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IGAC News

Coordinating and fostering atmospheric chemistry research towards a sustainable world

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Carlo Carmagnola (left) and Patrick Wright (right) doing the first simultaneous measurement of snow specific surface area and albedo at Summit (Greenland). With additional measurements of soot in snow and atmosphere, this work hopes to quantify the role of soot in the atmosphere and in the snow on the energy budget of the surface and of the troposphere.

Photo: F. Domine

In Cooperation with IAMAS Commission on Atmospheric Chemistry and Global Pollution

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IGAC News

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Canadian Ozone Monitoring Network

Observations are a corner stone of global environmental change research. Long-term atmospheric measurements provide an essential view of change in the atmosphere. It was with some worry that IGAC joined the international condemnation of the reported closure of the well-known and respected Canadian ozone monitoring network (http://www.nature.com/news/2011/110912/full/477257a.html). We at IGAC recognize the value of the Canadian network for the important observations it provides for monitoring both stratospheric ozone depletion and the short-lived climate forcer tropospheric ozone across the arctic region. The network in Canada is one of the longest running having been active since 1966. Therefore, it provides one of the most important records to be able detect not only climate change but also the effects of global environmental change. The IGAC Co-Chairs and Executive Officer, on behalf of the IGAC Scientific Steering Committee (SSC), wrote a letter to The Honourable Peter Kent, Minister of the Environment in Canada, expressing our concerns about the potential damage to scientific knowledge that will result from the reduction or closure of the Canadian ozone monitoring network (http://igac.jisao.washington.edu/igacsresponse.php).

In a wider context this highlights a critical issue that when budgetary constraints are tight, monitoring can seem an easy target, especially in-situ monitoring. These records, especially the long-term records, are key components of quantifying climate and global environmental change across our planet and many cannot be replaced or substituted by, for example, satellite measurements. We at IGAC are committed to supporting and exploiting these measurements to fulfill our scientific goals. We do understand there may be a need to rationalize observations, but this must be done on a scientific basis, not from a management perspective. We ask that the community be given the opportunity to determine what the scientific impact of reducing network size or coverage would be for any measurement. If we are to respond to future challenges we need good long-term observations to build on.

Paul S. Monks, IGAC Co-Chair
University of Leicester, UK

IGAC welcomes two Student Assistants to the International Project Office

June Landenburger is a second year undergraduate student at the University of Washington. She is majoring in Biology with a focus in environmental science with a minor in marine biology. She one day hopes to have the opportunity to study and work with sharks. Growing up on Vashon Island in Washington State deeply rooted her love for the water and marine life. Having little experience in Atmospheric Chemistry, she is learning vast amounts and finding interesting connections between chemistry and biology. June joined the IGAC Core Project Office in May of 2011. She is excited to become a part of the atmospheric chemistry community and learn more about current climate issues.

Steven Brey is an undergraduate student studying Atmospheric Science and Applied Mathematics at the University of Washington. Steven first took an interest in the atmosphere when at the age of five he saw a tornado touch down a short distance from his home in Central Minnesota. While Steven currently lives in Seattle, a city that deprives him of any more close encounters with tornados, he is lucky to have the unique opportunity of studying orographic effects mountains like Rainier and the Cascade Range have on clouds and weather. Steven joined IGAC Core Project Office in May 2011. He is excited to work with an organization that facilitates discussions and communicates essential information on climate and air quality.

Launch of the new IGAC Mailing List

IGAC has new options for staying in touch with the community. We have recently launched a new email based mailing list that gives you control over just how much you hear from us. You can choose to receive a hard or digital copy of our newsletter, which is published three times per year. Or you can decide to keep in closer touch with the IGAC community by signing up to be notified of upcoming IGAC related conferences, workshops, and other grand gatherings!

If you are currently receiving IGAC notifications for workshop, conferences, etc., then make sure to click update subscription preferences in the lower portion of these mailings to choose your level of communication with the IGAC community.

If you have not been receiving email notifications from IGAC, then you are not signed up for our new email service and we only have a snail mail address for you. In this case, you need to join the new emailing list by going to http://eepurl.com/eu3U6.

If you have any questions about our new mailing service, or IGAC in general, please contact us at info@igacproject.org.

Submit articles to the next IGAC Newsletter

The next upcoming IGAC newsletter is now open for article submissions! Workshop Summaries, Science Features, Program News, and Editorials are all acceptable and desired. Science Features are to be submitted at a recommended length of approximately 1500 words with 1-2 images. All other submissions must be approximately 600 words and have a maximum of 1 image. Images must be high resolution in the format of a .png file. The deadline for submissions for the February Issue of the IGAC Newsletter is 15 January 2012. Any questions concerning content or formatting may be sent to info@igacproject.org.

The 3rd Workshop on Air-Ice Chemical Interactions (AICI)

Columbia University, New York, NY USA · 6-7 June 2011

V. Faye McNeill

Department of Chemical Engineering, Columbia University, New York, NY USA

The 3rd Workshop on Air-Ice Chemical Interactions (AICI) was held at Columbia University in New York City from 6-7 June 2011, followed by a one-day workshop on snow modeling on 8 June 2011. The meeting was organized by V. Faye McNeill of Columbia University, Thorsten-Bartels Rausch of the Paul Scherrer Institut, and Hans-Werner Jacobi of the Laboratory of Glaciology and Geophysical Environment (LGGE) in Grenoble, France.

Air-ice chemical interactions play key roles in several key phenomena in atmospheric chemistry, including stratospheric ozone depletion, the chemistry of cirrus clouds in the upper troposphere, air-snowpack exchange, and halogen activation in the polar boundary layer. The first AICI workshop was held in 2006 at LGGE. The product of this meeting was a set of review articles that comprise a special issue in Atmospheric Chemistry and Physics in 2007 (http://www.atmos-chem-phys.net/special_issue80.html). The second meeting was held in 2008 at the British Antarctic Survey in Cambridge, UK. The goal of the 2008 meeting was to provide an update on the state of the science, continue to foster open communication among modeling, field, and lab groups, and to promote the participation of young scientists.

This year's meeting provided a forum to bring together new insights from AICI studies, including work carried out as part of the Ocean-Air-Sea Ice-Snow project (OASIS), Halogens in the Troposphere (HitT, another IGAC activity), and the International Polar Year (IPY). Like the first AICI workshop in 2006, the goal of this year's meeting was to produce a set of review articles, which are currently in preparation. The articles will be featured in a joint special issue between Atmospheric Chemistry and Physics and Earth System Science Data entitled "New Perspectives on Air-Ice Chemical Interactions (AICI)" (http://www.earth-system-science-data.net/submission/scheduled_special_issues.html#1). The editors are V. Faye McNeill, Thorsten Bartels-Rausch, Eric Wolff, and Hans Pfeiffenberger. Rather than an update to the 2007 special issue, the articles are intended to be retrospective, but written from the newly update perspective of the current state of the science. The articles are organized by science topic and integrate lab, field, and modeling perspectives. Planned articles include:

- Halogen-Ice Interactions in the Polar Boundary Layer
- Influence of Snow and Ice Microstructure on AICI
- Organic Material in Environmental Ices: Sources, Chemistry, and Impacts
- Polar Measurements
- AICI and climate
- AICI and Persistent Organic Pollutants

The workshop offered ample opportunity for discussion, and challenges for the AICI community were identified. The cryosphere is particularly vulnerable to climate change; therefore emphasis will be placed on understanding AICI in a changing environment. There is a need going forward to strengthen connections between the AICI community and other related Earth System areas (for example, biology in the sea ice, snow pack, and oceans) encompassed by OASIS and SOLAS. Future emphasis will also be placed on advancing snow chemistry modeling with the goal of interfacing with Earth System models. The participants agreed that one of the best ways to move the state of the science forward is to continue promoting productive communication among field, lab, and modeling groups through workshops like this one.

Presentations from the 3rd Workshop on AICI can be found at

http://mcneill-lab.org/aici-2011-archive/.

Tara Kahan

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This newsletter's young scientist spotlight is on Tara Kahan who received an IGAC Young Scientist Travel Grant to attend the 3rd Workshop on Air-Ice Chemical Interactions (AICI). Tara is from Regina, Saskatchewan (Canada). She received her undergraduate degree at the University of Regina and her PhD at the University of Toronto. She is currently a postdoctoral fellow at the University of Colorado's Cooperative Institute for Research in Environmental Science (CIRES).

What is your current area of research, i.e. what topic really gets your motor running?

I'm currently measuring weak absorptions of atmospherically important molecules, and investigating the effects of water on their absorption spectra. I'm also still dabbling in heterogeneous chemistry: I'm using molecular dynamics simulations to investigate physical interactions of hydroxyl radicals with ice surfaces in order to better understand their reactivity there.

Where are you most at home? Is it in the lab, writing papers, during field research or in front of a crowd giving a talk on your research?

I enjoy playing in the lab and writing papers, but my favourite part of science is discussing results and ideas with other researchers.

With absolutely no context, simply making the decision on the fly, would you choose a volumetric flask or an Erlenmeyer? Please explain your choice.

I would choose the volumetric flask. I find their shape more aesthetically pleasing.

Was there an event, influential individual or childhood dream that lead you to become a scientist? If not, what led you to pursue a career in science?

When I was quite young (up to grade 3 I think), I wanted to be a mathematician when I grew up. Then we learned long division and I changed my mind. I enjoyed science (and especially chemistry) throughout high school, but never considered pursuing it until two years after graduating. I'd been working as a legal secretary, and was ready for a change. I ran into my high school calculus teacher who suggested I study math at university (I had been intending to enroll as an arts or humanities student). I started looking at math classes, which led to looking at physics classes, which led to looking at chemistry classes, and that was that.

How did you become a member of the IGAC community?

I went to my first IGAC conference in 2008 in Annecy, and haven't missed a conference since!

To you, what is the ultimate goal of science? Does this goal have anything to do with why you became a scientist?

To me the ultimate goal of science is to improve our understanding of how the universe works. I don't think I really knew what the goal of science was when I first started taking science classes at university, but as soon as I started my first research project I knew that this was what I wanted to do.

What is your favorite hobby?

I guess I have lots of hobbies. What I consider my favourite is pretty circumstantial though. It could be hiking, or reading, or several other things. I think eating would be up there too.

Where do chemical reactions occur in ice and snow?

Jonathan Abbatt[1] and Thorsten Bartels-Rausch[2]

[1]University of Toronto, Canada

[2]Paul Scherrer Institute, Switzerland

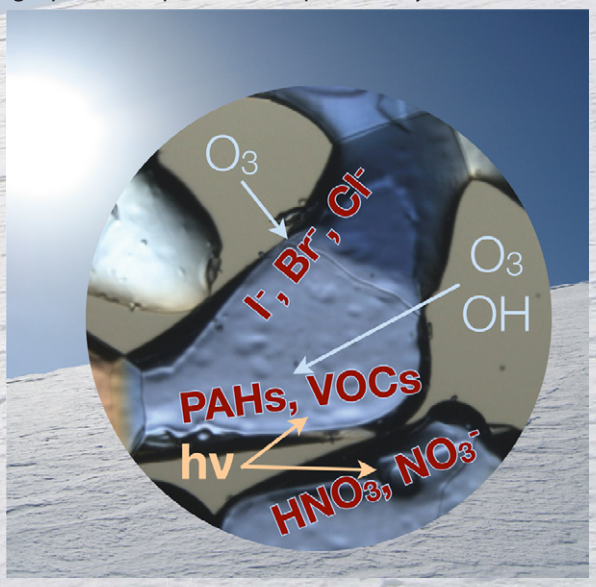
This question was the focus of much discussion at the recent IGAC-sponsored Air-Ice Chemical Interactions (AICI) workshop held at Columbia University. While of considerable interest from a purely academic perspective, the question is also of genuine relevance to understanding the nature of chemistry that occurs in the polar boundary layer, and probably mid-latitude snow chemistry as well. It is also relevant to reactions that occur with ice clouds throughout the atmosphere.

The subtlety of the question lies with the associated complexity of the composition, gross morphology, and surface structure of ice and snow substrates in the environment. As is well recognized, a pre-melting phenomenon gives rise to increased mobility and disorder of water molecules at the surface of ice, as the temperature approaches the melting point [*Hobbs*, 1974]. While not a true liquid, this surface layer a few molecule diameters thick is frequently referred to as the 'quasi-liquid layer (QLL)'. The QLL has been studied by a wide array of analytical techniques that differ over their characterization of its thickness and liquid-like nature, but all are in agreement that it is a persistent feature of pure ice surfaces. And, there is agreement that the QLL is important in driving some atmospheric reactions, such as chloride activation and SO_2 oxidation on ice clouds [*McNeill et al.*, 2006b; *Clegg and Abbatt*, 2001]. Other processes, such as the purely physical uptake of atmospheric trace gases, such as small organic species, to clean ice, seem to be perfectly well described as interactions with a solid surface, without invoking the QLL. But, in less pristine real-world environments such as the Artic snow pack, where considerable levels of adsorbed gases and deposited particles are present, what is the surface structure? Should it be described still as the QLL, or is a brine better suited to represent stable liquid solutions that may form when soluble species interact with ice?

While the difference may be purely semantic, and a continuum likely exists between the surface liquid layer and true liquid as solute concentrations increases, there is nevertheless direct relevance to the atmosphere. In particular, reactive processes may be accurately described by well-known, bulk aqueous-phase kinetics in a brine, whereas reactions occurring in/on the QLL will proceed at rates that remain poorly defined. What approach is most appropriate for snowpack photochemical models? In sea ice brine certainly is relevant to Artic boundary layer chemistry. This solid is percolated by concentrated brine channels that form when salts are excluded upon freezing. But it is not known whether the surface is better described as a brine or as ice. Similarly, liquid surfaces likely prevail on small, cm-scale frost–flowers that form on freezing seawater, where solid ice cores lead to the wicking upwards of concentrated salt solutions [*Roscoe et al.*, 2011].

Results were presented by a number of scientists, to illustrate the difference in reactivity between the surface and the bulk. In one study, the kinetics of bromide oxidation driven by exposure to gas phase ozone were presented, for both frozen solutions with close to seawater composition and their unfrozen counterparts [*Oldridge and Abbatt*, 2011]. This reaction is of considerable importance as a bromine activation mechanism that may be tied to springtime ozone depletion events. In both the frozen and liquid cases, the formation of gas-phase bromine occurred at rates consistent with both a bulk phase reaction, presumably within the brine, and a surface phase reaction occurring either on the surface of the ice or the brine itself. This is the first study to demonstrate quantitatively that, under atmospheric conditions, the surface phase reaction kinetically dominates.

Photochemical processes also occur at different rates, depending on their chemical environment. A clear example was given of work by a young scientist on the direct photolysis of aromatic compounds, such as anthracene (a three-ring polycyclic aromatic hydrocarbon, PAH) on ice [*Kahan et al.*, 2010]. It was found that this photolysis loss rate in the bulk phase, either frozen or liquid, was considerably slower than when the process occurs instead with high surface area substrates, such as small ice crystals, or at the interface itself. When a salt solution is frozen, these effects are not as pronounced, presumably because of the



Snow grains observed under polarized light in a microscope, with potential chemical processes. Thanks to Martin Schneebeli and Fabienne Riche for the photo (copyright by SLF, Davos).

brine that forms.

A higher level of chemical complexity, of relevance to sea ice chemistry, is the enhancement of the rates of reaction that occur upon solution freezing. In part this effect may arise from electric potentials that are known to arise across the liquid-ice interface that form in these situations [*Hobbs,* 1974]. However, the "freeze concentration" effect is also of potential importance, whereby freezing creates brine solutions that are orders of magnitude more concentrated than seawater. New results were presented that illustrate that halide oxidation can occur in such systems, associated with NO_x conversion chemistry. For example, when acidic solutions containing bromide, iodide and nitrate are frozen, oxidized halogens are formed [*O'Sullivan and Sodeau*, 2010]. The importance of this novel chemistry to the polar boundary layer is unclear but intriguing. Solute concentrations in natural brine should reach the same levels as in these laboratory experiments, so similar reactions might be expected.

One direction for the future is the application of new, state-of-the-art analytical techniques to better study the reactants under atmospherically relevant conditions. One example presented at the meeting was a new application of X-ray photoelectron spectroscopy (XPS) and near edge X-ray absorption fine structure (NEXAFS) spectroscopy to describe the chemical environment of small molecules at the ice surfaces. Normally these analytical techniques are restricted to high vacuum environments, i.e. not conducive to ice which has a sizeable vapor pressure, but technical advances now allow these experiments to be performed at synchrotron facilities. One result presented was for nitric acid, where it was shown that the adsorbed molecule is best represented as in the nitrate form [*Krepelova et al.*, 2010]. This is important because it is now known that photolysis of nitric acid within snow leads to the release of NO_x and OH. This likely occurs readily because the absorption cross section of nitrate is red-shifted into the actinic part of the spectrum relative to that of nitric acid. This study also revealed new insights to the restructuring of the ice surface due to the presence of impurities. Even though the nitrate was associated with water clusters just as in a solution, the measurements showed that there was still solid ice present at the surface. The presence of nitrate did not seem to have thickened or induced a complete surface disorder within the probing depth of a few tens of water molecule diameters. Certainly new experiments are needed to fully characterize the ice surface upon absorption of trace gases.

