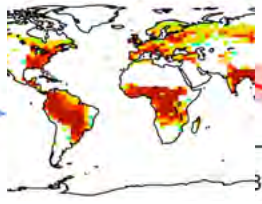
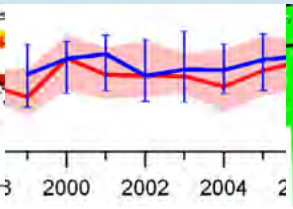
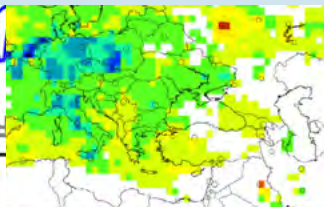
Issue No. 43
January 2011

IGACtivities Newsletter

of the International Global Atmospheric Chemistry Project







A Note from the IGAC Co-chairs: Tong Zhu and Paul Monks

It's been some time since our last newsletter, but this is not a symptom of inactivity. It's been a busy year for IGAC. Foremost, we had our 12th International Symposium, joint with iCACGP, at Dalhousie University in Halifax, Nova Scotia, Canada last July. In this issue, Dentener et al. give an overview of the conference, which was full of challenging science that reflected the changing landscape of our subject area. In many ways the conference did "Challenge the future" and mapped, at in important time for the IGBP family, some of the directions atmospheric chemistry will and should go. We give our heartfelt thanks to Jim Drummond (lead local organizer), Frank Dentener and Randall Martin (scientific program leads) and the many others who contributed to the success of the conference. Preparations are already underway for the next IGAC conference, which will be held the week of 17th-21st September 2012, in Beijing, China, hosted by IGAC co-chair Tong Zhu (University of Beijing).

The New Year marks a significant change with the end of the more than 7-year tenure of Sarah Doherty as Executive Officer of IGAC. On behalf of current, past chairs and SSC members we would like to register our gratitude to Sarah for her unwavering efforts on behalf of IGAC over her tenure. She truly has made the organization tick and made sure that the international collaborative science fostered by IGAC achieves outcome. In February, 2011 we look forward to welcoming Megan Melamed on board as the new IGAC E.O.

In this issue are articles based on two talks given in Halifax, by Bill Collins (UK Met Office) and Hiroshi Tanimoto (NIES, Japan). Collins et al. address a specific aspect of air quality-climate interactions by examining how ozone damage to plants affects the natural carbon cycles through altering plant primary productivity, through to the net effect of reductions in ozone precursor emissions on climate at different timescales. In particular, they look at the very interesting case of NOx emissions, which cool climate by driving decreases in methane concentrations but warm climate by increasing ozone and thereby decreasing the natural carbon sink. The results are somewhat surprising and make a great read! Appropriately following on this, Hiroshi et al. report on tropospheric ozone trends in Japan, where they observe increases in the free troposphere that are most pronounced in the past ten years and for high-ozone events. They conclude that this is likely owing to increasing emissions in East Asia and emphasize the importance of high-altitude sampling sites for quantifying, understanding and predicting regional trends in ozone resulting from changes in emissions.

Randel et al. present a report from a workshop on the upper troposphere/lower stratosphere, where the coupling between atmospheric dynamics and chemistry are complex, making both measurements and modeling a challenge. Much has been learned in recent years about this region through a series of focused, intensive field campaigns and modeling improvements. This article is a great way to get up to speed on what is known and what critical open questions remain. We thank our colleagues at WCRP-SPARC for their work on this contribution.

Finally, Wilson and Monks provide us with a brief overview of the European GEOmon (Global Earth Observation and Monitoring of the Atmosphere) project, which is supporting and helping make more readily available to users the suite of observations available from European ground-based observations of atmospheric composition. Such efforts are critical in helping our community quantify and understand continental-scale long-term chemical changes.

GLOBAL CHANGE IGBP
A Core Project of the Internations Geosphere-Biosphere Programme

In Cooperation with IAMAS Commission on Atmospheric Chemistry and Global Pollution

Report on the 12th Symposium of the International Commission on Atmospheric Chemistry and Global Pollution (iCACGP) and 11th Science Conference of the International Global Atmosphere Chemistry (IGAC) Project:

"Challenging the future"

Contributed by **Frank Dentener** and **Randall Martin** *(scientific co-chairs of the iCACGP-IGAC conference),* **Jim Drummond** (*conference local organizer*), and **Maria Kanakidou**, **John Burrows, Paul Monks, Tong Zhu** and **Sarah Doherty** (*of the iCACGP/IGAC steering groups*).

What was the conference about?

