cyclization, and branching, providing nearly complete resolution of hydrocarbon mixtures. In addition, we use high resolution analysis to extract the hydrocarbon fraction of the oxidized motor oil particles and compare relative reaction rates of various compound classes. Branched alkanes are found to have heterogeneous oxidation rates of 1.6 ± 0.3 to 2.1 ± 0.2 times faster than straight-chain alkanes depending on degree of branching, while cyclic compounds oxidize at 0.7 ± 0.2 times the average rate of alkanes. Higher volatility alkanes are observed to oxidize faster, while cyclic compounds do not strongly exhibit volatility dependent oxidation rates. These relative rates are shown to have significant effects on aerosol composition following multi-day atmospheric aging, demonstrated by differences in branching between Pasadena, Bakersfield, and fresh diesel exhaust. High resolution analysis of the GCxGC plane is used to estimate the oxidation pathway of motor oil by parameters typically used in current models (volatility and polarity). Lower estimates for fragmentation and functionalization of this complex hydrocarbon mixture are compared to simple model compounds.

P-6-025 Analysis of aerosol characteristics during the Spring Festival

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Key words
fireworks ATOFMS

In China, the Spring Festival is the most important festival. With the shutdown of most factories and reduction of vehicle traffic during this period, emission sources related will significantly decrease. Meanwhile, large amounts of burning of

fireworks will result in a significant increase in atmospheric aerosols. Therefore, aerosols'variation characteristics will be very typical during this period. In this study, more than 10 million aerosols with diameters in the range of 0.2－2μm were statistically analyzed by ATOFMS from Jan. 19 to Feb. 10, and the main sources and chemistry characteristics of aerosols were identified. In addition, time-varied of aerosols from the burning of fireworks were discussed particularly, and aerosol concentration was also quantified combining with WPS.

P-6-026 Measurement of NO_3 using cavity enhanced absorption spectroscopy with a red LED light source

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Key words
incoherent broadband cavity-enhanced absorption spectroscopy (IBBCEAS) NO_3 radical light emitting diode (LED)

The nitrate radical (NO_3) plays a dominant role in nighttime chemistry of the atmosphere. Besides OH radical, NO_3 is regarded as an key species for its oxidation ability in the atmosphere. The local NO_3 measurements with high sensitivity and high time-resolution which are imperative and important for investigating atmospheric chemistry processes, verifying different meteorological models and atmospheric chemistry models. With the characteristic of its changeability and the limitations in present measurements, NO_3 data in different atomospheric conditions are still insufficient.

In this work, the method for measurement of NO_3 with light emitting diode (LED) based cavity enhanced absorption spectroscopy (CEAS) under laboratory conditions is described. A long light path can be provided for CEAS by using a high finesse optical resonator. The effective path length of up to 7.3km at 662.09nm is determined from the known Rayleigh scattering of He and N_2. Combining the small size optical resonator with the established analysis method of differential optical absorption spectroscopy (DOAS), the approach was demonstrated for detection of different amounts of NO3 and NO_2 in laboratory conditions in the 645-680nm spectrum range. For a 300s sampling time, the 1σ statistical uncertainty of NO_3 is generally on the order of 2ppt.

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P-6-027 Aircraft measurements of gaseous pollutants over the coastal and offshore area of Yellow Sea

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Key words
aircraft measurement gaseous pollutant Yellow Sea

In order to investigate the regional air pollutant situation as well as transport in East Asia, aircraft measurements were conducted above the coastal and offshore area of Yellow Sea in April, 2011. A Yun-12 aircraft that has a cruising speed of about 180km/h was used for all flight missions and the base airport was in Qingdao, China. During nearly one month's campaign, 9 days aircraft observations were performed and the flight hours were totally 30 hours. In this report, from April 11 to April 15, 5 days' continuous observation data of gaseous pollutants, including nitrogen oxides (NO_x = NO + NO_2), sulfur dioxide (SO_2), carbon monoxides (CO), ozone (O_3) and volatile organic compounds (VOCs) are investigated. Temporal variations of NO_x and SO_2 concentrations are found and the range is from 0.5 to 10 ppbv and 0.1 to 16 ppbv, respectively. The high concentrations of CO are measured, ranging from 0.7 to 2 ppmv. All of these three pollutant species show obvious special variations, that is the concentrations over the coastal area are higher than those observed over offshore area. The analysis result shows that the measured O_3 concentrations range from 60 to 127 ppbv and the vertical variation. The total concentrations of VOCs range from 5.8 to 14.5 ppbv which are higher than those reported in some other papers. Among the 5 flights, the obvious cases of pollutant concentrations increasing were found during 3 flights and they were on April 11, 13 and 15, respectively. With the help of the meteorological analysis and back trajectory analysis results, it indicated that the aircraft had flown through some air pollution plumes. And the flight during April 13 was a little different from the other 2 flights. During that flight, O_3 didn't show obvious change, while increasing peaks were found for NO_x, SO_2 and CO at same time. It indicated that the observed air pollution plume on April 13 probably was a relative fresh one and the O_3 forming reaction had not been much proceeded yet.