As mentioned above, the challenge in the community is to now connect these exciting, novel laboratory results to the atmosphere/snow-pack/seas-ice environment, through the use of appropriately parameterized models. The challenges are considerable given that the reactions are not elementary processes, i.e. do not proceed via a single step. Instead, for example, in the case of the freeze-concentration effects, the reactions undoubtedly proceed via many steps, with the rate of each dependent on temperature, solution composition, ionic strength, etc. Similarly, characterization of true surface-phase processes requires knowledge of surface layer partition coefficients and rate constants. To date, these quantities are only just being measured in the lab. In the past, snowpack models worked within a paradigm of liquid phase chemistry occurring within a thin aqueous solution layer existing on the top of ice. The next generation of models will have to encompass more of the true chemical and physical complexity described above to more accurately describe the processes.

For example, for one day after the AICI workshop, scientists from outside the atmospheric chemistry community presented ideas on how to model snow metamorphism [*Kaempfer and Plapp*, 2009]. Dynamics of the snow cover are currently not implemented in the chemistry snowpack models. By doing so changes in optical properties, air flow, and heat and mass transfer could be more precisely captured. These properties all influence the reactivity of the snowpack. Another line of modeling research tackles the question of the global importance of snow-atmosphere interactions. Here developments over the recent years to implement a detailed parameterization of snow chemistry in large-scale models were presented.

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The Atmospheric Chemistry and Climate Model Inter-comparison Project (ACCMIP)

Drew Shindell[1] and Jean-François Lamarque[2], co-chairs

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[2]NCAR, Atmospheric Chemistry Division, Boulder, CO USA

Introduction

The ACCMIP activity aims to better evaluate the role of atmospheric chemistry, both gases and aerosols, in driving climate change. In particular, the intercomparison is designed to facilitate analyses of the driving forces of climate change in the simulations being performed in the Climate Model Intercomparison Project phase 5 (CMIP5, see Figure 1, from http://cmip-pcmdi.llnl.gov/cmip5/docs/Taylor_CMIP5_design.pdf) in support of the IPCC AR5. ACCMIP consists of a set of experiments designed to provide insight into the CMIP5 simulations of historical and future climate change, along with additional simulations to better understand the role of particular processes and to constrain uncertainties. After a preliminary meeting held in Paris, France in June 2009, the first ACCMIP workshop took place 13-15 April 2011 in Toulouse, France and was hosted by Meteo-France.

Motivation for ACCMIP

The simulations performed for the Climate Model Intercomparison Project (CMIP) phase 3 activity in support of the IPCC AR4 provided a tremendously useful resource for exploring issues of climate sensitivity, historical climate and climate projections. However, the radiative forcings imposed in both the simulations of the 20th century and the future projections varied from model to model due to varying assumptions about emissions, differences in the behavior of physical processes affecting short-lived species that were included, and differences in which processes and constituents were included at all. For example, only 8 of 23 CMIP3 models included black carbon while less than half included future tropospheric ozone changes. Furthermore, the CMIP3 archive does not include diagnostics of radiative forcing from aerosols, ozone, or greenhouse gases other than carbon dioxide. Hence it is not straightforward to understand how much of the variation between simulated climates in the models results from internal climate sensitivity and how much results from differences in the forcings.

The CMIP5 project similarly will have a knowledge gap when it comes to atmospheric chemistry, with relatively little information on aerosols or on gases other than carbon dioxide. As models progress to a more Earth System approach including more interactions with the biosphere, a larger number of climate-sensitive emissions are also being incorporated into models, which will lead to diversity in the projected emissions even though anthropogenic emissions should be quite uniform. Hence there is a need for characterization of the forcings imposed in the CMIP5 historical and future simulations, and for diagnostics to allow us to understand the causes of the differences in forcings from model to model. There is also a need to better constrain uncertainties due to natural emissions, projections of anthropogenic emissions, etc.

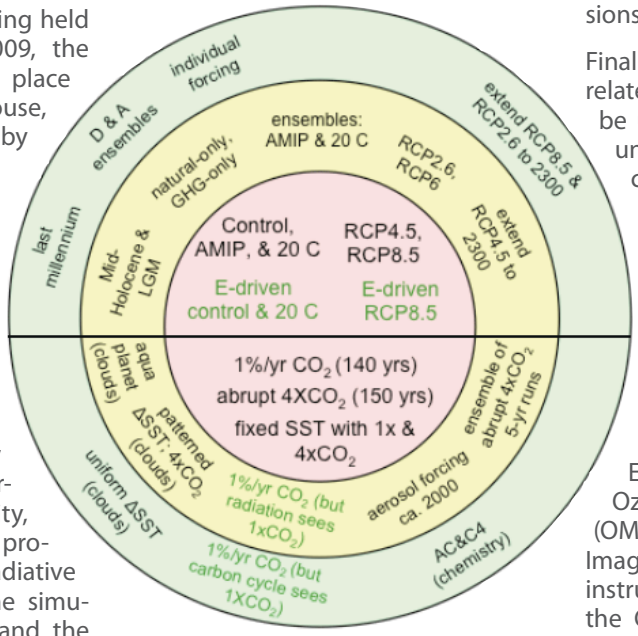

Figure 1. The design of the CMIP5 project, with the central area (pink) indicating the CMIP core simulations, the first ring (yellow) indicating Tier 1, and the outer ring (blue) shows Tier 2 simulations (including ACCMIP, then called AC&C4).

Finally, a wealth of new observations related to atmospheric chemistry can be used to evaluate and further our understanding of chemistry and climate. ACCMIP will take advantage of these measurements by performing extensive evaluations of the models, especially as regards their simulations of tropospheric ozone and aerosols, both of which have substantial climate forcing that varies widely in space and time. Sources such as the Tropospheric Emission Spectrometer (TES), Ozone Monitoring Instrument (OMI), and Moderate Resolution Imaging Spectroradiometer (MODIS) instruments on the Aura satellite, the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO), and the ground-based Aerosol Robotic Network (Aeronet) will be used, requiring the input of both the modeling and observational communities. The ACCMIP attempts to meet these various needs through

a set of coordinated simulations, diagnostics and evaluations.

Workshop outcomes

The workshop further defined the experimental setup of the ACCMIP projects currently underway, along with the delivery schedule and format, while focusing primarily on discussion of specific topics for analyses that would be performed on the ACCMIP dataset.

The ACCMIP Experiments

Complete descriptions of the experimental specifications and output protocols are maintained at the ACCMIP website (http://www.giss.nasa.gov/projects/accmip). Here we provide a brief overview and highlight updates from the workshop.

Emissions

Consistent gridded emissions dataset from 1850 to 2100 for modeling studies in support of CMIP5/IPCC AR5 have been recently created [*Lamarque et al.,* 2010a]. Emissions of gaseous and particulate species (i.e. aerosols, ozone and aerosol precursors) from anthropogenic activities and biomass burning have been estimated over the full period, using the 2000 dataset for harmonization of the 1850-2000 emissions with the future emissions determined by the Integrated Assessment Models (IAMs) for the four Representative Concentration Pathways (RCPs); all these scenarios are described in *van Vuuren et al.* [2011] and associated papers. These emissions are being used as boundary conditions for chemistry/aerosol model simulations in ACCMIP.

ACCMIP_1

The first set of ACCMIP simulations consists of timeslice simulations including detailed chemistry diagnostics to provide information on the forcings of historical and future climate change in the CMIP5 simulations (see Table). Each requested simulation is assigned a C (core, i.e. essential) or 1 (Tier 1, i.e. useful). Each run is 4-10 years with prescribed SSTs taken from CMIP5 runs. Note that output from experiments performed with coupled chemistry-climate-ocean models run in transient mode (e.g. CMIP5 runs) is also part of the protocol.

Additional runs for 2000 with 1850 climate and for 2030 and 2100 (RCP8.5) with 2000 emissions are designed to separate the effects of climate change on constituents and for isolating aerosol indirect effects (AIE). For other time periods and RCPs, these diagnostics will again be used to diagnose the AIE, but removal of climate-induced cloud feedbacks will be required based on the transient 1% per year CO_2 CMIP5 runs.

During the recent workshop, it was decided to request future simulations from the RCP6.0 scenario and to de-emphasize the RCP4.5 scenario. Indeed the RCP6.0 shows the largest divergence from the other scenarios for many short-lived species emissions. It was also decided to add timeslices at 2050 to the requested output as this time horizon is of interest to policy makers. The workshop also concluded that we would follow the suggestion of the Ozone Trends Group led by *J. Stähelin et al.* to determine which tropospheric ozone to use in model evaluation.

ACCMIP_2

The second set of ACCMIP simulations is designed to characterize the sensitivity to fully or partially natural emissions that will vary between models. These were initially planned to be performed for year 2000 conditions only, but the workshop participants decided to add similar experiments with 1850 and 2100 conditions to explore how sensitivities may vary with time.

The set of five simulations following discussion at the workshop, including substantial revision of the methane-related runs, is:

- 2.1: +100 Tg/yr isoprene (scale existing source to add 100 Tg/yr)
- 2.2: +20% biomass burning (all species).
- 2.3a: +8 ppb (mol/mol) or 50 Tg/yr methane (depending on if running with prescribed concentrations or emissions)
- 2.3b: 2100 RCP8.5 methane concentration (for chemistry only, not radiation, all else at 2000)
- 2.4: +2 TgN/yr lightning NO_x (scale existing lightning source to add 2 TgN/yr)

Historical Simulations

Emissions/Configuration	1850	1890	1910	1930	1950	1970	1980	1990	2000
Emissions and SSTs/GHGs for given year	C	1	1	C	1	1	C	1	C
Year 2000 emissions/1850 SSTs & GHGs									C

Future Simulations

Emissions/Configuration	2010	2030	2050	2100
RCP 2.6		C	1	C
RCP 4.5	1	1	1	1
RCP 6.0	C	C	1	C
RCP 8.5		C	1	C
Year 2000 emissions/RCP 8.5 SSTs & GHGs		C		C

C = core, 1 = Tier 1, blank = not requested

While these simulations emphasize emissions that affect gas-phase chemistry, ACCMIP strongly endorsed the AeroCom simulations with prescribed optical properties that will help characterize the sources of diversity in aerosol models.

Three additional sets of ACCMIP simulations have been proposed during the original ACCMIP meeting, but it was decided at the workshop to concentrate initially on these first two sets.

Output

The ACCMIP runs include output of concentration/mass of radiatively active species, aerosol optical properties, and radiative forcings (clear and all sky) as well as important parameters that do not directly influence climate such as the hydroxyl radical, ozone budget production and loss rates, specific chemical reaction rates, deposition rates, emission rates, high-frequency surface pollutants and diagnostics of tracer transport. CMOR tables have been created, based in part on fields archived for HTAP, AeroCom, and/or CCMVal. All data follow standardized formats and use CF-compliant names whenever available. The diagnostics include a stratospheric ozone tracer and a passive tracer of transport within the troposphere as defined in the HTAP project.

Current status

Data is currently being archived at the British Atmospheric Data Center, with a data access policy providing one year of access to participating groups only followed by general public access. To date, data from nine models has been deposited with the BADC, including the models used at CICERO (Norway), GFDL (USA), GISS (USA), LSCE (France), Meteo France (France), NCAR (USA), NIES (Japan), NIWA (New Zealand), and the UKMO (UK) (although not all models have completed the process).

Important dates regarding the ACCMIP activity include:

- Apr 13-15, 2011 — ACCMIP workshop, Toulouse
- Sep 1, 2011 — Submission of past and future core simulation (most groups)
- February 2012 — ACCMIP 2nd workshop (currently being planned)
- July 31, 2012 — Papers must be submitted by this date for use in IPCC AR5 WGI
- 2012 — Post-AR5 ACCMIP activities (ACCMIP_3, ACCMIP_4, etc)

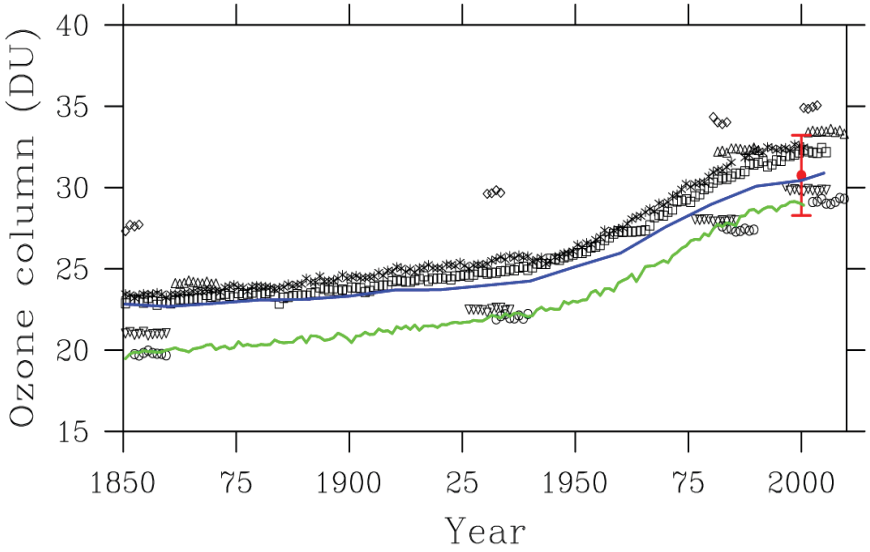

Figure 2. Time evolution of tropospheric ozone column (O_3 < 150 ppbv) from ACCMIP simulations (symbols), annual values (green) from *Lamarque et al.* [2010a], decadal values (blue) from *Kawase et al.* [2011], ACCENT/AR4 results (red). ACCMIP simulations are preliminary results.

ACCMIP Science

Several specific analyses were discussed at the workshop, with volunteers agreeing to lead the investigations in particular areas with the cooperation of other interested ACCMIP participants. The science topics and their leads are:

D. Stevenson (University of Edinburgh, School of GeoSciences): ***evaluation of ozone budget***

Tropospheric ozone is an important greenhouse gas and air pollutant, affecting human health and vegetation. Tropospheric ozone has increased substantially since preindustrial times (Figure 2). This increase is thought to be mainly due to increases in anthropogenic emissions of its precursors: methane, nitrogen oxides, carbon monoxide, and non-methane volatile organic compounds. Attribution of the increase to specific causes is complex, as the ozone budget has several inter-related components. In addition to chemical production from anthropogenic precursors, there are also significant natural sources of precursors, and ozone is also added to the troposphere from the overlying stratosphere. Ozone is removed by chemical destruction and dry deposition to the Earth's surface, particularly to vegetation. All of these budget terms are sensitive to global change. The ACCMIP model diagnostics allow the evolution of individual budget terms to be tracked in detail, to understand their influence on ozone concentrations. Understanding how the ozone budget and concentrations respond to changes in emissions and climate are necessary prerequi-

sites to the development of ozone control strategies.

W. Collins (United Kingdom Met Office): ***air quality and climate penalty***

The ACCMIP experiments cover many short-lived species responsible for ground level pollution, particularly ozone and fine particulate matter ($PM_{2.5}$). These can be harmful to people and to vegetation (including crops). The experiments cover both changes in emissions and in climate, thus enabling us to separate the impact of climate change on surface concentrations. We will analyze the changes in concentrations from the pre-industrial to the present and future, and will compare the present day results with observational datasets of surface pollution.

J.-F. Lamarque (NCAR, Global Tropospheric Modeling Group) and V. Eyring (DLR): ***comparisons with ice core observations and modern aircraft/satellite data (troposphere & stratosphere)***

Using the 2000 time slice experiments, we will perform an extensive evaluation of the model performance against standard tropospheric (Figure 3 for a comparison against ozone sondes) and, if applicable, stratospheric datasets (based on the CCMVal diagnostics). In particular, we will make use of the extensive satellite datasets (sampled as climatologies as the simulations do not capture interannual variability in emissions). Using the additional historical simulations, we will use ice-core measurements (e.g., sulfate and black carbon deposition, hydrogen peroxide, see *Lamarque et al.,* 2010a and b) to capture long-term changes in atmospheric composition. Finally, using the guidance from the Tropospheric Ozone Sondes group (see this issue), we will make use of the 1980 and 2000 time slices to identify if models are able to capture recent changes in tropospheric ozone.