The most recent iCACGP/IGAC conference took place in Halifax, Canada, 12-16th of July, 2010. The conference, with the overarching theme "Challenging the future", was organized around five sub-themes:

1. *Climate chemistry interactions*
2. *Observing atmospheric composition*
3. *Chemistry at the interfaces*
4. *Trace gas and aerosol source strengths*
5. *Pollutant transformation and loss.*

With ~ 370 participants, 65 oral presentations, and over 400 posters the conference had a very busy schedule. The conference agenda is available at: https://www1.cmos.ca/abstracts/congress_schedule.asp

A remarkable feature was the large number and high quality of young scientists' contributions. A poster competition resulted in six young scientist winners:

Alexander Archibald: *Investigating the impacts of HOx recycling in the oxidation of isoprene: Sensitivity studies of past, present and future atmospheres using the UKCA model)*

Nicholas Boeke: *Investigating Ozone Chemistry with Measurements of NO_2 and HCHO from the Ozone Monitoring Instrument and GEOS-Chem*

Ian Burling: *Results from a large, multi-platform study of trace gas and particle emissions from biomass burning*

Yuanyuan Fang: *Impacts of changing transport and precipitation on pollutant distributions in a future climate*

Roisin Walsh: *Year long measurements and analysis of OVOCs at Mace Head, Ireland*

Zhihui Wang: *Record of Concentration $\delta^{13}C$ and $\delta^{18}O$ of Atmospheric CO over the last millennium from Antarctic Ice Cores*

These young scientists were given the opportunity to give a short presentation during the last conference day and we hope to feature some of their work in the next IGAC newsletter.

While the conference spanned a huge range of topics in atmospheric chemistry, there was overall a lot of attention to formation processes of organic aerosol, measurements (techniques) and modeling frameworks. A number of presentations talked about the increasing importance of satellite observations and their synergetic use in models. Elegant techniques for source apportionment of aerosol-gas, using isotopes and other markers received much attention. Modeling and measurements of HOx radicals, and the potential role of recycling reaction pathways was a fourth major theme.

A major initiative in this conference was to archive all posters, to share them with the broader scientific community. High-resolution photographs of the posters can be viewed online at:

http://www.icacgp-igac-2010.ca/page3aa.html
username: icacgp@igac2010.ca
password: igac2010. theme.

If it had any weaknesses, the conference somewhat under-represented the important area of underlying basic laboratory work, e.g. determining rate constants and cross sections. Also, cross-disciplinary talks linking to other parts of earth system (land, ocean, stratosphere) were somewhat underrepresented. Given its educational role, this is an issue that should be addressed in future editions of the conference in close collaboration with our fellow IGBP/WCRP projects SOLAS, iLEAPS and SPARC.

What are the challenges for the future?

All invited and keynote speakers were encouraged to reflect on the future challenges for atmospheric chemistry. While we have enormously increased our knowledge during the last 3-4 decades, numerous examples were given where we still have to improve our

observational and modeling capacities, to provide evidence-based, credible and useful results for policy making.

On the other hand, there were several examples of where integration of our knowledge with other fields is now happening: air pollution and health, the role of pollutants as short-lived climate forcers, and the interactions of atmospheric chemistry with biosphere and cryosphere. However, much remains to be learned in order to sufficiently assess future impacts on atmospheric composition and climate. The two key-note speakers John Seinfeld, and Douglas Dockery well summarized these challenges in the fields of climate modeling and understanding of pollution and health effects.

What is next?

The IGAC project and the iCACGP will actively keep identifying new directions for international atmospheric chemistry research: reaching out to other research fields, building on a strong disciplinary understanding. At the same time, Ian Galbally's inspiring lecture on the history of atmospheric chemistry made it clear that there is a lot to be learned from the ingenuity and ideas of the 'heroes' of atmospheric chemistry. The more than 50-year-old International Commission on Atmospheric Chemistry and Global Pollution will take it as a challenge to promote these historical perspectives in future conferences.

We thank everyone – participants and organizers- for making this conference such a big success. We are looking forward to meeting you again during the next IGAC conference, in Beijing, September, 2012.

Linking air quality, vegetation, and climate change

Contributed by **Bill Collins** (bill.collins@metoffice.gov.uk) *U.K. Met Office Hadley Center for Climate Prediction and Research, Exeter, U.K.*, **Stephen Sitch** (s.sitch@leeds.ac.uk) *School of Geography, Univ. of Leeds, Leeds, UK,* **Olivier Boucher** (olivier.boucher@metoffice.gov.uk) *U.K. Met Office Hadley Center for Climate Prediction and Research, Exeter, U.K.*

Introduction

Human activity has at least doubled the concentrations of ground level ozone over the 20th Century. Ozone has a climate effect in its own right, being the third most important man-made greenhouse gas after carbon dioxide and methane. It is also poisonous to people and plants. Much research has looked at the impact of ozone on plants in terms of crop yields, but here we summarise our recent work on the consequent impacts on the carbon cycle and ultimately climate (Collins et al. 2010).