P-6-028 Elemental composition of HULIS in the Pearl River Delta Region, China: Results inferred from positive and negative electrospray high resolution mass spectrometric data

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Key words
HULIS UHRMS

The HUmic-LIke Substance (HULIS) fraction isolated from aerosol samples collected at a rural location of the Pearl River Delta Region (PRD), China, during the harvest season were analyzed by both positive and negative mode electrospray ionization (ESI) coupled with an ultra-high resolution mass spectrometer (UHRMS). With the remarkable resolving power and mass accuracy of ESI-UHRMS, thousands of individual elemental formulas were identified. Formulas detected in the positive and the negative ESI mode complement each other due to differences in the ionization mechanism and the use of both provides a more complete characterization of HULIS. Compounds composed of C, H and O atoms were mainly detected in the negative mode by deprotonation, implying their acidic properties. Tandem MS and Kendrick Mass Defect (KMD) analysis suggested that carboxyl groups are abundant in the CHO compounds. This feature is similar to those of natural fulvic acids but with relatively smaller molecular weights observed in the HULIS samples. A greater number of reduced nitrogen organic compounds were observed in the positive mode compared to the negative mode analysis. Compounds with biomass burning origin including alkaloids, amino acids and their derivatives are their probable constituents. Sulfur-containing species were dominantly detected in negative mode. The presence of sulfate fragments in the MS/MS spectra of these species and their high O/S ratios implied that they are mainly organosulfates. Organosulfates and nitrooxy-organosulfates were often the most intensive peaks in the ESI- spectra. They are believed to be products of reactive uptake of photooxidation products of reactive volatile organic compounds by acidic sulfate particles. The elemental compositions deduced from the UHRMS analysis confirm the conclusion from our previous study that biomass burning and SOA formation are both important sources of HULIS in the PRD region.

P-6-029 Simultaneous Desulfurization and Denitrification under Ultraviolet Light Irradiation

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Key words
Desulfurization   Denitrification   Ultraviolet Light Irradiation

SO_2 and NO_x are the two most important air pollutants in the atmosphere. Due to the adverse environmental and health effects caused by the emission of SO_2 and NO_x from coal-fired power plants, desulfurization and denitrification techniques (such as Wet-FGD, SCR, EBA, etc) have been investigated during the past 50 years. Among these methods, UV irradiation is simple-operation, energy-saving and environment-friendly. Most of the desulfurization and denitrification studies involving UV irradiation have been focusing on the UV/O_3, UV/H_2O_2 or UV/TiO_2 systems, in which UV lights were used to illuminate O_3, H_2O_2 or TiO_2 photocatalyst to trigger the oxidation reaction of SO_2 and NO_x. However, the effect of UV irradiation in the co-existence of oxygen and water, which are the two main components in the flue gas, has to be investigated yet. It is known that both oxygen and water can proceed with photolysis under UV (185nm & 254nm) irradiation to generate reactive oxygen species. Therefore, it is of significance to investigate the desulfurization and denitrification processes under 185/254nm UV irradiation, considering the photolysis effect of oxygen and water.

In this study, a simple and highly effective simultaneous desulfurization and denitrification method under UV irradiation was presented. The simulated flue gas composition is $N_2/O_2/H_2O/SO_2/NO_x$=87.5%/10%/2%/0.05%/0.04% (v/v) and the flow rate is 0.2 L/min. The simulated flue gas were irradiated by a 15W low-pressure mercury lamp (λmax=185 and 254 nm, with the light intensity of 254 nm 10 times higher than 185 nm) in a 17L cylinder stainless steel reactor. The SO_2, NO, NO_2 concentrations were monitored by MRU Optima 7 flue-gas detector every 10 minutes. In the desulfurization experiment, 98% of the SO_2 was effectively removed within 20 minutes. In the denitrification experiment, 97.5% of the NO_x was removed within 20 minutes. In the co-existence of SO_2 and $NO_{x,}$ SO_2 and NO_x were removed by 97% and 95% respectively within 30 minutes. The mechanisms can be proposed as follows. Upon UV irradiation, O(3P) and OH radicals were generated by photolysis reaction of oxygen and water. O_3 was formed when O(3P) reacted with O_2. The O(1D) was generated from the the photolysis of O_3 and can further react with water to generete more OH radicals. O_3 can react very fast with NO ($k_{NO}=1.96\times10^{-11}$ cm^3•molecules^{-1}•s^{-1}). •OH can react fast with SO_2 and NO_2 ($k_{SO2}=3.6\times10^{-12}$, $k_{NO2}=6.9\times10^{-10}$$cm^3$•molecules^{-1}•s^{-1}). The final products were H_2SO_4 and HNO_3, which can be easily removed or recycled. Since the experiments were carried out in room temperature (about 20 °C), the results might not be the same for the real flue gas emission condition. Higher temperature (60-90 °C), water content (8-10%) and the introduction of CO_2 (10%) are required to simulate a more real flue gas in the further research.