K. Bowman (NASA, Jet Propulsion Laboratory): ***measurement/model comparison of ozone RF from TES***

The radiative forcing from tropospheric ozone (with respect to the tropopause, after stratospheric adjustment) estimated in the IPCC TAR (Third Assessment Report) [*Ramaswamy et al.,* 2001] was +0.35 ± 0.15 W/m^2 and did not change significantly in the IPCC AR4 due in part to continued uncertainty in preindustrial emissions and lack of global present day ozone observations. *Worden et al.* [2011] introduced global instantaneous radiative kernels, which represent the sensitivity of outgoing longwave radiative flux to the vertical structure of ozone, from the Tropospheric Emission Spectrometer (TES) - launched aboard the NASA Aura spacecraft in 2004. These kernels were applied to the comparison of four chemistry-climate models in Aghedo et al. [2011] with TES ozone profiles that showed vertically dependent biases in ozone led to regional biases of up to 0.7 W/m^2 for August 2006. This methodology will be applied to the ACCMIP chemistry - climate model simulations to evaluate ozone and ozone radiative effect bias over the TES epoch.

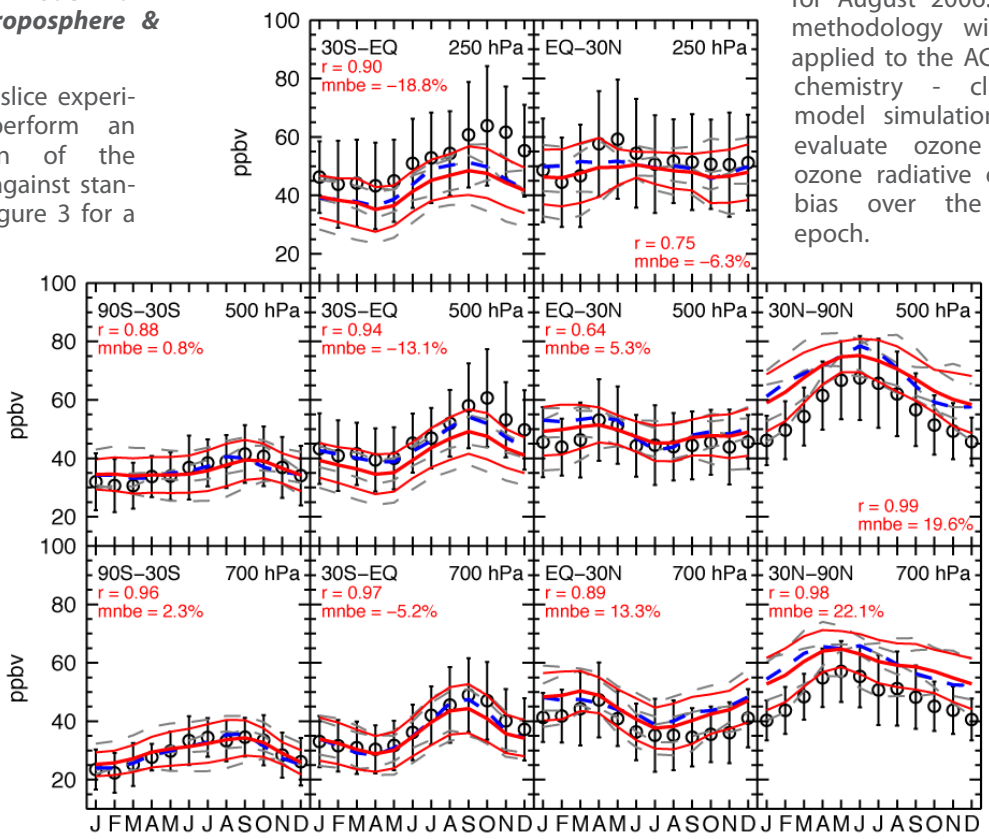

Figure 3. Comparison of model simulations (2000 time slice, red for multi-model mean, blue for multi-model median) against climatology of the ozone sondes in the given latitude bands. Error bars are the mean of the temporal standard deviations of the ozone sondes. Correlation coefficient and mean bias are also indicated for each region. Results are preliminary.

F. Dentener (Joint Research Centre): ***deposition of nitrogen and sulfur***

Deposition of nitrogen- and sulfur-containing compounds plays a prominent role in a host of environmental effects, including enhancement of carbon uptake by vegetation, eutrophication and acidification. Using the ACCMIP dataset we analyze the global and regional changes in deposition of these components from the pre-industrial to the present, and compare the model results with observational datasets.

M. Schulz (Norwegian Met Office): ***AeroCom-style evaluation of models against multiple datasets***

The ACCMIP dataset will create a new set of simulated of aerosol distributions based on different emission assumptions and for time slices across the 20th century and into the future. Comparison to aerosol observations is only possible for the period 1980-2010 based on very heterogeneous datasets of surface concentration, deposition and optical properties. Such evaluation has been used recently within the AeroCom initiative to analyze hindcast and present day simulations (http://aerocom.met.no) and is currently written up in several scientific papers. AeroCom tools and datasets will be made available and applied to the ACCMIP analysis. Providing a comparative analysis for the time slices around present day time periods would allow to establish model bias and a characterization of intensive properties of the aerosol in the ACCMIP model simulations. Doing this will make the ACCMIP future aerosol distributions comparable to other multi-model aerosol forcing estimates (AEROCOM, CMIP5). Ultimately it is hoped that the aerosol field differences between detailed offline chemical transport models and GCMs become better characterized. The realism of aerosol in fully interactive GCMs and thus the role of aerosol for the evolution of climate shall be explored.

P. Young (NOAA, Earth System Research Laboratory, Chemical Science Division): ***response to isoprene change***

Biogenic VOCs (BVOCs) are of interest due to their high emissions (~9 times that of anthropogenic VOCs), their generally high reactivity, and the sensitivity of their emissions to climate change (e.g. temperature, CO_2 level, changing distribution of plant species). While the atmospheric chemistry of BVOCs has consequences for both air quality (e.g. ozone, aerosols) and climate (e.g. methane lifetime, ozone), including BVOCs in a global model is complicated by the complexity of their chemistry, and the parameterization of the emission rate. The different models within ACCMIP have developed several strategies to deal with modeling the impact of BVOCs, and one of the main goals of the project will be to document these and analyze potential sources of differences. Analysis of the impacts of changing BVOC emissions will be closely coupled to analysis of changes in methane lifetime and OH, and the ozone budget.

A. Voulgarakis (NASA, Goddard Institute for Space Studies) and V. Naik (NOAA, Geophysical Fluid Dynamics Laboratory): ***methane lifetime***

The atmospheric concentration of methane (CH_4) has more than doubled since preindustrial times, which is of concern because of its impact on atmospheric chemistry and climate. The atmospheric lifetime of methane is controlled primarily by the hydroxyl radical (OH), the most important oxidant in the atmosphere. OH removes a wide range of pollutants and non-CO_2 greenhouse gases from the atmosphere, and is key to the formation of tropospheric ozone and aerosols. We analyze the results from ACCMIP models to understand how global and regional OH, and methane lifetime have evolved from preindustrial to present day and how they may change under future scenarios. A range of model diagnostics allow us to identify the key drivers of atmospheric OH and methane lifetime variability, including different emissions, water vapor, stratospheric ozone, clouds, aerosols and surface albedo.

D. Shindell (NASA, Goddard Institute for Space Studies): ***evaluation of radiative forcing and of climate response***

The ACCMIP dataset includes the full geographic distribution of radiative forcing by each gas and aerosol component, as well as the total aerosol indirect effect. Our first effort will be simply to characterize the various forcings and explore the reasons for diversity in those cases where it is greatest. We also intend to compare the spatial distribution of forcing with the climate response realized in the companions CMIP5 simulations. CMIP5 includes simulations driven solely by increasing CO_2, which will allow us to determine how much of the spatial pattern of climate response is due to variations in climate sensitivity in different places. We hope that that geographic pattern of climate sensitivity can then be removed from the full climate response to historical and/or future forcings to diagnose the regional climate impact of the inhomogeneous forcings from ozone (Figure 3) and aerosols. Results will be compared with CMIP5 'individual forcing' experiments that will be performed by at least some groups.

Conclusion

The simulations performed for ACCMIP are intended to provide a better understanding of both the factors driving projected climate change in the CMIP5/AR5 simulations and of

the strengths and weaknesses of the current generation of chemistry-climate models and/or their boundary conditions. This represents a community-driven effort to provide more insight into the overall chemistry component of the climate models whose output is used in major policy decisions, and is complementary to evaluations focused on specific portions of the chemical system (AeroCom on aerosols, CCMVal on the stratosphere, HTAP on long-range transport in the troposphere).

The structures built for ACCMIP have been designed to follow the conventions used in the climate modeling community as much as possible. This should greatly facilitate comparisons between the ACCMIP models and CMIP5 models, as well as between ACCMIP models and the many datasets that are being used for evaluation of CMIP5 models. We are also optimistic that the many tools developed for the ACCMIP activity, including the CMOR tables, the archive structure, and analyses codes, can provide many years of continued support of chemistry-climate model intercomparison and evaluation against observations. By aligning the activity with CMIP5, we also hope the ACCMIP infrastructure can help serve as a bridge between the climate-centered CMIP and the chemistry modeling community as the community continues to move towards more fully interactive Earth System Models.

ACCMIP is organized under the auspices of Atmospheric Chemistry and Climate (AC&C), a project of International Global Atmospheric Chemistry (IGAC) and Stratospheric Processes And their Role in Climate (SPARC) under the International Geosphere-Biosphere Project (IGBP) and World Climate Research Program (WCRP).

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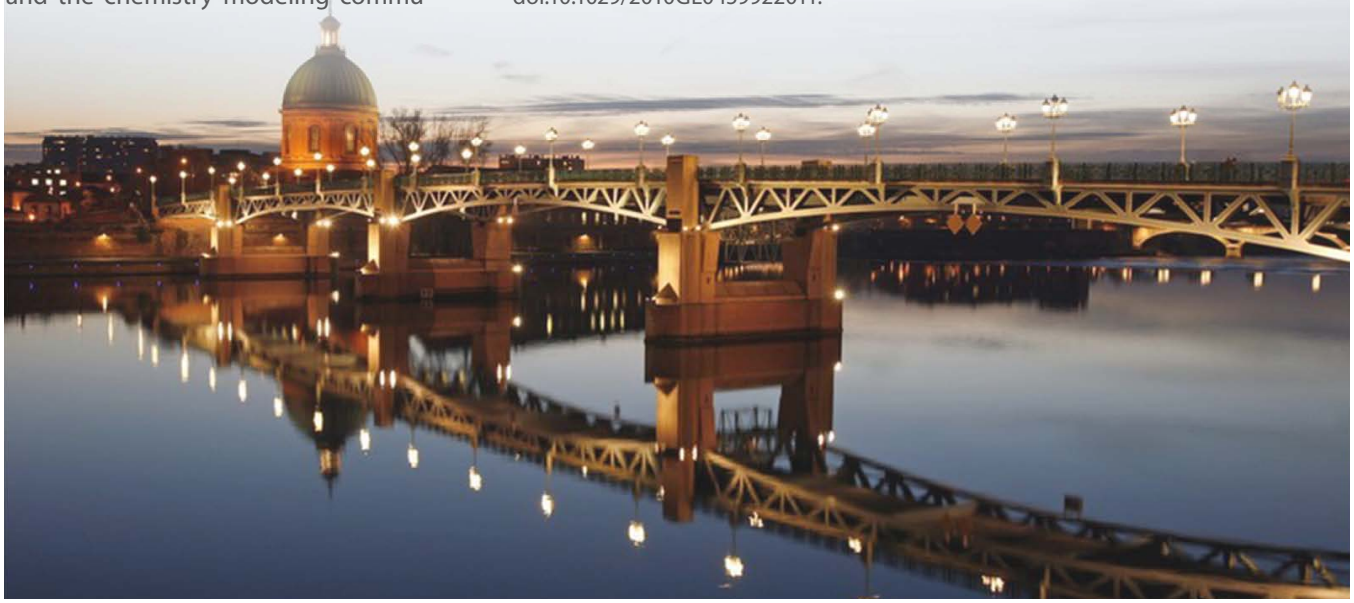

Workshop on Tackling the Air Pollution and Climate Change Challenge

Arona, Italy • 9-10 June 2011

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As part of the International Geosphere - Biosphere Programme's (IGBP) Air Pollution & Climate Initiative, a two-day workshop was held in Arona, Italy on 9-10 June 2011 to discuss the development of an effective science-policy dialogue to address the Air Pollution and Climate Change Challenge.

The workshop had 22 participants across the science-policy spectrum representing 13 different countries. Participants were given the opportunity to present their perspective on the Air Pollution and Climate Change Challenge. Perspectives were varied but the general consensus was there is still a separation between air pollution and climate change in both the policy and scientific communities. This separation is reflected in the temporal and geographic scales of interest: with air pollution efforts focused on the near-term and the local and regional scales, whereas climate change efforts are focused on the long-term and global scale. As with many issues, there also exists a divide between the scientific and policy communities that hinders communication and understanding. The aim of the Air Pollution and Climate Initiative is to break down these divides (Figure 1) and clarify the synergies and trade-offs of research and mitigation efforts across a spectrum of air pollution and climate change policies (Figure 2). The Air Pollution & Climate Initiative seeks to build upon current efforts tackling these issues and to provide continuity between present and future efforts. Current efforts include the United Nation Environmental Program (UNEP) Integrated Assessment of Black Carbon and Tropospheric Ozone, the UNEP Atmospheric Brown Clouds (ABC) Impact Assessment Report, the Arctic Monitoring and

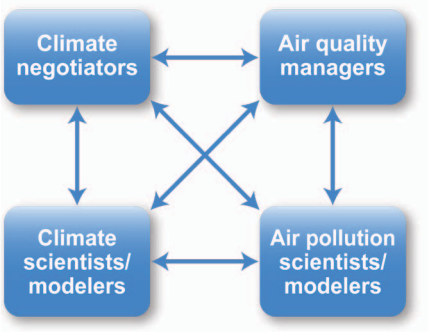

Figure 1. Linkages needed to facilitate simultaneous efforts to address air pollution and climate change in both the policy and scientific communities. Without these linkages opportunities for co-benefits or unintended negative consequences may be overlooked.

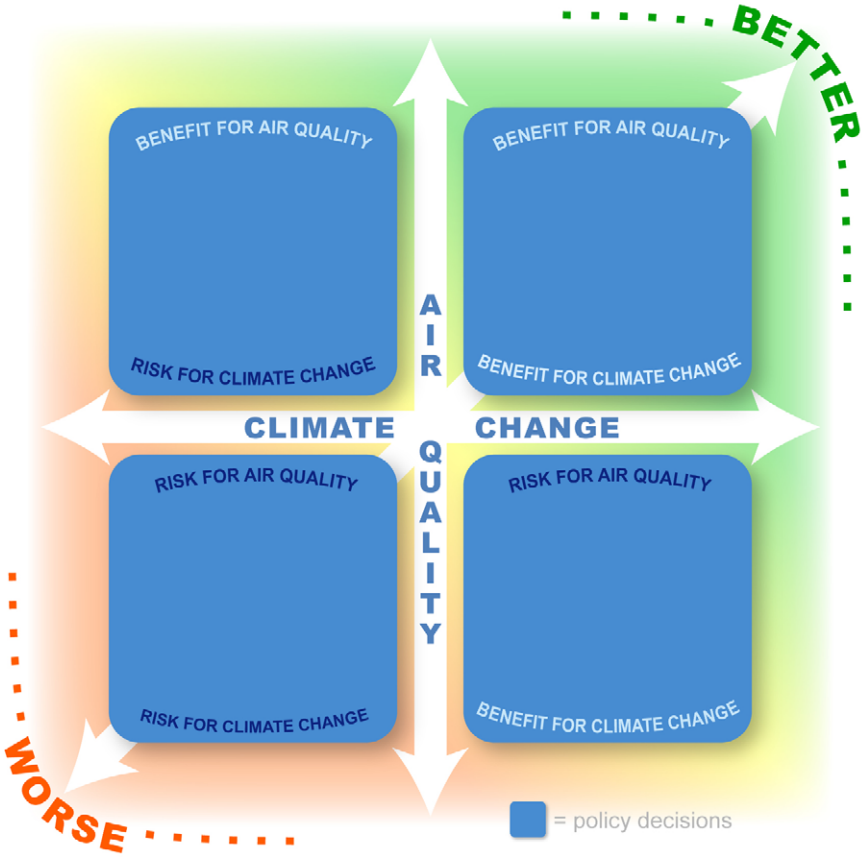

Figure 2. Schematic of the synergies and trade-offs of air pollution and climate change policy decisions.

Assessment Programme (AMAP) report on The Impacts of Black Carbon on the Arctic Climate, the International Global Atmospheric Chemistry (IGAC) and Stratospheric Processes And their Role in Climate (SPARC) Atmospheric Chemistry and Climate (AC&C) Activity, the US Environmental Protection Agency (EPA) Black Carbon Report to Congress, the EU Atmospheric Composition Change the European NeTwork Plus (ACCENT Plus), and the Long Range Transboundary Air Pollution (LRTAP) and European Monitoring and Evaluation Programme (EMEP) Task Force on Hemispheric Transport of Air Pollution (HTAP). By building upon these current efforts, the Air Pollution & Climate Initiative frames the Air Pollution and Climate Change Challenge as a problem comprising one atmosphere, same pollutants, and multiple effects.