About half of the carbon dioxide emitted from fossil-fuel burning to date has been taken up by the land and oceans through the natural carbon cycles. On the land this is because increasing levels of carbon dioxide encourage plant growth (the CO_2 fertilisation effect) and this eventually increases the amount of carbon stored in vegetation and in the soil. Here we show that damaging levels of air pollution in the form of ground level ozone may reduce this ability of vegetation to take up CO_2.

Ozone damage

Ozone enters the plant leaves through holes called stomata where it causes cellular damage, reduces the photosynthetic rates and lowers plant production (Figure 1). We parameterised the sensitivities of plants to ozone

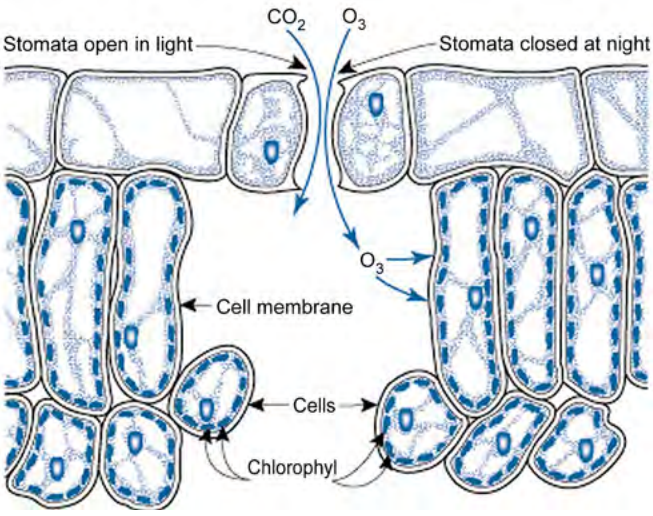


Figure 1. Schematic diagram of a leaf showing a stomatal opening.

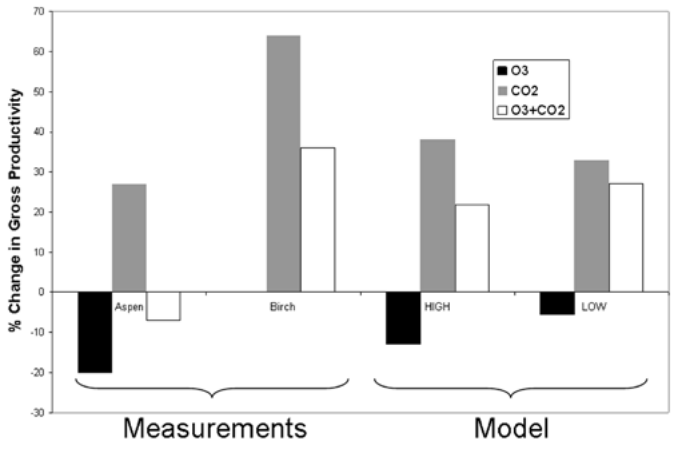


Figure 2. AspenFACE experiments, showing a comparison of JULES simulations against data from Karnosky et al. (2005).