P-6-030 Heterogeneous Oxidation of SO_2 by Ozone on the Surface of Black Carbon Particles

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Key words
Heterogeneous reactions SO_2 oxidation Ozone Black Carbon

Black carbon (BC) produced by incomplete combustion processes is an important aerosol species. The heterogeneous reactions of gaseous pollutants (such as SO_2, O_3 and NO_x) on the surface of BC particles are of significance because these reactions can change the particles' morphology, chemical composition, hygroscopicity and optical properties, and thus alter their climate or health effect. The internal mixing state of BC particles with secondary particles (e.g., sulfate and nitrate) was observed under field investigations. Additionally, it is believed that BC particles coated with sulfate can experience large hygroscopic size and mass growth at subsaturated conditions (<90% relative humidity) and act efficiently as cloud condensation nuclei. These heterogeneous reactions have attracted attention since 1970s, mostly focusing on the reaction products and the influence of NO_x. While it is of importance to further investigate the influence of some atmospheric oxidants such as O_3 on the reaction process, including the measurements of uptake coefficient and sulfate formation rate.
We investigated the heterogeneous oxidation of SO_2 by O_3 on the surface of BC (using Printex U as the model particles) by the method of Diffuse Reflectance Infrared Fourier Transform Spectroscopy (DRIFTS). The surface product was monitored, and confirmed by Ion Chromatographic (IC) and X-ray photoelectron spectroscopy (XPS) techniques. It was found that the main product of SO_2 heterogeneous reaction on the surface of diluted BC (with one part of BC diluted in 399 parts of infrared transparent NaCl powder, denoted as BC(1:400)) was sulfate and the oxidation reaction rate could be greatly enhanced by O_3. The simultaneous presence of ozone and water vapor are necessary for the sulfate formation and the reaction rate can sustain for several hours. In the condition of 1020~ 1021 molecule/m^3 SO_2/1020~ 1021 molecule/m^3 O_3/40% RH, the steady state uptake coefficient of SO_2 on BC(1:400) was 1~ 6×10-6 using BET surface area as the reacting surface. The reaction order of O_3 was found to be 0.5. The sulfate formation rate on the surface of BC particles was calculated up to 1015 ion/(s•g), indicating that this kind of heterogeneous reaction may play an important role in altering the physicochemical properties of BC particles in the highly air-polluted weather conditions.

P-6-031 Compound specific condensation to secondary organic aerosols providing size dependent information

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Key words
SOA Condensation and growth Box modeling

As until now, modelers have mostly simulated the aerosol mass balance using one or a few compounds (most often water, sulphuric acid and one or two organic compounds) as a representation for all condensing vapors. Also the Volatility Basic Set (VBS), which does not require any compound specific information, is widely used. Those few studies that do take compound specific condensation into account, use an equilibrium partitioning approach for condensation, and generate a bulk aerosol, whereby all size information is lost. If we want to understand aerosol growth on a more process based level, we need to know what molecules contribute significantly to the aerosol mass in each size bin, and therefore compute the individual contributions from all possible candidates.

We are presenting the next version of the zero-dimensional model MALTE–BOX with implemented temperature depended saturation vapor pressures for atmospherically relevant organic compounds. MALTE–BOX includes three parts: 1) emission (using MEGAN), 2) chemistry (using KPP and MCM), and 3) aerosol dynamics (using UHMA). The boiling temperature and following saturation vapor pressure for all organic compounds listed on MCM's website has been calculated using Nannoolal’s vapor pressure method. This includes 5707 chemicals. We calculate size dependent secondary organic aerosol growth (from nucleation to accumulation mode) for an alpha-pinene + ozone chamber experiment, using a kinetic condensation approach, based on an open number of condensible organic vapors. We characterize the chemical composition of the formed aerosols. We have also investigated the sensitivity of the surface tension, vapor density, activity coefficient, mass accommodation coefficient and diffusion coefficient on the aerosol mass.