Over the next two years, the Air Pollution & Climate Initiative will produce two documents:

1. IGBP Statement on the Air Pollution and Climate Change Opportunity
2. Strategic Plan for a Multi-Disciplinary Program on Air Pollution & Climate Change

The IGBP Statement on the Air Pollution and Climate Change Opportunity will provide a concise assessment of the benefits and risks associated with mitigating air pollutants for human health, agriculture, ecosystems, and climate. The statement will be released as a briefing document at the ICSU Planet Under Pressure Conference March 2012 in London.

At the same time the Air Pollution & Climate Initiative will develop and publish a strategic plan for a multi-disciplinary program on Air Pollution and Climate Change that will engage the international earth system science, social science, and policy communities. This will build on and take account of other international efforts coupling air quality and climate research such as the ICSU-Belmont Earth System Visioning process and provide specific recommendations and methodologies for creating and sustaining such a multi-disciplinary international program.

A follow up workshop on the IGBP Air Pollution & Climate Initiative is scheduled to take place 7-10 November 2011 in Taipei, Taiwan. This workshop will focus on Air Pollution & Climate: A Science-Policy Dialogue in Asia. The Taiwan Environmental Protection Agency (EPA) is sponsoring the workshop.

For more information visit http://www.igbp.net/4.1b8ae20512db692f2a6800018410.html

or contact megan@igacproject.org.

Methane Mitigation – Benefits for air quality, health, crop yields, and climate

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Air pollution contributes to climate change and climate change will likely exacerbate air pollution in some regions of the world, even if emissions of reactive air pollutants remain constant. As a result, there is an increasing dialogue between the scientific and regulatory communities to coordinate efforts to reduce emissions of reactive air pollutants, greenhouse gases and fine particulates and their precursors so that controls are beneficial for both air quality and climate. The newly launched IGBP Air Pollution & Climate Initiative is intended to facilitate such discussions and coordination.

Mitigation of methane (CH_4) emissions provides an opportunity to simultaneously improve air quality and reduce the rate of climate change. In addition, CH_4 is the primary constituent of natural gas and an important energy source. As a result, efforts to prevent emissions or capture and use CH4 offer significant environmental, energy and economic benefits [*USEPA*, 2006].

At approximately 1.8 ppm, CH_4 is the most abundant non-carbon dioxide (CO_2) greenhouse gas (GHG) in the atmosphere today [*Montzka et al.*, 2011]. CH_4 accounts for approximately 15% of current radiative forcing from GHGs in the atmosphere and comprises 63 percent of annual CO_2eq (equivalent CO_2 emissions calculated using a 100-year time horizon global warming potential, GWP_{100}) emissions of non-CO_2 GHG [*WWS*, 2011]. Methane is also a precursor of tropospheric ozone (O_3) and contributes to the growing global background concentrations of tropospheric O_3, itself a GHG and air pollutant with detrimental impacts on human health and vegetation. A strong positive feedback on radiative forcing (RF) through atmospheric chemistry is found following increased emissions of methane [*Isaksen et al.*, 2011]. This occurs because methane is a GHG, the O_3 it produces is a GHG, and increased CH_4 concentrations depress concentrations of the hydroxyl radical (OH), the primary sink of methane, which thus increases the lifetime of methane. In addition, methane oxidation produces CO_2 and leads to increased stratospheric water vapor, which contributes to destruction of stratospheric O_3 and to surface warming [*Shindell*, 2001].

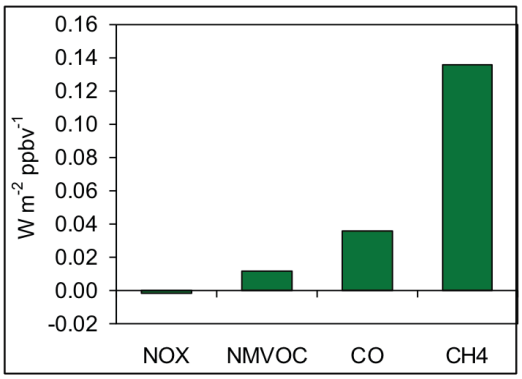

Figure 1. A 20% global reduction in anthropogenic emissions of NO_x, NMVOC, CO and CH_4 results in varying radiative forcing decreases per unit decrease in global surface O_3 concentration ($Wm^{-2}ppbv^{-1}$). Methane emission reductions result in the largest decrease in RF per unit decrease in surface O3 concentration of any O3 precursor. Results are from global model calculations discussed in *West et al.* (2007) as presented in *Jacob et al.* (2011).

O_3 is produced via the catalytic reaction of nitrogen oxides (NO_x=NO+NO_2) with non-methane volatile organic compounds (NMVOCs), carbon monoxide (CO) or CH_4 in the presence of sunlight. The effect of O_3 precursor emission reductions on RF per unit reduction of surface O_3 concentrations vary. Shown in Figure 1 is the calculated decrease in RF per unit (part per billion by volume, ppbv) decrease in global surface O_3 concentrations resulting from a 20% global decrease in anthropogenic emissions of each of the key O_3 precursors: NO_x, NMVOC, CO and CH_4. Of all O_3 precursors, CH_4 emission reductions result in the largest decrease in RF per unit reduction in surface O_3 [*West et al.*, 2007]. Thus, of all O_3 abatement strategies, methane controls reduce the rate of climate warming most.

Model simulations indicate that had global anthropogenic methane emissions been reduced by 20% beginning in 2010 the average daily maximum 8-h surface ozone would decrease by approximately 1 ppbv globally [*West et al.*, 2006]. By using epidemiologic ozone mortality relationships, this ozone reduction was projected to prevent approximately 30,000 premature all-cause mortalities globally in 2030, and 370,000 between 2010 and 2030 [*West et al.*, 2006].

Increasing evidence points to elevated O_3 concentrations as an important and usually overlooked stress on global crop yields [*Avnery et al.*, 2011a; *Van Dingenen et al.*, 2009; *Wang and Mauzerall*, 2004]. Recent model simulations quantified the present and potential future (year 2030) impact of surface O_3 on the global yields of soybean, maize, rice and wheat given both upper- and lower-boundary projections of reactive O_3 precursor emissions [*Avnery et al.*, 2011a; b; *Van Dingenen et al.*, 2009]. *Van Dingenen et al.*, 2009; and *Avnery et al.*, 2011b projected substantial future yield losses globally

for these crops: 10-16% for soybean, 3-6% for maize, 4-6% for rice, and 4-18% for wheat, even under scenarios of stringent O_3 controls via traditional pollution mitigation measures (i.e. reductions in NO_x, CO, and NMVOCs). In addition to reductions in short-lived O_3 precursors, further calculations indicate that mitigation of surface O_3 through gradual reductions in methane emissions between 2006 and 2030 could increase global production of soybean, maize and wheat by 23-102 Mt in 2030 – the equivalent of a ~2-8% increase over year 2000 production of these crops, worth US$3.5-15 billion worldwide (USD_{2000}) [*Avnery et al.,* submitted 2011].

With a lifetime of about a decade and a GWP_{100} of over 20, methane mitigation provides an opportunity to slow the acceleration of climate change. Because neither the air quality nor climate benefits of CH^4 mitigation depend strongly on the location of the CH_4 emission reductions, the lowest cost emission controls can be targeted [*Fiore et al.*, 2008]. Large potential for methane emission reductions exists, including the recovery of methane from coal, oil and gas extraction and transport, methane capture in waste management, and modifications of some rice cultivation and livestock management practices [*UNEP/WMO*, 2011]. Widespread implementation is achievable with existing technology but requires significant strategic investment and institutional arrangements [*UNEP/WMO*, 2011]. Many measures achieve cost savings over time, however initial capital investments are necessary in some cases. Figure 2 provides a cost curve for various methane mitigation options and indicates that at least 10% of projected 2030 methane

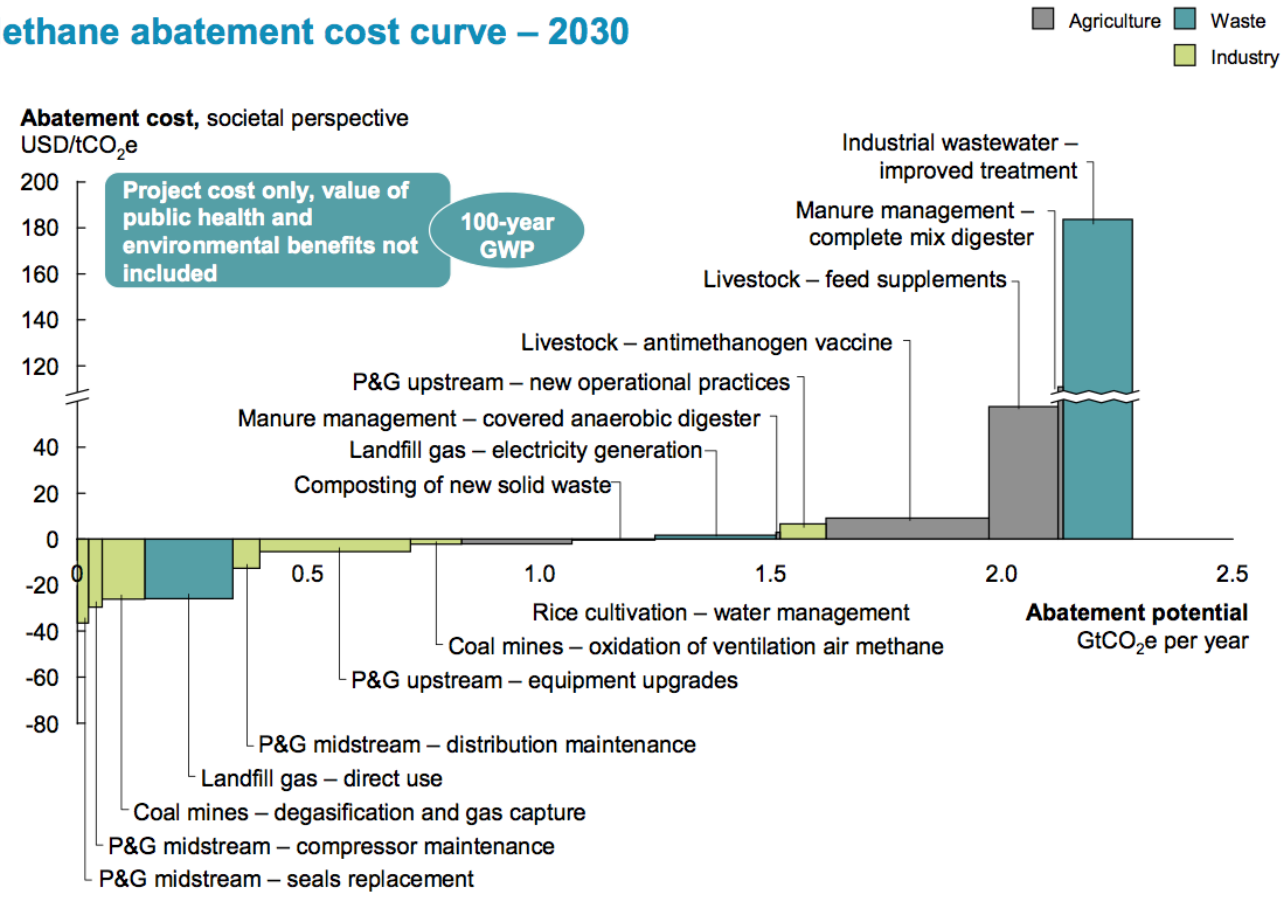

Figure 2. Global methane abatement cost curve. According to these estimates, methane mitigation of over 1.0 Gt $CO2_{eq}$ (approximately 10% of business-as-usual CH_4 emissions in 2030) can be achieved at a net cost savings. P&G = Petroleum and Gas [*ClimateWorks,* 2011].

emissions can be eliminated at a net cost saving [*ClimateWorks,* 2011].

Given the challenges of successfully implementing these mitigation strategies globally, further research which spans the scientific and stakeholder communities is needed to optimize near-term mitigation strategies in countries around the world and to evaluate the cost-benefit ratio for individual measures. This is an area where the newly launched IGBP Air Pollution & Climate Initiative, whose members span the scientific and stakeholder communities and include representatives from developed and developing countries, will have an opportunity to facilitate the implementation of cost-effective methane mitigation strategies which benefit air quality, human health, agricultural yields and climate.

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Atmospheric Brown Clouds: An Integrated Approach for understanding and managing climate change and other environmental problems resulting from atmospheric pollution

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Environmental issues such as climate change and air pollution are acknowledged facts. Also accepted as, highly likely, is the prediction that the observed warming trend of the last few decades will continue unabated during this century. It is also now accepted worldwide that human activities are the major source of the observed and projected climate changes. We have now entered an era in which sufferings of humans and all other animals and unprecedented stresses on eco-systems provide a powerful motivation for climate change research, which will be needed to provide a deeper understanding of the driving factors and the climate system responses. Research in atmospheric sciences take a central role since atmospheric pollution is the dominant source of manmade climate changes and the impacts on health, agriculture and water. Given its long history, the atmospheric science community should also focus directly on the human dimensions and applications aspects of environmental issues such as impacts analysis and decision making to address the adverse impacts. Towards this growing societal need, the world community of atmospheric scientists should take an overarching view of atmospheric problems, fill in major gaps in the application of the science, and develop practical solutions for challenges and problems that confront humans and ecosystems. This is the guiding vision of project Atmospheric Brown Cloud (ABC). While the objective of most air pollution institutions and organizations has been on the local and regional air quality, ABC's objective is to add value to this vast body of knowledge, by addressing the role of air pollution in climate change.

Project ABC was initiated by United Nations Environment Programme (UNEP) in response to the findings of the Indian Ocean Experiment (INDOEX), which documented exhaustively for the first time the trans-continental and trans-oceanic Atmospheric Brown Clouds (ABCs) over continental South Asia and the Indian Ocean in 1999 [*UNEP-ABC*, 2002 report]. ABCs include the manmade aerosols that are included in reports by the Intergovernmental Panel on Climate Change (IPCC). It also includes air pollutants such as tropospheric ozone. The major focus of the ABC project thus far has been on soot. Soot results from the incomplete combustion of fuels and consists of nano- to a few micro-metre (millionth of a metre) size particles. Black carbon and many organic aerosols are the main constituents of soot. The brownish colour of ABCs is mainly due to the absorption and scattering of solar radiation by black and organic carbon (BC), fly ash, and nitrogen dioxide gas. Typical background concentrations of aerosols are in the range 100 - 300 cm^{-3}, whereas in polluted continental and some marine regions the concentrations are in the range 1,000 – 10,000 cm^{-3}.

UNEP, in collaboration with INDOEX scientists established Project ABC to address the emerging issue of ABCs in 2001. The first project to start is ABC_Asia that includes scientists from China, Germany, Japan, India, Italy, Republic of Korea, Maldives, USA and Sweden. Plans have now been finalized to start ABC_Africa and ABC_Latin America and include scientists from these two continents. Over the last 10 years, the project has made significant progress on the enhancement of science, capacity, and awareness. The project results have greatly contributed to the elevation of short-lived climate forces high on the policy agenda at the national, regional, and global levels. Major contributions of the research conducted under ABC studies are summarized below:

> *The main finding as documented in a first of its kind regional assessment report [UNEP, 2008], is that ABCs and their interaction with the build-up of greenhouse gases have significant impacts on regional climate systems including the monsoon and the Himalayan glaciers, water budgets, agricultural production, and human health. The deaths attributed to ABCs (indoors as well as outdoors) are of the order of 2 million annually.*

The other principal outcomes from ABC are:

- **Increased capacity to study ABCs in developing countries:** Establishment of an integrated network of 12 strategically located ABC surface climate observatories throughout the Indo-Asia-Pacific region, supported by 2 super site observatories in the Maldives and the Cheju Island in S. Korea – a major achievement in this relatively poorly monitored part of the world – that are now operated by national scientists with regular training through the project and support from Science Team. ABC observatories include the first aerosol observatory in the

Indian Ocean with a 6-year time series data on the seasonal cycle and inter-annual variability in transport, aerosol chemistry and forcing, and the highest ABC observatory in the Himalayas documenting high soot levels at elevations as high as 5 km a.s.l. Observatories have enhanced the environment monitoring in developing countries, and they are providing useful information to assess the impacts of ABCs / air pollutants on climate change, water security, and food security in developing countries in Asia-Pacific. Observatory programme is being expanded to Africa and Latin America.