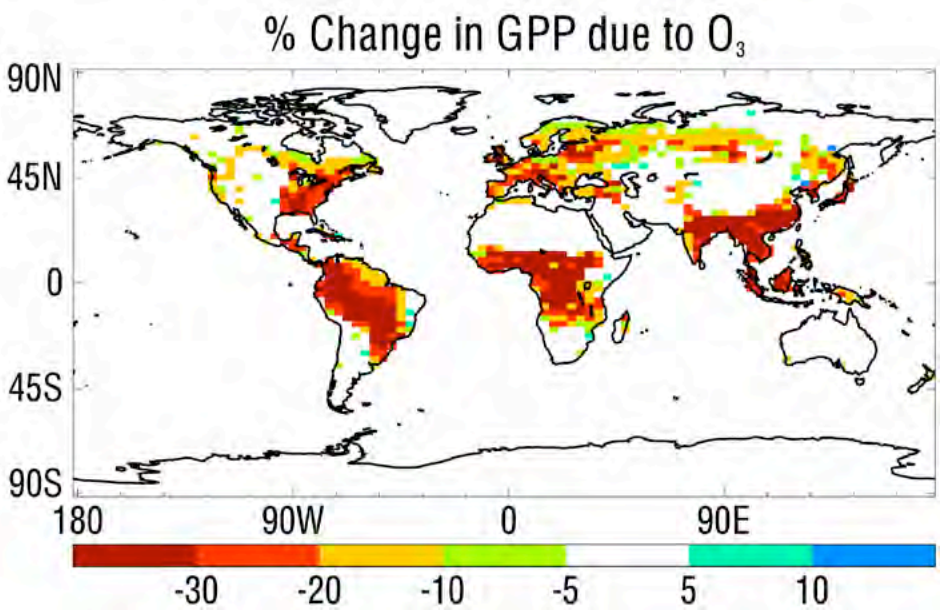


Figure 3. Difference in Productivity (GPP), in percent, of the terrestrial vegetation between a model simulation assuming levels of ozone for the year 1900 and for the year 2100. The future ozone levels were based on a scenario without air quality legislation (SRES A2).

Table 1. Anthropogenic CO_2 emissions over the 20th century in Gt carbon. Equivalent reductions in land uptake (net ecosystem productivity) of carbon due to ozone changes over the same time period assuming a "low" or "high" sensitivity of plants to ozone damage. Data from Sitch et al. 2007.

Changes 1900-2000	Anthropogenic emissions	Reduction in uptake "Low"	Reduction in uptake "High"
CO_2 (Gt C)	387	50	93

in Sitch et al. (2007) from many measurement studies. There we grouped plants into those that had high sensitivity to ozone and those that had low sensitivity. We implemented these effects in the JULES vegetation model and tested the results against "free air CO_2 enrichment" experiments from Karnosky et al. (2005). Figure 2 shows the separate and combined effects of carbon dioxide fertilisation and ozone damage on aspen and birch trees. The modelled changes in productivity for "low" and "high" sensitivity compare reasonably against the observed responses of birch and aspen.

Impact of pollutant emissions

With this model set up we can examine the induced climate impacts from pollutant emissions. Ozone changes for the SRES A2 emission scenario (which assumes no pollution controls) were calculated by the STOCHEM chemistry model (Collins et al. 1997) out to 2100. If pollutant emissions (NO_x and hydrocarbons) were to grow without legislative controls, the resulting increases in ozone by the year 2100 could reduce the productivity of the more ozone sensitive vegetation by over 30% compared to pre-industrial levels of ozone. Figure 3 shows that this damage is large over the tropical forests.

The lower plant productivity in polluted air means less carbon is stored in the plants and soil, and so more carbon dioxide remains in the atmosphere (see Table 1). The climate change due to this extra carbon dioxide can be important. Figure 4 (top) shows the decrease in the carbon stored on land (in vegetation and soils) as ozone pollution levels are increased from 1900 levels to 2050 levels according to the A2 scenario (solid lines). The lower panel of Figure 4 shows that the resulting increase in atmospheric CO_2 leads to a significant radiative forcing of climate, comparable to that from the greenhouse effect of ozone itself. The red and blue lines show the range of values obtained by assuming a high (red) or a low (blue) sensitivity of plants to ozone.

This future damage to plants and climate is not inevitable. Legislation to control air quality is in place in most countries. These measures (which are designed to protect both people and crops) will slow down the damage and reduce the climate impact. The dotted line shows the impact on land carbon and climate of an alternative emission scenario which assumes strict compliance with current air quality control legislation. It was generated by IIASA for the Royal Society report on ozone (Royal Society 2008). This legislation was designed for its air quality benefit