P-6-032 Aqueous chemistry and kinetics of atmospheric isoprene, its descendants and sulfur dioxide

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Key words
isoprene methacrolein oxygenated polar organic compounds sulfur dioxide aerosol rainwater

Introduction Isoprene is one of the most important precursors of organic carbon in the atmosphere. It is emitted mainly by terrestrial vegetation, as well as by sea

algae, humans and animals. Once in the atmosphere, it enters numerous chemical reactions in the gas-phase, on solid and liquid surfaces and within the liquid phases. Products bearing the isoprene parenthood have been often detected in atmospheric and smog-chamber aerosols, and in rainwater samples. Gas phase chemistry and kinetics of isoprene and its descendants is quite well understood. Heterogeneous transformations have been characterized mostly through the product studies base on smog-chamber experiments, while aqueous processes are far less understood. This work extends previous studies on kinetics and chemistry of aqueous-phase reactions of isoprene with radicals derived from sulfur dioxide, a natural and anthropogenic air pollutant, and includes a similar study done for methacrolein.

Methods

Isoprene and methacrolein were reacted with sulphoxy radical anions generated in situ by autoxidation of inorganic sulfite catalyzed by transition metal ions and/or by decomposition of Oxone by cobalt(II) cations, or by photodissociation of peroxydisulphate ions. The reactions were run in a classical stirred tank reactor or in a stirred photoreactor equipped with a mercury UV lamp. Kinetics of the reactions was followed using high-resolution UV spectroscopy as well as electrochemical determination of oxygen consumption. The rate constants were estimated using the reversed rates method. Products of reactions were analyzed using liquid chromatography with mass-spectrometric detection as well as mass spectrometric techniques including secondary fragmentation and collision-induced dissociation of individual ions. Molecular structures of the products were deduced and confirmed using true synthesized standards.

Results

The results of kinetic studies include the estimated values of rate constants for reactions of isoprene and methacrolein with sulfate radical anions, evaluated over the range of temperatures. Product studies resulted in confirmed molecular structures of reaction products, including oxygenated polar species containing sulfate or sulfite groups.

Discussion

The results obtained were discussed to evaluate plausible chemical mechanisms behind the transformations studied. Atmospheric significance of the reactions was discussed in terms of participation of aqueous bulk and surface processes in formation of oxygenated polar species occurring in ambient aerosol and rainwater samples. The oxy-sulfur groups in these species can originate from atmospheric sulfur dioxide and products of its oxidation.

Conclusions

The main conclusion from this work is that the current understanding of formation of oxygenated polar components of ambient aerosol and rainwater underestimates the role of aqueous-phase and aqueous-interface processing of trace volatile organic components of the atmosphere. The aqueous transformations of volatile organic compounds involving inorganic pollutants, such as sulfur dioxide, can increase the number of secondary organic compounds in the atmosphere.

P-6-033 Stable isotopic composition of δ15N-NO_3^- and δ18O-NO_3^- in bulk precipitation of Kathmandu valley, Nepal

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Key words
bulk precipitation Kathmandu valley sources

We collected bulk precipitation samples from different places of rapidly growing urban Kathmandu valley during June 2010- August 2011. The δ15N measured was in the range of -13.9‰ to +9.3‰ for NO_3^- with an average of + 0.03 ‰. Though seasonality was not clear positive values of δ15N during winter reveal that NO_x emitted from the vehicles sources and negative values as an influence of agricultural activities during monsoon season. The δ18O values of NO_3^- ranged from +13.0‰ to +64.8 ‰ with an average of + 34.5 ‰. Though values are in broad range of previous results, still the values are lowest for δ18O atmosphere NO_3^- based on the denitrifier method (+60.3‰ and +86.5‰). The seasonality was clear as the value reaching a minimum during (summer) monsoon due to this the value of δ18O -NO_3^- was significantly higher in non-monsoon (winter) season. The minimum δ18O value of atmospheric NO_3^- could be accepted during winter season when the area is highly polluted but still during summer monsoon too shows low which still the lowest value when the air pollution is minimum in the world. Our results emphasized either the importance of peroxy radicals' pathway where O_3 is totally replaced for the formation of δ18O values of NO_3^- or dust particles in Kathmandu valley.

P-6-034 EUROCHAMP-2 – A European Infrastructure Project for Atmospheric Simulation Chambers – An Appraisal and Future Directions

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Key words
EUROCHAMP-2 Simulation chambers

Since June 2004 the European Commission in Brussels is funding the Integrated Infrastructure Initiative EUROCHAMP (http://www.eurochamp.org), whose overall goals are:

-the integration of existing environmental reaction chambers into a Europe-wide

infrastructure

-the initiation of an effective interdisciplinary collaboration between the community of atmospheric scientists and colleagues from other disciplines (e.g. cultural heritage protection and human health).