- **Identification of regional hotspots:** Spatial distribution of ABCs and regional ABC hotspots around the world have been identified (Figure 1). The aerosol regional radiative forcing to climate system and associated impacts are significant, often higher than the global values, due to heterogeneous spatial and temporal distributions of aerosol loading. The particularly affected regions with high values of aerosol burden caused by human activities are Asia (e.g., South Asia, East Asia, Indonesian region), Africa (Southern and Central Africa), and South America (Amazon basin) and the oceanic region downwind. These are the regional hotspots, and the ABC problem is not restricted to these regions only.
- Increased understanding on the impacts of ABC in Asia: A better understanding of the science of ABC with new findings on regional climate change through the studies by ABC scientists; such as
  - Fossil fuel combustion, biofuel cooking and biomass burning are the sources of ABCs.
  - Globally, BC in ABCs has a net warming effect on the climate system. The magnitude of its current warming effect is subject to uncertainty, ranging from about 25% to as much as 60% of the warming effect of CO_2 increase.
  - BC in ABCs adds significant solar heating to the atmosphere and causes large dimming at the surface.
  - While the earlier studies under ABC focused on South Asia, the East-Asian team of ABC have documented even large effects of ABCs on regional radiative forcing.
  - The world's highest ABC observatory is near the base camp of Mt. Everest and has recorded high BC concentrations of about 1000 ngm^{-3}, similar to values found in polluted regions.
  - ABCs (i.e., BC and other man-made particles) lead to large dimming at the surface and the global average effect of this is to decrease rainfall.
  - The large solar heating of the air by ABCs have been documented by aircraft data and has been shown (using models) to contribute to warming of the elevated

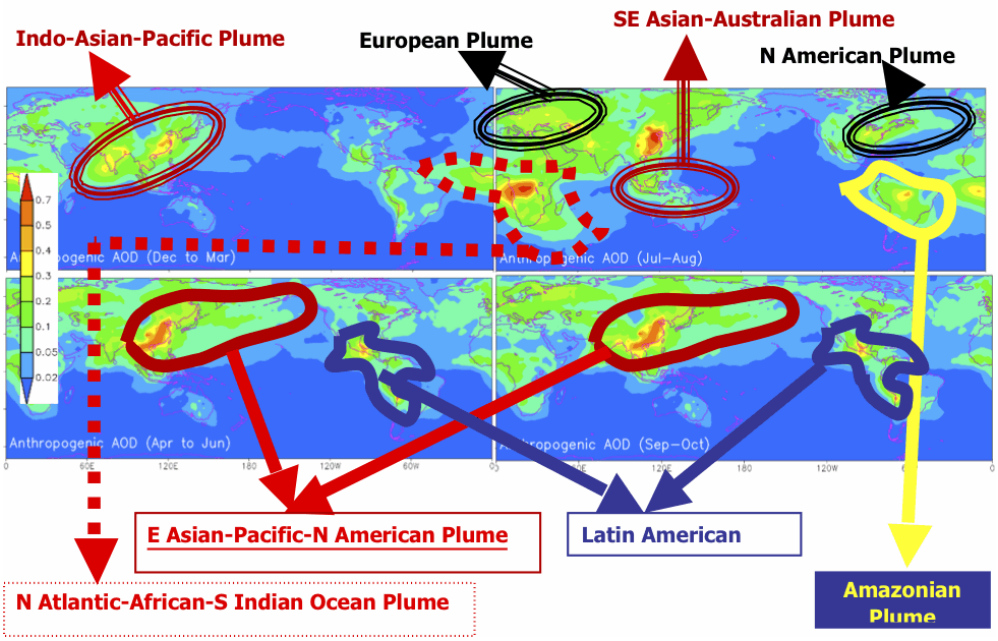

Figure 1. ABC hotspots [*V. Ramanathan et al.,* 2008].

regions of Himalayas and Tibet and thus amplify the CO_2 induced melting of glaciers.

- Deposition of BC on sea ice and snow darkens the surface and leads to more solar absorption and melting of sea ice and snow. This effect also contributes to melting of snow packs and ice in the Mountain glaciers, for instance Himalayas.
- Innovative isotope studies have provided convincing evidence for the major role of biomass combustion in the regional concentrations of BC over South Asia.
- Life time of BC is of the order of few days to few weeks. Hence, benefits of BC emission reduction can be seen within weeks of emission reduction.
- Reducing ABCs emissions will result in significant co-benefits on human health, water security, and food security.

- **Knowledge concerning mitigation measures developed:** Because ABCs are short-lived, atmospheric concentrations, and in turn ABC's effects, would be reduced shortly after emissions are reduced. Adding to the near-term promise of addressing ABCs is the fact that cost-effective technologies are presently available, which are being used in many parts of world, to reduce emissions from many key sources. The analyses show that available mitigation measures for residential sector has been identified as the most potential area to introduce interventions due to multiple co-benefits, technical feasibility and social and political acceptances. In the residential sector, solid biomass plays a key role in the energy mix of developing countries. Project Surya, mitigation component of project ABC, has been initiated to develop the knowledge concerning mitigation measures for solid biomass uses in the residential sector. Project Surya (www.projectsurya.org) is an internationally recognized cook stove project sponsored by the UNEP. The goal of Project Surya is to scientifically demonstrate the environmental and health benefits of the introduction of clean cooking technologies, with the ultimate goal of providing a rigorous evidence base for large-scale action in this area. Project Surya aims to deploy improved cooking technologies in a contiguous region with a population of approximately 50,000. The resulting "black carbon hole" that will be created in the otherwise omnipresent pollution cloud will be measured across space and time to quantify the multi-sector impacts of better cooking technologies. Project Surya will use cell phones, instrument towers, and satellites, and will empower village youth to work with world-class experts in documenting the impacts. A pilot phase was successfully completed in 2010 in a village in North India, one of the poorest and most polluted regions in the Indo-Gangetic plains. This pilot phase has already achieved a number of ambitious and measurable outcomes including documentation the connection between indoor air pollution from cooking and ambient outdoor pollution levels; identification of improved cooking technologies that reduce pollution significantly; deployment of improved cook stoves in all households in the pilot village (about 500); and verification that we will be able to measure the impacts of a larger-scale intervention. In addition, a parallel pilot test has been started in Nairobi, Kenya. Our recent data has additionally shown that the measured black carbon emissions are three to five times higher than the climate models predicted, making it all the more urgent to take action now to target black carbon and other short lived climate forcers.

Studies conducted by ABC scientists and other studies conclude that particles (or aerosols) and other pollutants (Ozone, CO, NO_x) in ABCs have major adverse impacts on human health, water security, food security, and climate at the global scale as well as at regional to local scales. On the global scale, ABCs may have masked as much as 50% of the global warming due to greenhouse gases. Thus disconnected and isolated policy responses to global warming and to health/eco-system effects of indoor and outdoor air pollution will have unintended consequences for the climate and possibly trigger non-linear changes. An example of an isolated policy response is the reduction of sulphur-dioxide emissions (to mitigate health impacts of air pollution) without a reduction in climate warming pollutants, which can lead to a large increase in the warming during the coming decades. On the regional scales, however, the ABCs may intensify effects of global warming on glacier retreat by soot deposition on snow and ice; and soot induced solar heating of the atmosphere, may led to major droughts in Africa and Asia by asymmetric alterations of sea surface temperatures and land surface temperatures, and have been shown to suppress formation of rain clouds. Finally, on local scale, inhalation of particles (indoors and outdoors) in ABCs has been linked with over a million deaths annually worldwide.

Clearly, what is needed is a common framework for addressing the combined impacts of due to greenhouse gases and air pollution. The need for an integrated approach was recom-

mended by the project ABC for the first time in 2002 (Figure 2). With the recent scientific developments, the need for an integrated approach is constantly being renewed. Now, more than ever, there is a need to help policy makers and decision makers formulate an effective integrated approach for atmospheric issues in the context of sustainable development.

References

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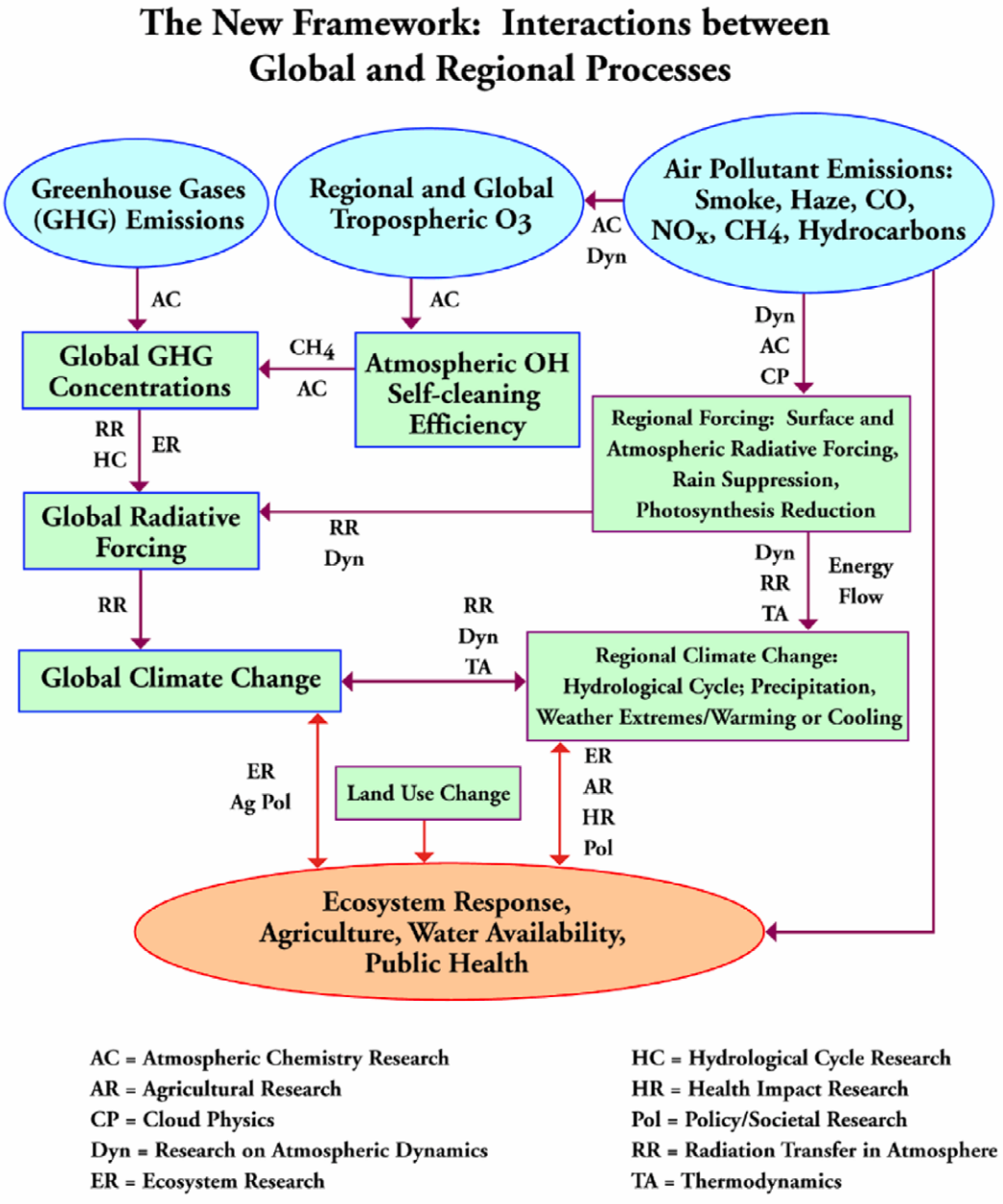

Figure 2. The New Framework: Interactions between Global and Regional Processes [*UNEP and C[4]*, 2002].

International workshop on biogenic volatile organic compound emissions models and their applications

Lancaster Environment Centre, Lancaster University, UK May 17th-18th 2011

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A two-day workshop was held at Lancaster University, UK, on 17-18 May 2011 to discuss biogenic volatile organic compound emissions models and their applications.

Over 60 attendees from 16 different countries heard presentations on the atmospheric chemistry of biogenic volatile organic compounds, the models used to estimate the emission rates of these compounds and their evaluation against measurements, and the application of emissions models for air quality and climate modelling. Discussions focused on the need to reduce the uncertainty in estimates of bVOC emissions and how this could be achieved.

The main points agreed in the discussion sessions were:

1. More inter-comparisons are required of both the components of the models and the full emissions models themselves. The results of these should be used to elucidate which of the underlying processes are most responsible for the differences between emissions estimates from the various models, and to generate realistic estimates of uncertainty.
2. The general consensus was that while the different emissions models could capture the short timescale fluctuations in emissions caused by instantaneous fluctuations in temperature and light, the models are not as successful in simulating lower frequency fluctuations such as seasonality. This is in part a result of the scarcity of flux measurements outside of the Northern Hemisphere summer or over sufficiently long timescales.
3. There was strong agreement that long-term measurements of the fluxes of isoprene and other bVOCs from different biomes are essential to enable the community to address the current limitations of the emissions models. The funding difficulties of this were recognised but it was felt that a framework such as FLUXNET could be successfully adapted for at least isoprene measurements, but infrastructure funding would be required. The need for simultaneous measurements of the fluxes of NOX, ozone and of speciated monoterpenes was also highlighted.
4. There is a pressing need for all of the bVOC flux data from the various campaigns to date to be collated into a single database, and that it is clear which data are used for developing the models and which can then be used for validating models.
5. A further inconsistency arises from the switch from predominantly leaf-level flux measurements to more canopy-scale and ecosystem-scale measurements. It was suggested that a model monoculture plantation of, for example, poplar would allow the community to make long-term measurements at all spatial scales within and above the canopy to gain an insight into canopy processing and to bridge the gap between scales.
6. The modelling community need to identify exactly what measurements are required (in terms of both fluxes and micrometeorology or plant physiology) and where. Are we missing representative locations (such as high-latitude remote sites) or particular ecosystems?
7. The development of the MEGAN model at NCAR and its availability as a community resource is greatly appreciated by the community. However, there was a feeling that parts of the model are opaque to users, making it difficult to apply and develop. This particularly applies to the derivation of the emission factors used for the various plant functional types. It was also felt that there are too few plant functional types used (although the treatment of plant types will change in MEGANv2.1) and that the leaf area index used (one value per grid cell) is too coarse and fails to capture the limited growing seasons at mid to high latitudes. It was suggested that a MEGAN user group be convened and that an interactive community forum be developed.
8. There are still uncertainties in the chemistry of many bVOCs (including isoprene) and it was felt that atmospherically realistic chamber and lab experiments were needed, as well as comprehensive within canopy measurements and modelling to allow theory to be tested against observations. There was also a feeling that many chamber experiments still use unrealistically high ratios of oxidants to VOCs, and also tend to focus on single compounds rather than mixtures. Experimental confirmation of the recently proposed recycling of oxidants during the oxidation of isoprene is essential.

The meeting was financially supported in part by the UK's Natural Environment Research Council's National Centre for Atmospheric Science. The next workshop in this series is planned for spring 2013 at the UK Met Office in Exeter.

Report on the second international workshop on tropospheric ozone changes

Toulouse, France • 11-14 April 2011

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Summary

Tropospheric ozone has long been recognized as a harmful air pollutant affecting human health and terrestrial ecosystems. It also plays a role in climate change as the third most important greenhouse gas. In spite of several observational records spanning a period of more than 30 years by now, many features of the ozone variability remain poorly understood. Measurements from ground stations, balloons and aircraft cannot always be reconciled and sometimes give conflicting information regarding the long-term changes of tropospheric ozone concentrations. Satellites have begun to provide some useful information regarding ozone in the troposphere, but the accuracy is still not ideal and the retrievals offer poor vertical resolution. Numerical models show some skills in predicting ozone concentrations and they are contributing to our understanding of the global tropospheric ozone budget. However, the spread among individual models is still large and up-to-now they cannot satisfactorily reproduce past ozone changes.

The lack of consensus in the scientific community on how tropospheric ozone has changed in recent decades stimulated the organization of two open scientific meetings on this topic. The first workshop took place in October 2009 and was hosted by NOAA in Boulder, Colorado [*Logan et al.,* 2010]. At that meeting about 40 participants predominantly from Europe and North America gathered to assess the status of available data sets from in-situ measurements and to attempt a first qualitative synthesis of tropospheric ozone changes across the world. Although this workshop highlighted many ongoing issues, due to inconsistencies in the statistical treatment of the data, differing lengths of time series, a lack of data series from Asia and some issues concerning the quality of the data record prior to the mid 1990s the outcome was not very conclusive. It was therefore agreed to organize a second workshop in order to (i) enhance participation from Asia and other regions of the world, (ii) attract members of the satellite data community and atmospheric modellers to provide their views, and (iii) refine the analysis of datasets from individual regions by providing standardized analysis templates.