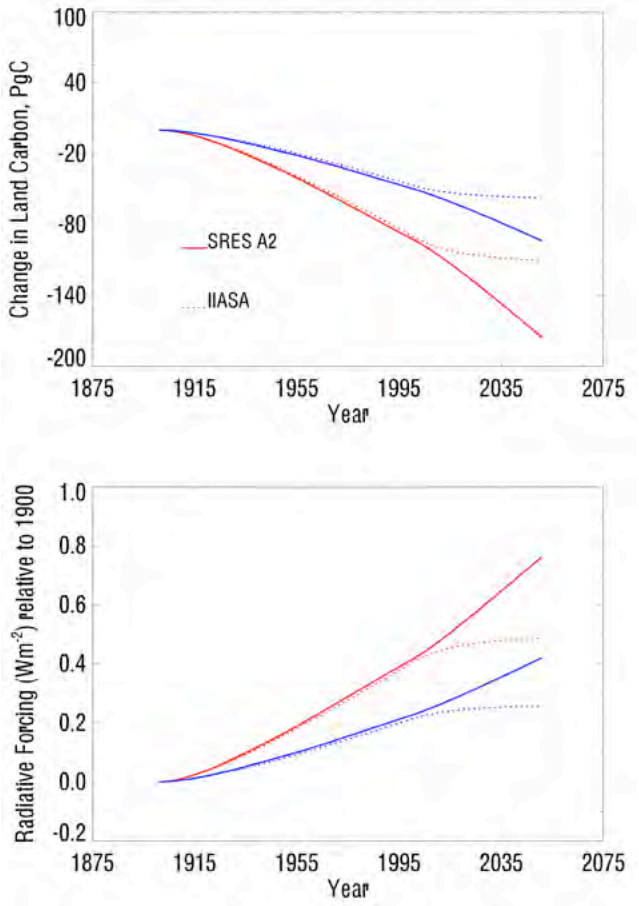


Figure 4. Change in land carbon (top) and consequent change in radiative forcing (bottom) due to increasing levels of ozone pollution. Red lines are for a high sensitivity of plants to ozone, blue are for a low sensitivity. Solid lines into the future assume no controls on air pollution (SRES A2), dotted lines assume air quality controls according to IIASA (Royal Society 2008).

in reducing ground level ozone, but as can be seen in Figure 4 this reduction in ozone also has an important climate benefit.

Climate response to emission changes

NO_X emission controls are often effective at reducing ozone, but NO_X emissions increase the removal of methane (also a greenhouse gas). Previous reports without including the carbon cycle impacts (eg. IPCC 4th Assessment Report) found the impact on methane dominates, so that NO_X emissions on balance cool climate. Therefore reducing NO_X emissions would benefit air quality, but warm climate. We are now in a position to calculate whether this is still true when we account for the damage of ozone to plants.

We run a step change in NO_X emissions through the STOCHEM model to calculate changes in ozone and methane, and use the JULES model to calculate the impact on carbon uptake. We then use simple analytical representations of the carbon cycle and climate (Joos et al. 1996; Boucher and Reddy 2008) to calculate the impact of these changes on global surface temperatures. These are shown graphically in Figure 5 for a NO_X change of 5Tg/yr (around 20% of the anthropogenic emissions). The tropospheric ozone burden and hence the radiative forcing responds almost immediately to the emission change. The climate response to this (diagnosed with the analytical climate response function) is an increase in surface temperature with a longer timescale that plateaus after around 20 years. The methane responds more slowly, decreasing with a 12 year e-folding time, and hence the induced surface cooling takes longer to develop too. The analytical climate (and carbon cycle) response functions are also used to diagnose the warming from the extra CO_2 resulting from the decrease in carbon uptake. The cooling from the methane is stronger than the warming from the ozone, in agreement with the findings of the IPCC AR4. However when we take into account the extra CO_2, this induces sufficient warming to give an overall positive temperature change from NO_X. Now the conclusion is that reducing NO_X emissions is of benefit to both air quality and climate.

For simplicity, Figure 5 neglects further climate impacts of these NO_X emissions through the formation of nitrate aerosols, impact on sulphate aerosols, stratospheric water vapour and nitrogen fertilisation of plants. We can also take this study further by quantifying the vegetation responses to ozone changes resulting from emissions of other ozone precursors: VOCs, CO and CH_4. These precursors too can affect sulphate aerosols and stratospheric water vapour.

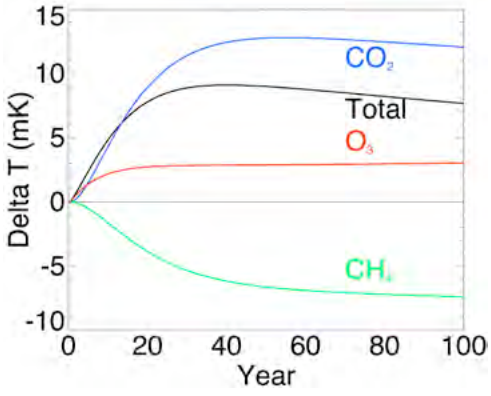


Figure 5. Temperature changes due to ozone, methane and carbon dioxide following a step change in NO_X emissions by 5Tg/yr. The change in carbon dioxide is due to damage to vegetation by the ground-level ozone.