In addition to the development of, for example, standardized data protocols, a central database for environmental chamber studies, infrared and mass spectral databases and instrument intercomparisons, the project includes two so-called Joint Research Activities (JRA).

- JRA1: Experimental Techniques

The main aim of JRA1 is the development and refinement of analytical instrumentation used in the investigation of atmospheric processes, e.g., methods for the characterization of oxygenated VOCs, atmospheric pressure ionization techniques for MS, UV and IR spectroscopic techniques in general, radical measurements, nitric acid measurements, and improved methods for the generation, detection and characterization of aerosols.

- JRA2: Development of Common Techniques for the Use of Chamber Measurements in Model Development and Evaluation

The main aim of JRA2 is the improvement of chemical modeling techniques, i.e. techniques for the evaluation and development of oxidant mechanisms and models, techniques for the evaluation and development of aerosol models, development and deployment of statistical tests and sensitivity analysis methods. A particular innovation of this activity has been the development and implementation of a modelling database Electronic lab notebook (ELN) which has been developed in parallel with AtChem Online (https://atchem.leeds.ac.uk/) making simulation of atmosphere chemistry phenomena with the MCM readily and easily accessible to the scientific community.

A very important activity within EUROCHAMP has been the so-called Transnational Access (TNA). This activity promotes scientific excellence through free access to the EUROCHAMP infrastructure for scientists within and outside EUROCHAMP. The TNAs have resulted in the use of the EUROCHAMP facilities by scientists not only from Europe but worldwide, have fostered high caliber science and have contributed extensively to our understanding of atmospheric chemical processes.

The presentation will review the current progress of the project, in particular, with respect to aforementioned very successful joint research and transnational access activities.

P-6-035 Quantification of carbonate carbon in aerosol filter samples using a modified thermal/optical carbon analyzer (M-TOCA)

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Key words
Carbonate carbon Thermal optical analysis

Measurement of carbon dioxide (CO_2) gas evolved from acidification is a method to quantify carbonate carbon (CC) in aerosols collected on quartz fiber-filters. This paper describes the installation of an addon device in a DRI Model 2001 Thermal Optical reflectance (TOR)/Thermal Optical Transmittance (TOT) Carbon Analyzer (M-TOCA) to facilitate a direct CC measurement. In each run, a maximum of 20 filter punches (each of 0.5 cm^2) were acidified with 1 mL of 20% v/v phosphoric (V) acid in a vial under a 100% helium gas environment. The CO_2 evolved was reduced to methane (CH_4) and detected by a flame ionization detector (FID). The optimum reaction kinetics were obtained under an operational temperature of 40 ^{o}C and ultrasonic agitation. Method precisions were 3.5% on average for carbonate standards ranging from 3.0 to 60.0 mg and 3.8% on average for ambient samples in masses ranging from 0.30 to 56.0 mg respectively. Method accuracy was on average 91.9%, ranging from 81.4 to 102.1%. Minimum detection limit (MDL) of the M-TOCA method was 0.048 mg cm^2, corresponding to an ambient concentration of 0.098 mg m^3 for a sampled volume of air of 7.2 m^3. The MDL is >22 times lower than the value obtained using the novel method with a regular TOCA

Comparison studies on standards and ambient samples have demonstrated that the two methods do not yield systematic differences in concentrations of the carbonate. The lower MDL value provided by the M-TOCA allows a simple, precise and accurate measurement for ambient samples having a low CC concentration.

P-6-036 Remote sensing of atmospheric aerosol composition from ground-based sun-sky radiometer measurements

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Key words
aerosol composition remote sensing

1. Introduction

Aerosol chemical composition play important role in many studies like climate change assessment and environment monitoring. Aerosols directly modify the earth's radiation balance through absorbing and scattering the incident solar light, and indirectly affect the climate by affecting formation and properties of clouds. Some aerosol components are potential threats to human health by acting as carrier for a variety of toxic and hazardous substances, which may cause serious health damage. In this study we introduce recent progresses on the remote sensing method to retrieve aerosol component fraction from ground-based radiometer belonging to China Sunphotometer Observation network (SOnet).