This second workshop on tropospheric ozone changes was organized by the Laboratoire d'Aérologie and took place at Météo France in Toulouse, France, from April 11 to April 14, 2011. More than 90 abstracts for oral and poster presentations were received and thanks to 7 organisations (Météo France, Laboratoire d'Aérologie and Université Paul Sabatier from Toulouse, the European ACCENT+ and IAGOS programs, CNRS-INSU and WMO) which sponsored travel and lodging for 14 participants, 70 people from 17 countries attended the meeting. Importantly, contributions from China, South Africa, Indonesia and Australia were received.

While there was again a strong focus on the analysis of individual data sets from different world regions, two sessions were dedicated to the specific needs for the evaluation of long-term model runs and to methodologies and benchmarking data sets. Links to the modelling community were strengthened by the back-to-back scheduling of this workshop with a meeting of the IGAC/SPARC Atmospheric chemistry and climate model intercomparison project (ACCMIP) as well as a joint session between the two gatherings.

The workshop made progress towards a robust and comprehensive analysis of tropospheric ozone changes during the recent decades but also identified several issues that limit the scope which such analyses can assume. The most serious concerns are related to insufficient geographical coverage of long-term observations and the lack of a community approach to evaluate tropospheric chemical processes in numerical models. Methods need to be developed, in a dialogue between the measurement and modelling communities, how to best confront simulation results with observational data. Other issues that were discussed concern the quality of various long-term data records; it was found that agreement between adjacent sites has improved considerably during the

late 1990s, but discrepancies remain in earlier portions of the record.

The workshop presentations and several posters are available from the meeting web site at http://mozaic.aero.obs-mip.fr/web/features/workshop.html. An important outcome of the workshop is the formation of several regional working groups to address unresolved scientific issues. It is possible to contribute to these discussions on the TROPO3 Wiki at http://icg-ii-wikis.icg.fz-juelich.de/tropo3. A WMO GAW report of the workshop proceedings is under preparation.

Key challenges to ozone observations

Many of the available measurements of tropospheric ozone concentrations are from regional air quality networks which were set up in North America, Europe and more recently also in Asia as a response to legal requirements to control air pollution. These sites are often located in urban or suburban environments where they are influenced by local pollution sources. As a consequence they are rarely representative of larger regions and poorly reflect large-scale long-term changes in tropospheric ozone concentrations. Only a limited number of long-term observations exist in the "unpolluted" troposphere outside of Europe and North America. Most of these are coordinated by the Global Atmosphere Watch (GAW) Programme of the World Meteorological Organization. The data are submitted to the World Data Center for Greenhouse Gases (WDCGG) in Tokyo, Japan. A snapshot taken from the WDCGG surface ozone database reveals that coverage of most world regions remains rather poor (Figure 2).

Ozone sondes, which provide some of the longest observational records on tropospheric ozone were originally designed to primarily measure ozone in the stratosphere. Sensor optimization and data processing routines still reflect this focus although significant efforts have been made recently to improve the quality of the tropospheric (as well as stratospheric) measurements. Insights from experiments like JOSIE [*Smit et al.,* 2007] have demonstrated that solution parameters and sonde preparation significantly influence the sensitivity of the electrochemical ozonesonde sensor. Even small changes, if systematic, can have significant impact on long term trends derived from sonde data. For this reason, lack of documentation about the details of sonde preparation and data processing often makes it difficult to reliably assess the uncertainty of some early records. Infrequent sampling (many stations launch ozone sondes once per week or less and time series are often interrupted, see Figure 3) adds further uncertainty to the derivation of ozone changes from sonde measurements.

Since 1994, measurements on board commercial passenger airliners (http://mozaic.aero.obs-mip.fr/web/features/information/overview.html) have provided another valuable data set for tropospheric ozone with more than 20,000 flights (i.e. 40,000 vertical profiles upon take off and landing) up to now. Regular instrument calibration and inter-calibration ensures homogeneous data quality with estimated accuracy of 2 ppbv ± 2% at 4 seconds time resolution. The sampling frequency exceeds that of ozonesondes for many locations. The geographical coverage is

Figure 1. Participants of the second international workshop on tropospheric ozone changes.

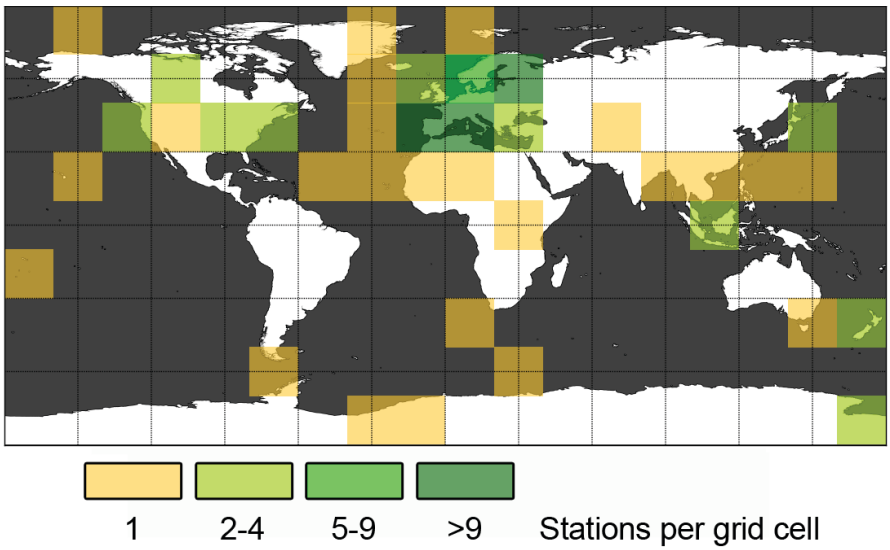

Figure 2. Number of surface ozone stations of the Global Atmosphere Watch network with valid data reported to the World Data Center for Greenhouse Gases in Japan segregated into boxes of 20° longitude by 20° latitude.

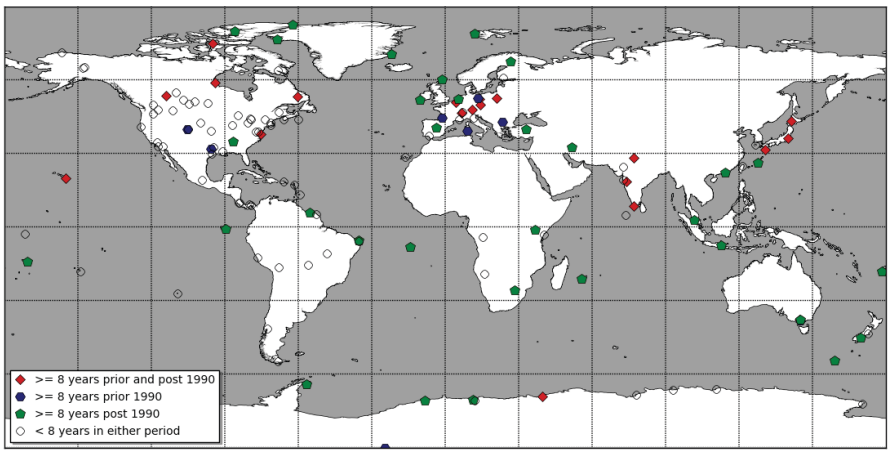

Figure 3. Ozone sonde launch sites and data coverage in the World Ozone and Ultraviolet Radiation Data Centre in Canada.

somewhat biased towards Europe, North America and Africa. There are no data in the Pacific region and only few over the Southern Atlantic and South America. While aircraft data are generally regarded as statistically robust in the free troposphere, they are biased towards polluted environments in the planetary boundary layer. The MOZAIC measurements are being phased out at present and will be continued under IAGOS (http://www.iagos.org/). Earlier aircraft measurement programmes (e.g. GASP, CONTRAIL, NOXAR, CARIBIC) are less suitable for the analysis of long-term tropospheric ozone changes due to short programme duration, low sampling frequency, missing ozone instrumentation or potential issues with data quality.

Besides the lack of data for some world regions and the scarcity of tropospheric ozone data prior to 1990, quality assurance and quality control remain challenging issues, in particular when measurements from different networks or different platforms are combined. The WMO GAW programme makes an effort to coordinate standard operating procedures, calibration and data processing across international and national ozone measurement programmes, but it is still a long way to the full implementation of QA/QC procedures worldwide.

Finally, there is some debate in the scientific literature about the best way to derive robust statistical trends from tropospheric ozone measurement series. Various approaches have been suggested and applied to individual data sets. To date there is little consensus regarding how the data should be treated prior to the trend analysis, which method is best suited to quantify long-term concentration changes and how to assess the robustness of a statistical trend. Often, tropospheric ozone changes are more pronounced in one season than in others, and the magnitude of the change also depends on the metrics that is being examined.

The second workshop on tropospheric ozone changes made important contributions to understanding the challenges described above, to closing gaps in the observational records, to harmonizing the analysis of existing data sets and to beginning to unravel the various causes leading to or inhibiting changes in regional ozone concentrations.

Overview of the workshop programme

The workshop programme consisted of seven thematic sessions:

- Data analysis in the upper troposphere and lower stratosphere, global analysis
- Data analysis over Asia
- Data analysis over the southern hemisphere and Tropics
- Updates of analyses over North America and Europe
- Discussion session: State-of-the-art and future needs

- Evaluation of long-term model runs
- Common methodologies and standard data sets

Common methodologies and standard data setsThe first three sessions were dedicated to the analysis of time series from regions that were not covered well in the Boulder workshop. Presentations included data summaries from the IAGOS/MOZAIC passenger aircraft sampling programme, comparisons of aircraft data with ozone sonde measurements, analysis of surface station observations and results from numerical model simulations.

As introduction to the global data analysis session **Oksana Tarasova** presented tropospheric ozone observations in the GAW Programme of WMO (http://www.wmo.int/gaw). These fall under the coordination of two expert groups: ozone sonde observations are supported by the scientific advisory group (SAG) for ozone while surface ozone measurements are addressed by the SAG for reactive gases. Continuous measurements from mobile platforms (aircraft vertical profiles) are considered by both SAGs as a "vertical dimension" link. The GAW programme has developed a stringent QA/QC system and supports its implementation via Central Calibration Laboratories which maintain the primary standards and World Calibration Centers which propagate these standards to the field observations.

Four baseline NOAA observatories (Barrow, Alaska; Mauna Loa, Hawaii; American Samoa and South Pole) with long records (35 years) were compared by **Irina Petropavlovskikh** and **Samuel Oltmans**. At Barrow and Samoa there has been no significant change during any of the considered periods. At Mauna Loa the largest changes (increases) occurred during the seasonal minimum in the most recent decade (2000-2009) related to changes in the transport patterns with more frequent flow from higher latitudes. Springtime ozone concentrations at Mauna Loa have slightly decreased in contrast to expectations based on increasing Asian precursor emissions. Summertime concentrations at South Pole rose after 2000 while winter and spring concentrations exhibited a minimum during the 1990s. The record of three Atlantic stations (Iceland, Bermuda, Barbados) indicates increasing concentrations after 1990.

The upper troposphere lower stratosphere (UTLS) region was assessed by **Valerie Thouret** utilizing data from the MOZAIC/IAGOS passenger aircraft sampling programme and by **Johannes Staehelin** based on GASP measurements between 1975 and 1979. MOZAIC data show a significant UTLS ozone increase in all regions between 1995 and 2000. After 2000 the rise continues only over the Black Sea region and over East Asia. Overlays of gridded data from GASP and MOZAIC indicate relative changes of up to 40% over the Middle Eastern region and some parts of Asia from the late 1970s to the 1990s, while changes are generally below 10% over Western Europe and North America. This is in contrast to the results from Brewer Mast sondes launched at Hohenpeissenberg and Payerne, which found larger increases. **Johannes Staufer** found good agreement between the ozone sonde time series of Payerne, Uccle and De Bilt and MOZAIC data between 2002 and 2007 in the UTLS.

Several presentations addressed the role of stratosphere-troposphere exchange (STE) for ozone variability in the troposphere. Aircraft observations in the UTLS region demonstrate a strong coupling between the upper troposphere and lower stratosphere, which may however be influenced by the data selection procedure. **David Tarasick** analyzed balloon-borne ozonesonde measurements at 10 stations in Canada to study ozone variability in the UTLS region between 1980 and 2009. He showed that both tropospheric and lower stratospheric ozone concentrations exhibit large interannual variability but no significant long-term trend. There appear to be systematic decadal changes which may be controlled by small alterations in the Brewer-Dobson circulation. According to his analysis, tropospheric ozone changes over Canada are largely controlled by stratospheric variability. Satellite retrievals from the SCIAMACHY instrument show some promising signatures of regional tropospheric ozone enhancements as demonstrated by **Felix Ebojie**. However, the comparison with ozone sondes or MOZAIC data is not satisfactory yet.

The asymmetry between tropopause level ozone concentrations in the northern and southern hemisphere can cause erroneous quantification of STE depending on the definition of the tropopause via the temperature gradient, a potential vorticity threshold or a tracer concentration threshold. **Michael Prather** proposed a new chemical diagnostic for the tropopause in numerical models based on an artificial "e90" tracer with uniform surface emissions and a 90-day e-folding lifetime. This approach helps obtain unbiased estimates of the STE contribution to tropospheric ozone trends. A detailed analysis of the STE impact from 1990 to 2009 using three different ozone tracers in the CAM-Chem model was presented by **Peter Hess**. According to his simulation, the stabilization of northern hemispheric tropospheric ozone levels after 1999 is due to a combination of decreasing tropospheric ozone production and a continued increase of lower stratospheric ozone concentrations. The stratospheric contribution to ozone concentrations in the middle free troposphere (500 hPa) can exceed 50% and is generally larger at higher latitudes. These model findings are consistent with some but not all observational data sets. A significant influence of the STE on lower tropospheric ozone concentrations modulated by El Niño Southern Oscillation variation was found by **Meiyun Lin** who also noted that her model could not reproduce the changes in surface ozone con-

centrations in the northern hemisphere mid-latitudes, presumably because of underestimated nitrogen oxide emission trends in Asia. The model yielded a very good representation of monthly mean ozone anomalies in the tropopause region but could not capture anomalies in the lower troposphere.

Various new data sets from the Asian continent fill important gaps in the observational network. **Hiroshi Tanimoto** analyzed surface ozone observations from 1998 to 2007 at seven remote sites in Japan and on two islands east of the main island. While the low-altitude sites do not show a significant change during this period the elevated site at Mt. Happo exhibits a distinct increase of ozone of 1.25 ± 0.53 ppbv yr^{-1}. **Jennifer Logan** reports that she finds similar changes in the ozone sonde observations from Tateno and Sapporo and in MOZAIC data. The sonde and MOZAIC measurements are similar over central Japan except for summer months when local pollution from Tokyo affects the soundings. This was confirmed by **Regina Zbinden** who analyzed MOZAIC and ozone sonde data at various altitude levels. Some inconsistency remains between the ozone changes derived from the low-altitude surface measurements and the trends that are calculated from low-altitude sonde data. This may be related to changes in the type and preparation procedures of Japanese ozonesondes.

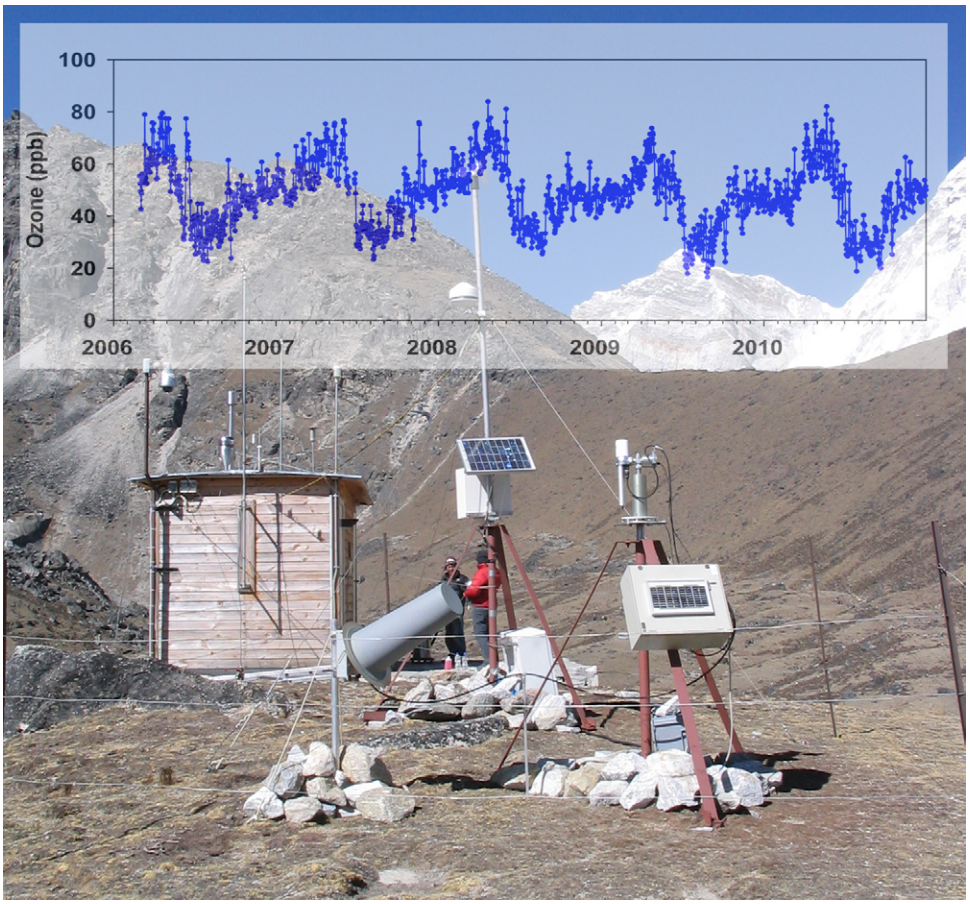

Figure 4. The Nepal Climate Observatory – Pyramid GAW Global Station at 27°57′ N, 86°48′E, 5079 m asl and the first five years of ozone measurements at this site. Photograph and data plot by ISAC-CNR/EV-K2-CNR.