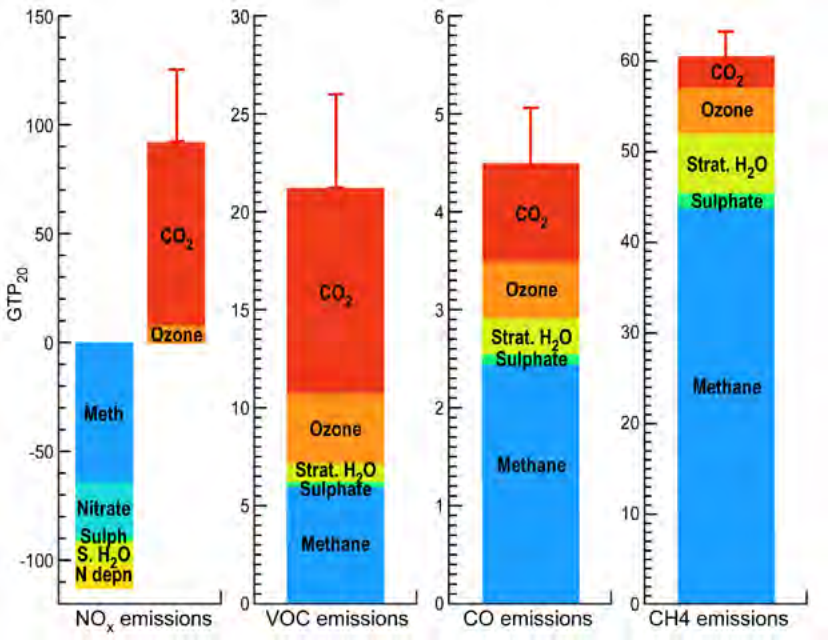


Figure 6. Components contributing to the GTP_{20} climate metric for emissions of ozone precursors. The components are: methane, nitrate aerosols, sulphate aerosols, stratospheric water vapour, nitrogen fertilisation of vegetation, ozone, and carbon dioxide from reduced vegetation uptake. The whisker on top of the CO_2 bar illustrates the range between the "low" and "high" sensitivity assumptions. Aerosol indirect effects are not accounted for. [From *Collins et al. (2010)*, Figure 10].

Figure 6 summarises the total climate impact of all these processes using the GTP_{20} climate metric (Shine et al., 2007), which compares the global surface temperature response after a number of years (20 in this case) after the emission of 1kg of a species to that of an equivalent emission of carbon dioxide. The red bars (labelled "CO_2") show the large contribution made by the extra CO_2 remaining due to the damaging effects of ozone on the vegetation. For NO_X and VOC emissions this is the largest term.

Summary

By damaging plants, ground-level ozone pollution affects the amount of carbon dioxide remaining in the atmosphere. This adds to the direct impact of ozone as a greenhouse gas. We have used a tropospheric chemistry model to quantify the climate impact of different ozone precursor emissions, taking into account their contributions to plant damage. We find that the effect on the carbon cycle makes a significant contribution to the overall climate impact. In the case of NO_X emissions the effect is large enough to turn NO_X emissions into a net climate warming on timescales up to 20 years.

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Tropospheric ozone trends over Japan during 1998–2007

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Introduction

Much attention has been paid to the evolution of tropospheric ozone (O_3), one of the most important constituents of the Earth's atmosphere. A number of studies have provided observational evidence of increases in boundary layer O_3 during recent decades in the northern hemisphere, with growth rates ranging from 0.5 to 0.8 ppbv yr^{-1} (e.g., Jaffe et al., 2003; Lelieveld et al., 2004; Oltmans et al., 2006; Zbinden et al., 2006; Derwent et al., 2007; Parrish et al., 2009; Cooper et al., 2010). Chemistry-transport model (CTM) simulations have not yet quantitatively explained the observed increases, with simulations of background O_3 during recent decades tending to underestimate the observed changes. The major causes for this discrepancy have not yet been identified, and reconciling the observed trends using models is still challenging.

In the northern hemisphere, Asia is now the only region where man-made emissions continue to substantially increase. Emissions of air pollutants from East Asia are estimated to have increased rapidly during the past several decades, in contrast to those from Europe and North America, which have shown decreasing and stabilizing trends, respectively (Akimoto, 2003). Especially in China, emissions of O_3 precursors (nitrogen oxides (NOx = NO + NO_2) and volatile organic compounds (VOCs)) have increased greatly in recent decades (Ohara et al., 2007; Zhang et al., 2007). The growth of NOx emissions in China has been more dramatic since 2000 than during the 1990's, qualitatively in agreement with satellite observations of tropospheric NO_2 columns over east-central China (Richter et al., 2005). These changes would have significant influence on not only local/regional air quality but also on hemispheric transport. Hence the resulting impact needs to be quantitatively assessed.