2. Methods

Based on the analysis of spectral behavior of aerosol refractive indices at 440, 670, 870 and 1020nm wavelengths retrieved from sun-sky radiometer remote sensing measurements in Beijing, we found that the real parts (n) show a flat spectral variation behavior, while the imaginary parts (k) have a strong wavelength dependent characteristic with k at 440 nm significantly higher than other wavelengths. This phenomenon suggests the presence of mineral dust component and supports us in extending previous 3-component retrieval algorithm (Schuster et al., 2005) to the 4-component algorithm. In practice, we assumed an internal mixture of aerosols including 4 components, i.e., light absorbing components like Black Carbon (BC) and Mineral Dust (DU), non-absorbing components like Ammonium Sulfate (AS), and Aerosol Water (AW). By fitting spectral behavior of refractive indices instead of wavelength-independent ones, we can retrieve fraction of BC, DU, AS and AW of the total atmospheric column simultaneously from remote sensing measurements.

3. ResultsPreliminary results at different aerosol conditions (such as clear, hazy and dusty days) and typical sites (e.g. megacity, industrial, arid and background sites) show considerable contrast between their aerosol compositions and agree with a priori knowledge. In addition, a detailed analysis for heavy haze events in Beijing winter was carried out. The haze aerosols are characterized by high Aerosol Optical Depth (AOD) of about 3.3 at 440nm. The chemical retrieval algorithm showed that BC, AS, DU and AW occupied about 2%, 27%, 40% and 31% respectively in Beijing winter heavy haze events. Moreover, the retrieved BC concentration was compared with the observed BC mass by aethalometer (Magee Scientific AE51) and showing good correlation. The influence of combustion generated absorption brown carbon to the dust retrieval and the rationality of

choosing ammonium sulfate to represent the non-absorbing components are also discussed.

4. Discussion and conclusion

Our study provides a tool to remotely determine chemical composition fraction of ambient atmospheric aerosols. This is especially important considering many vital processes and procedures occurred in the ambient atmosphere, instead of laboratory controlled environment, i.e. case of sampler chemical analysis. In principle, this method can also be used in current and future satellite observations, e.g. POLDER, APS-2, SGLI, DPC, 3MI etc. With the development of new generation satellite retrieval algorithm by Dubovik et al. (2011), aerosol refractive index could be determined reasonably from polarimetric satellites and thus provides possibility to map global aerosol chemical composition distribution from space.

P-6-037 Oxygen-containing polycyclic aromatic hydrocarbons (OPAHs) in ambient air samples from different ecological zones of China: influence of climate and anthropogenic activity

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Key words
Oxygenated PAHs, PAHs, air, China gaseous phase, particulate phase climate, meteorology

OPAHs are emitted together with parent-PAHs from combustion sources, but they are additionally formed from photochemical transformation of parent-PAHs by atmospheric oxidants (OH, O_3 and NO_3). The OPAHs have been frequently reported to have higher toxicity than the frequently measured parent-PAHs. Little is known about the status of contamination, composition pattern and the role of climatic/meteorological variables in the formation and partition of OPAHs in the atmosphere, especially in China.

We sampled air (gaseous and particulate phase) from temperate urban site (Xi’an), subtropical sites (rural and urban sites near Chao lake), topical site (rural and urban near Huguangyan maar lake) and temperate site (background sites near Qinghai lake). Samples were extracted by pressurized liquid extraction, fractionated/cleaned-up by silica gel based column chromatography. We quantified

15 carbonyl-OPAHs in extracts with GC-MS.

The average sum of OPAH concentrations in particulate phase vrs. gaseous phase in Xi'an, Qinghai lake, Chao lake, Huguangyan marr lake and were 5.5 vrs 19.3, 3.6 vrs 3.9, 2.6 vrs 12.1 and 0.4 vrs 8.2 ng/m3, respectively. On average the most abundant OPAHs in the gaseous phase was 1-indanone (37%), 1,4-naphthoquinone (16%) and 9,10-anthraquinone (11%) and the most abundant OPAHs in particulate phase was 6H-Benzo(c,d)pyren-6-one (24%), 9,10-anthraquinone (23%) and 7H-benzo(de)anthracene-7,12-dione (13%). Hence molecular weight plays a role in composition pattern in gaseous and particulate phases.

The sum of OPAHs in the gaseous phase was negatively correlated with the windspeed ($r = -0.63$, $p = 0.02$) and marginally to the relative humidity ($r = 0.5$, $p = 0.068$), however OPAH concentration in particulate matter showed no relationship to any meteorological factors. The gas/particle partition of OPAH concentrations was significantly correlated with relative humidity ($r =0.67$, $p = 0.009$) and marginally with the ambient temperature ($r = 0.52$, $p = 0.058$).

The results indicate that anthropogenic activity alone cannot explain the OPAH concentration pattern. We therefore hypothesize that climate/meteorological conditions and transport distance from emission source has an influence on the turnover of parent-PAHs and formation of OPAHs. The influence of meteorological factors and molecular weight on gas/particle partition is further confirmed by this study.