Aijun Ding and **Tao Wang** presented a comprehensive analysis of tropospheric ozone observations in China using data from MOZAIC, from the 2007 North China Aircraft study, from a background site at Hong Kong and from other Chinese sites. The average ozone trend at Hong Kong for the period 1994-2010 reached 0.52 ppbv yr^{-1}. The data from other stations of the national observational network are still too short to allow for trend analysis. Data from the Chinese mountain station Mt. Waliguan were presented by **Xiaobin Xu**. Daytime observations exhibit a continuous increase since 1994 while nighttime concentrations rise significantly only until the year 2000. The largest increase in daytime concentrations is observed in fall (+0.26 ppbv yr^{-1} from 1994 to 2009). This change is much smaller than the change observed at Mt. Happo in Japan.

Paolo Cristofanelli introduced a recent addition to the global GAW network of surface stations at the Nepal Climate Observatory – Pyramid (PYR) where measurements began in 2006 (Figure 4). The site is influenced by STE events and direct transport of pollution from the Indian subcontinent.

Mikhail Arshinov presented surface ozone measurements from the TOR station Tomsk, Russia, between 1990 and 2009 and data from regular aircraft measurements southwest of Novosibirsk between 1997 and 2009. From monthly mean anomalies a significant negative trend in the surface ozone mixing ratio was found in the 1990s (-0.51±0.42 ppb/yr) while the trend was insignificant for 2000-2009 and 1990-2009. The aircraft data exhibit small positive but statistically insignificant trends at all levels up to 7 km. The exceptional downward trend at Tomsk may be related to the absence of major upwind pollution sources which impact most other sites in Asia.

Data from the Indian subcontinent

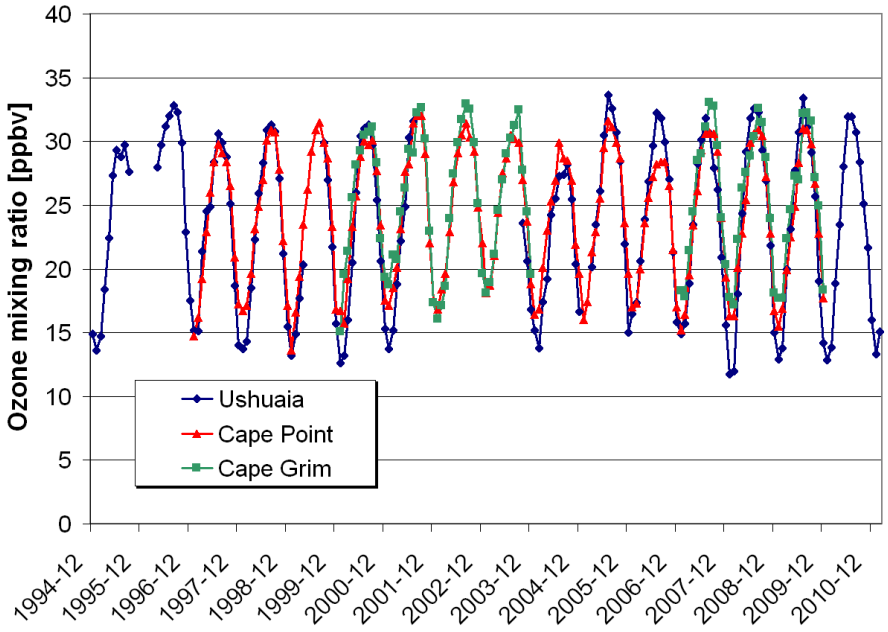

Figure 5. Comparison of surface ozone measurements from three stations in the extra-tropical southern hemisphere. Data selected for background conditions. Courtesy: S. Luppo.

were presented by **Valerie Thouret** who analyzed MOZAIC data over New Delhi. This location exhibits a substantial positive trend between 1 and 8 km altitude over the entire period from 1995 to 2005. The rate of change appears to slow after the year 2000.

Data from three extra-tropical stations in the southern hemisphere were presented by **Ian Galbally** (Cape Grim, Australia), **Ernst Brunke** (Cape Point, South Africa) and **Sergio Luppo** (Ushuaia, Argentina). All three stations show a rather coherent time series after 1995 if data are selected for background conditions (Figure 5). The measurements at Ushuaia experience baseline conditions only about 34% of the time so that data must be filtered before analyzing trends. No significant changes were found since 1994 when the measurements started at Ushuaia. Surface ozone at Cape Grim exhibits positive trends during the 1980s and 1990s in Austral spring (SON) and summer (DJF) and no significant changes during other periods and seasons. The time series of surface ozone at Cape Point (data since 1983) can roughly be divided into 3 parts with a significant rise only during the period 1990–2002. The ozone increase at Cape Point was 0.34 ppbv yr-1 compared to only 0.12 ppbv yr-1 at Cape Grim. As reported by **Ian Galbally** no significant trends were detected in ozone sonde data at 38°S, 54°S and 67°S after 2000 except for a springtime (SON) increase in the UTLS over Davis, 67°S, which may be related to circulation changes and stratospheric ozone recovery.

Surface ozone data from three tropical stations were shown: Bukit Kototabang in Indonesia by **A.C. Nahas** and Assekrem (Algeria) and Mt. Kenya (Kenya) by **Christoph Zellweger**. Due to low data availability, trends and monthly anomalies have not been calculated for Mt. Kenya. No significant surface ozone changes were observed at Assekrem between 1997 and 2009. Here also the data availability was low during the first years of measurements. At Bukit Kototabang a slight negative trend was found when all data are considered (-0.24±0.17 ppbv yr^{-1} during 1996-2010). This is driven by changes in daytime concentrations of up to -0.91±0.36 ppbv yr^{-1} after 2000.

Hiroshi Morioka presented ozonesonde data from 10 SHADOZ stations in tropical latitudes for the period 1998–2008, dating back to 1993 for some stations. Climatic patterns as expressed by the El Niño/Southern Oscillation index (ENSO) or the Indian Ocean Dipole mode (IOD) affect the transition between dry and wet seasons at these sites and complicate the derivation of robust trends. Significant increases were seen in the UTLS with a magnitude of 0.2-0.3 DU yr^{-1}. **Françoise Posny** took a closer look at data from La Réunion where negative trends of low significance are found before 2000 and statistically significant increases of 0.33±0.14 ppbv yr^{-1} (average over altitudes from 0 to 16 km) thereafter. MOZAIC data from the upper tropical troposphere over Africa were analyzed by **Jean-Pierre Cammas** who detected a widening of the tropical belt in association with ozone concentration increases of about 0.6 ppbv yr^{-1} over the past 14 years. It is speculated that this observation is a consequence of climate change and results from an intensification of the meridional circulation in the southern hemisphere. Long-term satellite data records from N7TOMS v8, EPTOMS v8, OMI v8.5 and Aura OMI/MLS were used by **Jerry Ziemke** (presentation by **Jose Rodriguez**) to investigate tropical tropospheric ozone trends based on various methods for the derivation of tropospheric column ozone. While large uncertainties persist in these methods, one can detect some robust change features such as a significant positive trend over Southeast Asia.

Europe and North America received most attention in the past and this was reflected by the large number of abstracts received for session 4. Owing to the priorities set out for this workshop and given that Europe and North America featured prominently during the Boulder meeting in 2009, oral presentations about these regions were limited and the reader is referred to the workshop website for detailed information. An analysis of surface measurements is also

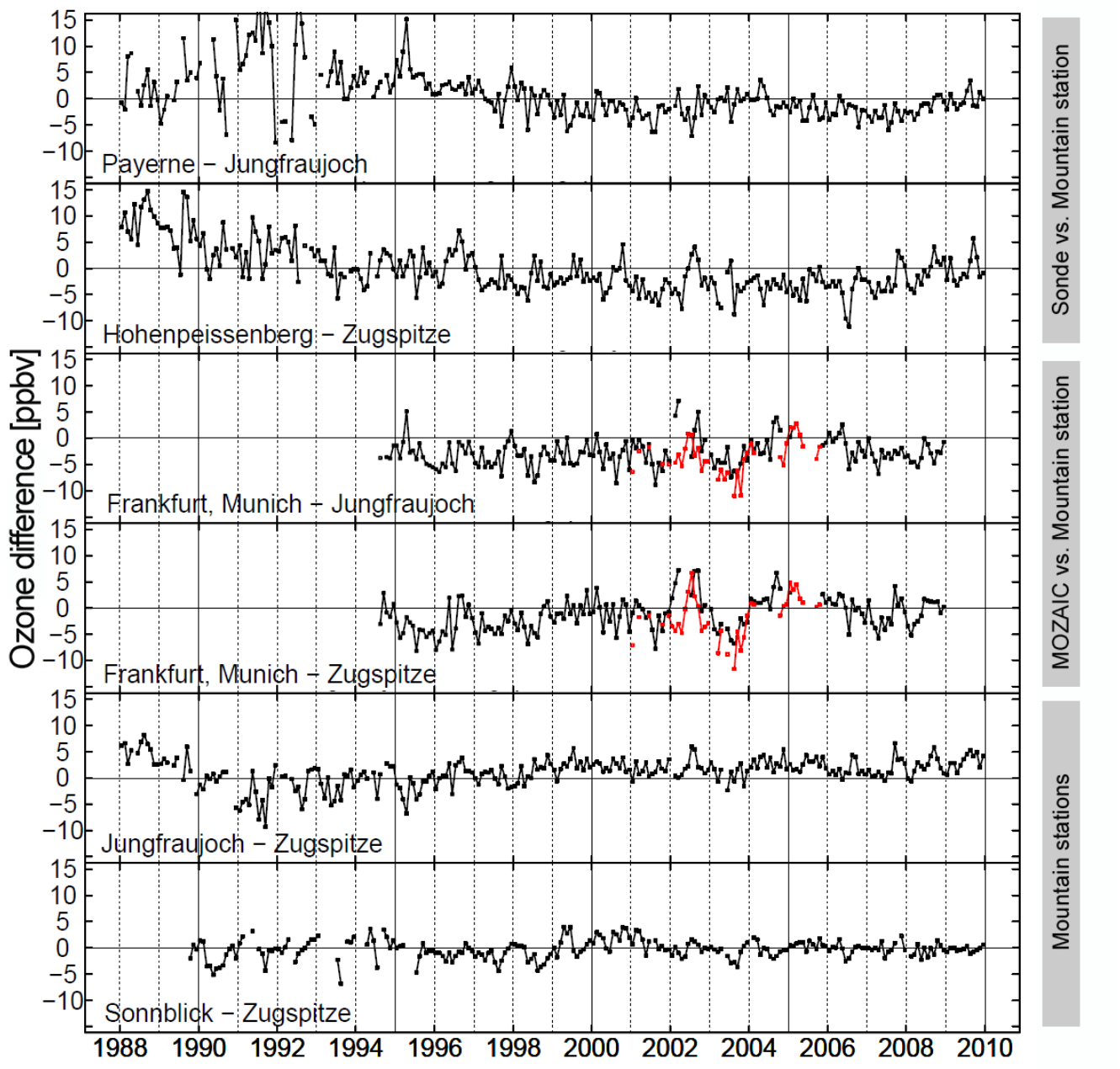

Figure 6. Comparison of time series from European ozone sondes and mountain stations with MOZAIC data from take off and landing at European airports. Courtesy: J. Logan.

contained in the recently published report of the task force hemispheric transport of air pollution [TFHTAP; *Keating et al.*, 2011] which was presented by **Kathy Law** and in a publication by *Rebecca Wilson et al.* [2011] who analyzed trends of mean concentrations, 5%-iles and 95%-iles from European surface sites. **Jennifer Logan**, **Stefan Gilge** and **Barbara Galleithner** summarized long-term changes in free tropospheric ozone over Europe by comparing measurements from sondes, MOZAIC and alpine surface stations. The different data sets show rather consistent behaviour after 1998 (Figure 6) while there are some unexplained differences of up to 15 ppbv in the monthly means during the earlier period. Most data sets show a general increase of free tropospheric ozone concentrations in the 1970s and 1980s (an exception are the ozonesondes from Hohenpeissenberg). Thereafter, the tendency flattens and a downward trend of about -0.2 ppbv yr^{-1} is found after 1998 (mainly during summer). This decrease is consistent with measurements at three French sites (Pic du Midi, Puy de Dome, and Donon) as shown by **François Gheusi**, with background data from the Czech

Republic (**Milan Vana**) and with baseline data from the Mace Head, Ireland station as observed by **Richard Derwent**. Both ozone sondes and a DIAL lidar instrument at the Observatoire de Haute Provence, France, document increases in ozone concentrations at 6–8 km altitude after 1990 as presented by **Gerard Ancellet**.

Rigel Kivi interpreted ozonesonde data from the Finnish Sodankylä station with a statistical model that takes into account the Arctic oscillation index and several proxies for stratospheric variability. The model can explain 75% of the variability in tropospheric ozone at this site. Trend analysis beginning in 1990 will be affected by a strong positive anomaly induced by the Arctic oscillation in 1991. Winter and spring season trends of ozone between 3 and 6 km are 0.36 ± 0.14 ppbv yr-1 (1989-2009) consistent with several other data sets in the high and mid latitudes of the northern hemisphere.

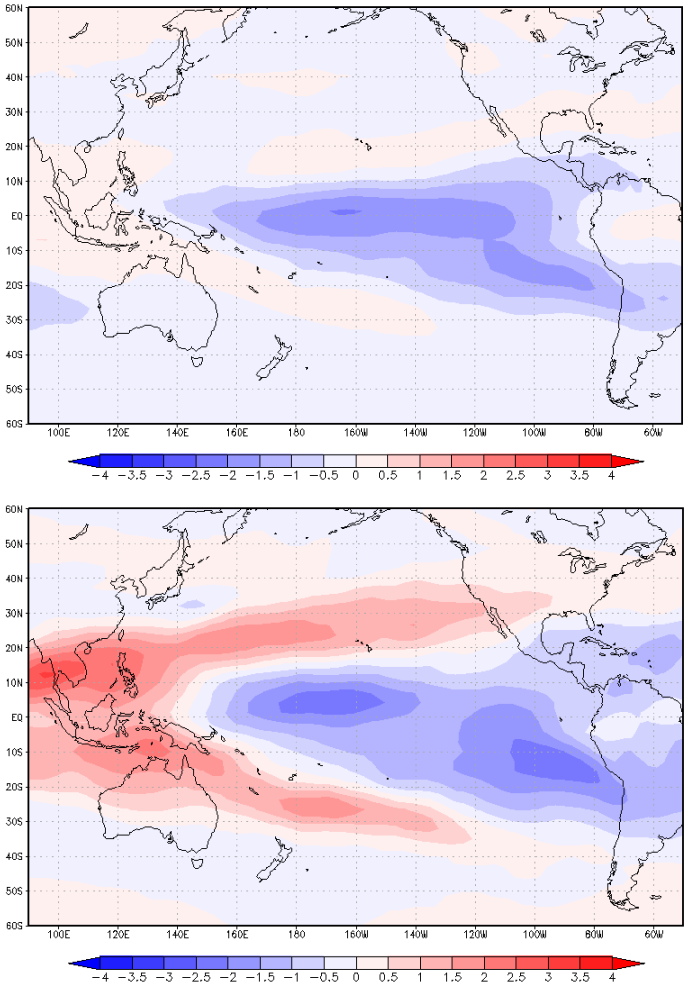

Figure 7. Contributions of ozone transport (top) and photochemically produced ozone (bottom) from precursors in North Africa and South America to the ENSO related ozone anomaly over the tropical Pacific at 500 hPa. Results from the CHASER model. Courtesy: K. Sudo.

A preliminary analysis of ozone changes over New York and Dallas, USA, based on MOZAIC profiles measurements performed since 1995 was presented by **Valerie Thouret**. Similar to Europe, free tropospheric ozone increased prior to 2000 and no significant change can be detected after that year.