In spite of this importance, fewer long-term records are available in Asia than in Europe and North America. Measurements by O_3 soundings were often used to infer O_3 trends in the lower troposphere during the past decades (Logan et al., 1999; Naja and Akimoto, 2004; Oltmans et al., 2006). Beginning in mid-1990s, the number of "regionally representative" monitoring sites for surface O_3 have increased in Japan, associated with emerging activities in the atmospheric chemistry research among Japanese scientists. Most of the stations became well established around 2000,

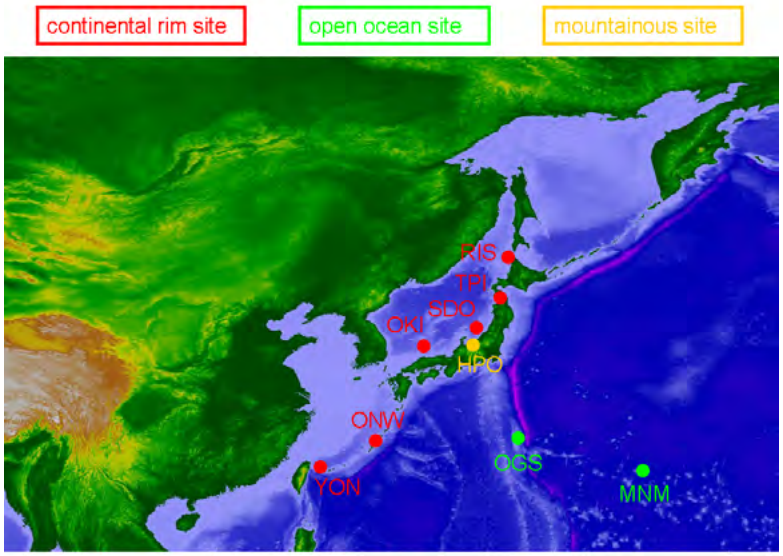

Figure 1. Locations of the stations used in this study. RIS, Rishiri Island; TPI, Cape Tappi; SDO, Sado Island; OKI, Oki Island; ONW, Okinawa Island; YON, Yonagunijima Island; OGS, Ogasawara Island; MNM, Minamitorishima Island; HPO, Mt. Happo.

Figure 2. A photo of Mt. Happo Observatory (HPO) (courtesy of Dr. Shungo Kato at Tokyo Metropolitan University).

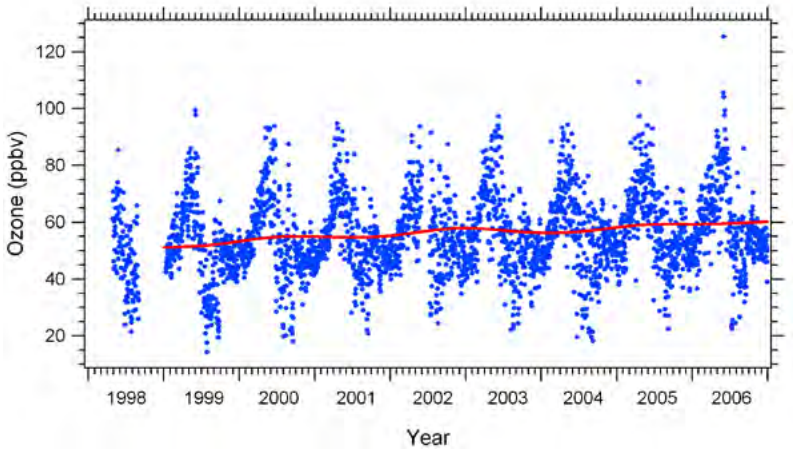

Figure 3. Daily (24-hour) mean ambient mixing ratios of ozone observed at Mt. Happo (dots) for the period 1998 to 2006. Also shown is the trend component (solid line) of the best-fit curve for daily means since 1999. The 1998 data were not used because only 37% of the hourly data were available due to instrument malfunction.

and now a decade of continuous data is available for analysis. Considering the rapidly changing anthropogenic emissions in continental Asia during the last decade, we posed the question "Can we detect an increasing trend in tropospheric O_3 exported from Asia during the last 10 years? If yes, where, how, and why?"

Monitoring sites and measurement quality

We compile measurements from two regional monitoring programs operated in the international framework. One is operated by the Ministry of the Environment of Japan as part of the Acid Deposition Monitoring Network in East Asia (EANET) program, and the other by the Japan Meteorological Agency as part of the Global Atmosphere Watch (GAW) programme of the World Meteorological Organization. We examine nine "remote" sites in different geographical regions where quality-controlled decadal records are available. Of these, six sites represent continental rim sites (i.e., Rishiri Island, Cape Tappi, Sado Island, Oki Island, Okinawa Island, Yonagunijima Island), two represent open ocean sites (i.e., Ogasawara Island, Minamitorishima Island), and one a mountainous site (Mt. Happo at 1850 m) in the Japanese mainland (**Figure 1**). The mixing ratios of O_3 are determined using photometric instruments based on absorption at 253.7 nm emitted by a low-pressure mercury lamp. The instrument is periodically referenced to a working standard placed at each site. The standards referenced in both EANET and GAW programs routinely go through intercomparison with a reference photometer, which is referenced to the Standard Reference Photometer (SRP) #2 at the National Institute of Standards and Technology (NIST) (Tanimoto et al., 2007). The data are scrutinized in the post-analysis, including through a formal Quality Assurance/Quality Control (QA/QC) activity.