P-6-038 Water-soluble organic carbon, dicarboxylic acids, ketocarboxylic acids, and α-dicarbonyls, OC, EC and inorganic ions of $PM_{2.5}$ aerosols in the Qinghai Lake: implication for sources, formation, and degradation during long-range transport

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Key words
dicarboxylic acids the secondary photochemical oxidation

Aerosol samples were collected from the Qinghai Lake located in the northeastern part of Tibetan Plateau from July to August 2010. To better understand the production of water-soluble organic aerosols, the samples were analyzed for water-soluble dicarboxylic acids (C_2-C_{11}) and related compounds (ketocarboxylic acids and α-dicarbonyals), as well as organic carbon (OC), elemental carbon (EC), water-soluble organic carbon (WSOC), and water-soluble inorganic ions. Distributions of dicarboxylic acids and related compounds were characterized by a predominance of oxalic acid (29.3 – 347.6 ng m^{-3}, 148.2 ± 83.9 ng m^{-3}) followed by Malonic (9.3 – 77.6 ng m^{-3}, 27.2 ± 15.5 ng m^{-3}) and Succinic (3.6 – 29.3 ng m^{-3}, 14.5 ± 8.0 ng m^{-3}) acids. The diurnal mean contributions of diacids, ketoacids and dicarbonyls to WSOC were 44.1%, 1.3%, and 0.4%, respectively. They were several times higher than those reported in north-western cities from which air masses were transported to the Qinghai Lake, indicating an importance of photochemical processing of aerosols during a long-range transport. The diacids/WSOC and C9/diacids ratios were higher than those reported previously in different atmosphere aerosols, which may indicate that the local aerosols were chiefly influenced by the biomass burning (herbages and dung) and photochemical oxidations were enhanced because of the stronger solar radiation. This study demonstrated that diacids were primarily produced by the secondary photochemical oxidation of organic pollutants and ω-oxoacids were likely intermediates to the production of dicarboxylic acids.

P-6-039 Comparison of OH concentration measurements by DOAS and LIF during SAPHIR chamber experiments at high OH reactivity and low NO concentration

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Key words:
chamber experiments hydroxyl radical measuerment comparison

During recent field campaigns, hydroxyl radical (OH) concentrations that were measured by laser-induced fluorescence (LIF) were up to a factor of ten larger than predicted by current chemical models for conditions of high OH reactivity and low NO concentration. These discrepancies, which were observed in forests and urban-influenced rural environments, are so far not entirely understood. In summer 2011, a series of experiments was carried out in the atmosphere simulation chamber SAPHIR in Juelich, Germany, in order to investigate the photochemical degradation of isoprene, methyl-vinyl ketone (MVK), methacrolein (MACR) and aromatic compounds by OH. Conditions were similar to those experienced during the PRIDE-PRD2006 campaign in the Pearl River Delta (PRD), China, in 2006, where a large difference between OH measurements and model predictions was found. During experiments in SAPHIR, OH was simultaneously detected by two independent instruments: LIF and differential optical absorption spectroscopy (DOAS). Because DOAS is an inherently calibration-free technique, DOAS measurements are regarded as a reference

standard. The comparison of the two techniques was used to investigate potential artifacts in the LIF measurements for PRD-like conditions of OH reactivities of 10 to 30 s^{-1} and NO mixing ratios of 0.1 to 0.3 ppbv. The analysis of twenty experiment days shows good agreement. The linear regression of the combined data set (averaged to the DOAS time resolution, 2495 data points) yields a slope of 1.02 with an intercept of 0.10x10^6 cm^{-3} and a linear correlation coefficient of R^2=0.86. This indicates that the sensitivity of the LIF instrument is well-defined by its calibration procedure. No hints for artifacts are observed for isoprene, MACR, and different aromatic compounds. LIF measurements were approximately 30 to 40 % (median) larger than those by DOAS after MVK and toluene had been added. However, this discrepancy has a large uncertainty and requires further laboratory investigation. Observed differences between LIF and DOAS measurements are far too small to explain the unexpected high OH concentrations during the PRIDE-PRD2006 campaign.

P-6-040 Hydrolysis of Organonitrate Functional Groups in Aerosol Particles.