The first attempt to simulate multi-decadal tropospheric ozone changes (1960-2000) with a set of three global chemistry models was undertaken in the European RETRO project in 2005 (http://retro.enes.org). The results from the TM4, LMDz-INCA and ECHAM-MOZ models were quite comparable in terms of long-term changes, but they differed in terms of the interannual variability and with respect to the sensitivity of each model to changes in stratospheric ozone content, lightning NO_x and other factors [see *Schultz et al.*, 2007]. None of the models was able to capture the apparent increase in background ozone concentrations that was observed at many surface sites in Europe and North America during the 1990s. Since then, a number of groups have performed similar model runs and investigated various aspects of the tropospheric ozone variability with a focus on the 1990s and more recent years.

Frank Dentener presented results from two 25-year simulations (1980 - 2005) with the ECHAM5-HAMMOZ model [*Pozzoli et al.*, 2011]. One of the simulations applied known changes in meteorology (ERA-40 reanalysis) and in emissions (RETRO emissions data), while the other one was run with constant anthropogenic emissions. According to this study anthropogenic emission changes can explain about half of the interannual variability over the entire period, whereas year-to-year changes are primarily caused by the natural variability (which includes meteorology, biogenic and biomass burning emissions and lightning). The model slightly overestimates ozone concentrations. It captures broad aspects of the variability at several surface sites in Western and Northern Europe, but it has difficulties in reproducing the ozone observations in the Mediterranean region. The different trends in winter and summer at European stations are qualitatively in agreement with observations, while the sign of the winter and summer trends at US stations is reversed.

A set of three reanalysis simulations covering the period 1970-2008 was presented by **Kengo Sudo**. These runs were performed with the CHASER-V3.0 model and used NCEP reanalysis for meteorological input. Tropospheric ozone columns and their interannual variability are reproduced well at selected sites across the globe. The differences between model and ozone sondes are generally smaller than the differences between ozone sondes and satel-

lite retrievals. Clear correlations are found between tropospheric column ozone and major meteorological patterns (El Niño/Southern Oscillation, Atlantic Oscillation, East Asian Jet Stream and Pacific/North American tele-connection). There is a significant contribution of photochemically produced ozone to these patterns (Figure 7). Between 1970 and 1989 surface ozone changes are largely positive throughout the northern hemisphere and tropics while they become slightly negative in many regions after 1990 when an emerging positive trend is detected in the southern hemisphere. Climate change mostly tends to reduce surface ozone concentrations although the opposite signal is seen for example in western boreal latitudes during the 1990s or in South America, South Africa, Australia and the eastern Indian Ocean after 2000. Sudo also highlighted the importance of methane for the tropospheric ozone budget and found a significant reduction of tropospheric methane concentrations due to changes in climate and stratospheric ozone.

Achim Strunk showed results from decadal simulations with the TM5 model (1999-2008) and investigated the impact of different Asian emission inventories on the vertical distribution of ozone and the ozone changes in the troposphere. Over selected airports in Japan and South Korea the model captured the monthly variations of ozone at 850 hPa very well. Differences between a simulation with increasing NO_x emissions from China and a simulation with constant year 2000 emissions amounted to 0.5 ppbv. When the emissions changes are taken into account the simulated decadal ozone change over the western US increases by 0.1 ppbv.

Prodromos Zanis presented an evaluation of the regional RegCM3/CAMx model over Europe based on EMEP surface ozone observations. The model reproduced monthly mean values very well and showed a satisfactory correlation with hourly daytime values while nighttime concentrations were less well captured. There is a seasonal bias with underestimated wintertime and overestimated summertime concentrations. Decadal changes resulting from this model set-up were presented in a poster by **Eleni Katragkou** who found differences of up to 4 ppbv for wintertime and summertime changes between a simulation with reanalyzed meteorology and a run with meteorology from a climate model.

Martin Schultz reviewed the current ability of global atmospheric chemistry models to simulate tropospheric ozone and ozone changes. While most models capture seasonal ozone changes relatively well, they all appear to disagree with observations in terms of long-term changes. While the sparse observations dating back prior to 1990 indicate increasing ozone concentrations well into the 1990s, the modeled trends flatten around 1980. The models react very differently to prescribed changes in climate and emissions and this appears to be related largely to differences in vertical exchange processes. The seasonal change of vertical gradients of tropospheric ozone was proposed as a diagnostic for evaluating model simulations. Schultz highlighted the importance of reliable high-quality data archives for tropospheric ozone observations, the availability of such observations in near-real time and the need for a community approach to standardize model performance metrics.

The importance of reliable global ozone precursor emissions estimates was emphasized by **Claire Granier** who also presented various model results for the 2000-2009 period including the global atmospheric composition reanalysis from the European MACC project.

Anne Douglass gave a presentation by **Luke Oman** on ENSO-related variability patterns of tropospheric ozone. They found a good representation of horizontal and vertical anomaly patterns in the GEOS-CCM when compared with SHADOZ ozone sonde and MLS satellite data.

The discussion on methodologies for the analysis of tropospheric ozone changes was preceded by three presentations covering the impacts of data sampling frequency

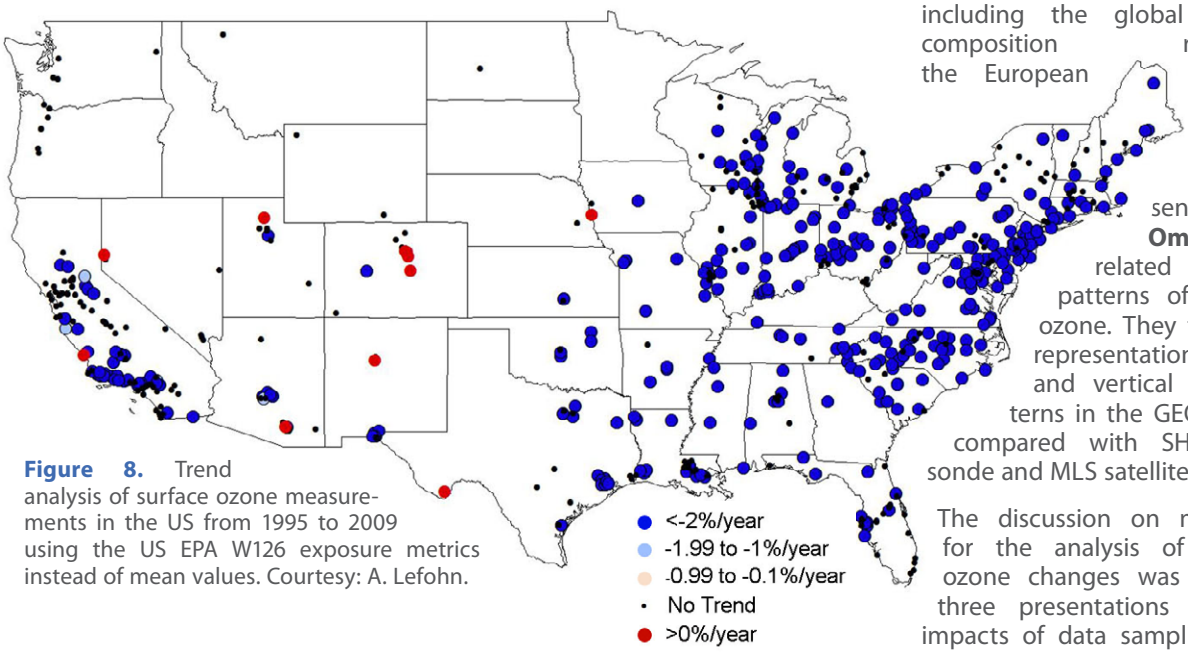

Figure 8. Trend analysis of surface ozone measurements in the US from 1995 to 2009 using the US EPA W126 exposure metrics instead of mean values. Courtesy: A. Lefohn.

(**Marielle Saunois**), the application of different metrics for the derivation of trends (**Allen Lefohn**; Figure 8) and a summary of audit results by the World Calibration Centre for Surface Ozone (WCC-Empa) which checked the quality of ozone measurements at several global GAW stations (**Christoph Zellweger**). In the discussion it was generally agreed that the presentations which made use of the analysis templates facilitated a comparative analysis among different regions. However, it must be acknowledged that any separation of time series into individual periods is somewhat arbitrary and can lead to misleading trend results, because the time series are still too short to be immune to end effects. Since many measurement series suggest some change of behaviour around the year 2000, several participants supported the suggestion to evaluate ozone changes separately for the periods before and after 2000. No unanimous recommendation was made about how the data should be treated statistically, but it became clear that tropospheric ozone changes should not be interpreted based on mean values only. A key question for further analysis is the consistency of ozone changes within and among various world regions. This question is intimately related to the data quality and measurement history. There are relatively few concerns about data taken after the mid-1990s, but some fundamental discrepancies exist between earlier data sets. Obviously this also raises the question how reliable reports are on long-term changes from individual sites in data-sparse regions.

Key outcomes

The workshop made progress towards a more systematic assessment of tropospheric ozone changes based on the in-situ and remote sensing measurements from various platforms and with information derived from global and regional scale numerical model simulations. In most regions of the world — the noteworthy exception being East Asia — surface and free tropospheric ozone concentrations have not risen significantly after the year 2000. Prior to the 1990s almost all records indicate a strong rise, while during the 1990s the picture is very diverse. These summary statements are made with caveats concerning the statistical robustness of the available data sets, the consistency of seasonal and regional changes and the metrics used to evaluate tropospheric ozone changes. In Toulouse it was suggested that we adopt a more community-oriented approach in order to improve our understanding of these issues.

In spite of several decades of tropospheric ozone research it must be recognized that the density of the surface and ozone sondenetworks remains sparse, especially outside of Europe and North America although several countries in Asia installed several new sites in recent years. Measurements in the free troposphere by ozonesondes are usually obtained with relatively low sampling frequency which raises concerns about the statistical significance of these data for the analysis of interannual variability and trends. Measurements on board commercial airliners can in principle be made more frequently, but sufficient geographical coverage has not yet been achieved and sustained. Satellite observations are of some value for tropospheric ozone research due to their global coverage and the ability to simultaneously observe ozone and ozone precursor species. However, in contrast to the situation in the stratosphere, vertical resolution and accuracy of tropospheric ozone retrievals are not yet sufficient to use these data for trend assessments or as constrains for numerical model simulations. Funding constraints lead us to predict that data scarcity will remain a problem in the years to come and it will be a challenge to ensure continuity of measurements at a reasonably large ensemble of core sites.

Global models have demonstrated their potential to successfully reproduce seasonal cycles and amounts of tropospheric ozone. More models are able to capture some aspects of interannual variability, in particular in the free troposphere, while surface ozone changes remain a challenge. It is not clear why most of the models fail to reproduce decadal surface ozone changes when compared to the few available multi-decadal observational records. This is a major challenge for our understanding of the tropospheric ozone budget as described in the current generation of global climate models.

The workshop initiated a dialogue between scientists who perform ozone measurements and the modelling community, a dialogue we hope will lead to improved methods to generate "meaningful" data sets on the one hand and to more a robust evaluation of model performance on the other. For example there was discussion about defining the terms "background ozone" and "trends". More interaction between the measurement and the modelling communities is required in order to identify those data sets which are representative for the typical scale of global or regional model simulations, respectively. Furthermore, a joint effort is needed to formulate suitable algorithms for data selection that can be applied to observational records and numerical simulation results, which then can be automated so that it becomes possible to extend tropospheric ozone studies across different regions of the world. First steps in this direction have been made by recent model intercomparison activities such as ACCENT, TFHTAP or ACCMIP, although a more systematic approach to evaluate the individual processes that contribute to tropospheric ozone (including STE changes, vertical mixing, anthropogenic and natural emission changes, chemical reactions, deposition and washout of precursor species) is needed. Here, the community could learn from the CCMVal concept for stratospheric ozone or the AEROCOM model for aerosol research. Research on tropospheric ozone in the coming years should focus largely

on regional patterns of seasonal and interannual variability and try to identify the processes which control this variability over larger scales. Exceptional episodes (e.g. 1991, 1998, 2003 or 2004) can help to elucidate the impact of weather patterns on tropospheric ozone and prepare for a more robust assessment of the potential impacts of climate change on the chemical state of the troposphere.

In order to facilitate the exchange of knowledge and new results six thematic working groups have been formed wit h the mandate to systematically investigate regional tropospheric ozone changes in Europe, eastern North America, Asia, the southern extratropics, the tropics and the Arctic. In addition, a cross-cutting group will deal with methodological issues and compare the surface ozone records of the different regions. All workshop participants and other interested researchers are invited to register for one or more of these working groups and contribute to the discussions on the collaborative wiki at http://icg-ii-wikis.icg.fz-juelich.de/tropo3. These activities will be coordinated via the GAW Programme with support of the Scientific Advisory Groups on reactive gases and on ozone who have a mandate to seek expansion of the measurement network and ensure data quality. Coordination of activities under GAW will ensure long-term continuity.

The next opportunities to meet, present and discuss progress will be during the Ozone Symposium and the IGAC conference in August and September 2012, respectively. As a result of the Toulouse meeting we expect a couple of key publications on tropospheric ozone changes over the next year, which will be just in time to be recognized in the forthcoming fifth assessment report of the Intergovernmental Panel on Climate Change.

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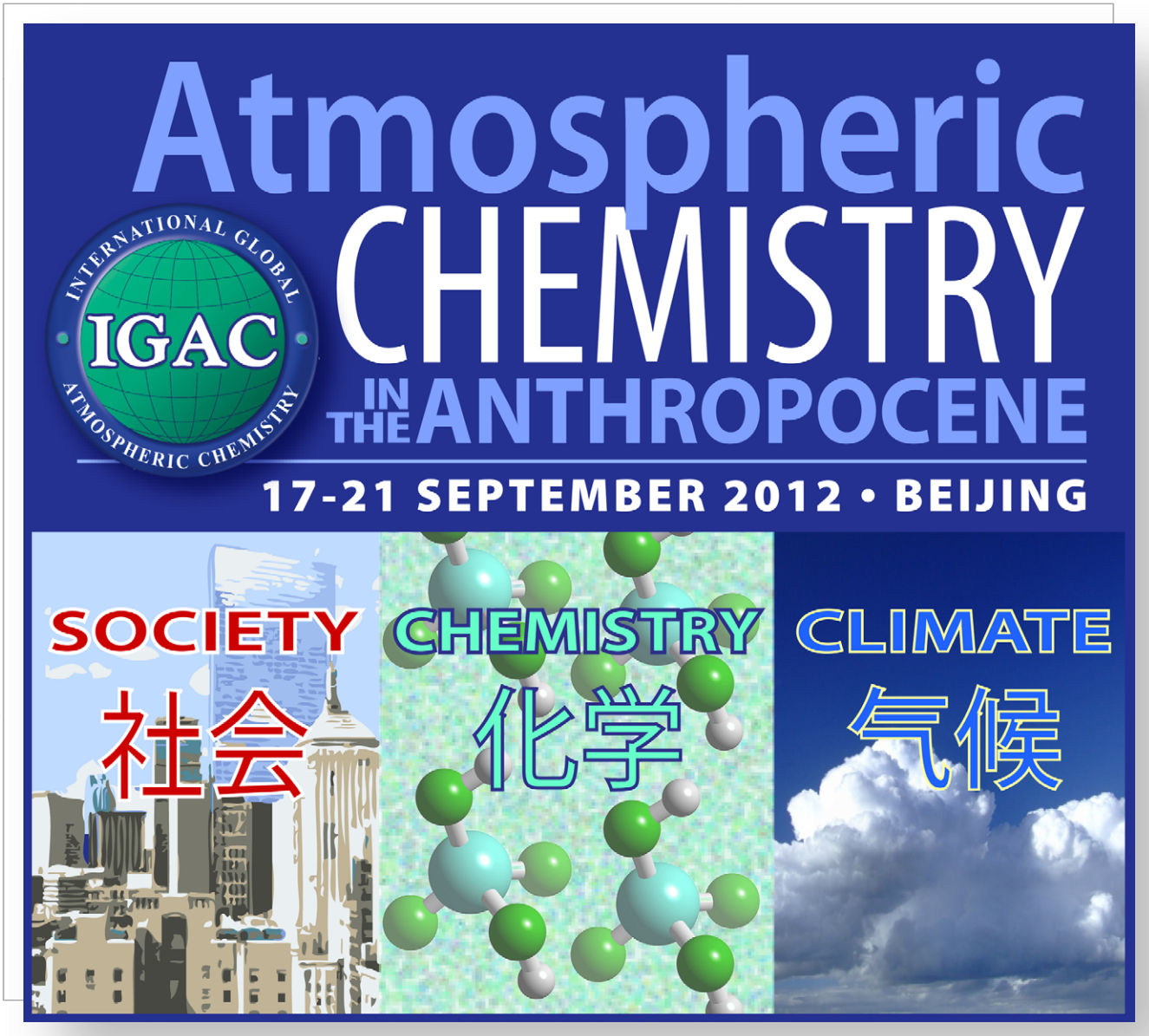
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