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Organonitrate (ON) groups are thought to be important substituents in secondary organic aerosols (SOAs).Model simulations and laboratory studies indicate a large fraction of ON groups in aerosol particles, but much lower quantities are observed in the atmosphere. Hydrolysis of ON groups in aerosol particles has been proposed recently to account for this discrepancy. To test this hypothesis, we simulated formation of ON molecules in a reaction chamber under a wide range of relative humidity (RH) (0 to 90%). The mass fraction of ON groups (5 to 20% for high-NOx experiments) consistently decreased with increasing RH, which was best explained by hydrolysis of ON groups at a rate of 4 day^{-1} (lifetime of 6 h) for reactions under RH greater than 20%. In addition, we found that secondary nitrogen-containing molecules absorb light, with greater absorption under dry and high-NOx conditions. This work provides the first evidence for particle-phase hydrolysis of ON groups, a process that could substantially reduce ON group concentration in atmospheric SOAs.

P-6-041 Atmospheric chemistry of $CF_3CH_2OCH_3$

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P-6-042 Atmospheric chemistry of halogenated propenes

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Introduction

Recognition of the adverse environmental consequences of the release of chlorofluorocarbons (CFCs) and Halon into the atmosphere (Molina & Rowland, 1974; Farman et al, 1985) has led to an international effort to replace these compounds with environmentally acceptable alternatives. While safeguarding the ozone layer has been at the center of these efforts, concerns related to global climate change are becoming an increasingly important consideration in the choice of alternative compounds. Saturated hydrofluorocarbons (HFCs), such as CH2FCF3 (HFC-134a), have become widely used CFC replacements. HFCs do not contain chlorine and therefore do not contribute to chlorine based catalytic destruction of stratospheric ozone (Wallington et al, 1995). Unsaturated hydrochlorofluorocarbons has been developed as a potential replacement for blowing agents CFCs and saturated HFCs in the manufacture of thermosetting foams. When compared to the corresponding saturated compound, the presence of a C=C bond leads to increased reactivity towards OH radicals, decreased atmospheric lifetime, and decreased GWP (Finlayson-Pitts and Pitts, 2000). Prior to the large-scale industrial use of unsaturated halogenated compounds an assessment of the atmospheric chemistry, and hence environmental impact, of these compounds is needed.

Experimental

FTIR-smog chamber techniques were used to study the kinetics, products and mechanisms of the Cl atom and OH radical initiated oxidation five halogenated propenes in 700 Torr of air or N_2/O_2 diluent at 296 ± 2 K. The reactions of Cl atoms and OH radicals with halogenated propenes occur via addition to the C=C double bond. Reaction with OH radicals is the major atmospheric sink for halogenated propenes. Infrared spectra, radiative efficiencies, and global warming potentials of the halogenated propenes are reported. The contribution of halogenated propenes to radiative forcing of climate change and to local air pollution is negligible. The results are discussed with respect to the atmospheric chemistry and environmental impact of halogenated alkenes.

Conclusions

The atmospheric lifetimes of these halogenated propenes are determined by reaction with OH radicals and range between 7 and 26 days. The lifetime together with the IR spectra allow us to estimate the GWP100 of these compounds ranging from 2 to 7. The products of the OH initiated oxidation of halogenated propenes have been identified and quantified. These products will have a negligible environmental impact. These new low-GWP compounds will help safeguard the ozone layer and climate system.

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P-6-043 Atmospheric chemistry and environmental impact of general anesthetic gases: Halothane, Isoflurane, Enflurane, Desflurane, and Sevoflurane

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The halogenated ethers, halothane ($CF_3CHClBr$), isoflurane ($CF_3CHClOCHF_2$), enflurane ($CHFClCF_2OCHF_2$), desflurane ($CF_3CHFOCHF_2$) and sevoflurane ($(CF_3)_2CHOCH_2F$), are inhalational anesthetics used in most medical settings worldwide for induction and maintenance of general anesthesia. Yearly global emissions of the anesthetic agents to the atmosphere are unknown, but based on consumption figures from a large US hospital (University of Michigan), combined global emissions of the three compounds are likely in the range of several ktons/year.

We have measured the infrared spectra of these anaesthetics and conducted the first calculations of their contribution to radiative forcing of climate change which recognize the important fact that radiative forcing is strongly dependent on the wavelength of the absorption features.

Atmospheric chemistry of anesthetics is well established. The atmospheric lifetimes of isoflurane, desflurane, and sevoflurane are estimated at 3.2, 14 and 1.1 years, respectively. The 100 year time horizon global warming potentials of halothane, enflurane, isoflurane, desflurane, and sevoflurane are 50, 680, 510, 2540, and 130, respectively. The atmospheric degradation products of desflurane, and sevoflurane are not of environmental concern. Assuming approx. 200 million anesthetic procedures worldwide on an annual basis: Climate impact of global emissions of anesthetics is equivalent to that from the emission of approximately 4.4 million tonnes of CO_2.

P-6-044 Atmospheric nutrient inputs to the western Pacific

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Conference Information