Decadal trends observed at Mt. Happo

In this work we focus on the data at Mt. Happo Observatory (HPO), where continuous measurements of surface O_3 started in 1998 (Tanimoto, 2009). The site is located on a ridge on Mt. Happo and faces the Sea of Japan (**Figure 2**). The HPO site is classified as a remote mountainous site because it is rarely impacted by the nearby pollution sources. Domestic pollution plumes are observed only during limited periods in summer (especially August), when land-sea air circulation over Tokyo Bay can occasionally produce medium-range transport (~ 200 km) of pollution from Tokyo to this region, causing a late-afternoon daily maximum. **Figure 3** shows the daily mean mixing ratios of O_3 measured at HPO during 1998–2006. Distinct seasonal cycles were observed, with a spring maximum and summer minimum. The overall trend indicates a continuous increase over the study period.

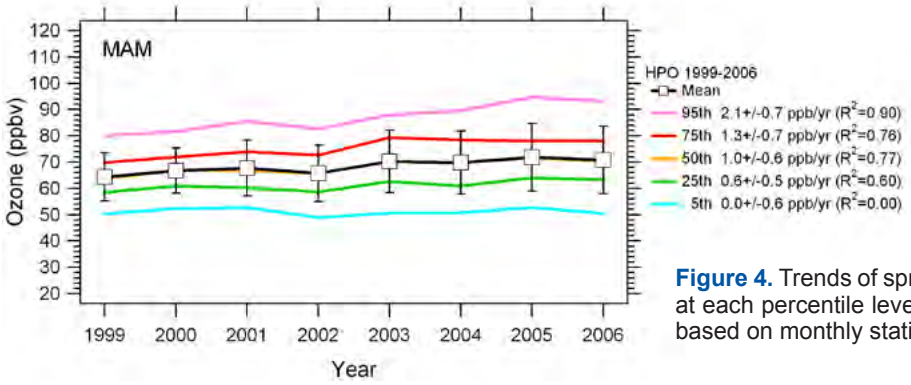

Figure 4. Trends of springtime (March–May) ozone mixing ratios at each percentile level at Mt. Happo for the period 1998–2006, based on monthly statistics.

The mean trend curve shows an initial value of 51–52 ppbv in 1999 and a final value of 59–60 ppbv in 2006, with a total increase of about 8 ppbv.

We analyze only the springtime (March–May) data to highlight broad-scale incidents (i.e., long-range transport). **Figure 4** illustrates the trends of the 5th, 25th, 50th, 75th, and 95th percentiles for the period 1999–2006. Linear regression for each percentile suggests that the trends differ substantially from the lower to the upper percentiles. The trends associated with the 5th percentile are negligible and statistically insignificant. The trends for the median and higher percentile levels are statistically significant with a high degree of correlation (R^2=0.8–0.9). In general, the higher percentile levels show larger increasing trends, with 1.3 and 2.1 ppbv yr^{-1} for the 75th and 95th percentiles, respectively, and 0.6 ppbv yr^{-1} for the 25th percentile. The overall mean trend was 1.0 ppbv yr^{-1} for the study period, in a good agreement with the median trend of 1.0 ppbv yr^{-1}. The increase in springtime O_3 mostly occurs starting in 2003; from 1999 to 2002, there was only a small increase.

Regional emission inventory in Asia

To examine a possible cause-and-effect relationship between the rapid increase in East Asian emissions and the significant O_3 increase observed at HPO, we use a three-dimensional regional CTM based on the Models-3 Community Multi-scale Air Quality (CMAQ) modeling system (Tanimoto et al., 2005; Uno et al., 2007). Briefly, the horizontal and vertical resolutions are 80x80 km^2 and 14 layers up to 23 km, respectively. The SAPRC-99 scheme is applied for gas-phase chemistry. The monthly averaged lateral boundary conditions are obtained from a global CTM. Stratospheric O_3 influx is thus taken into account, but no interannual variations are assumed. We use a yearly-dependent emissions inventory from the Regional Emission inventory in Asia (REAS), which was updated to 2006 by using the latest statistics of energy consumption and industrial activities, and employ the same emission factors (emissions per activity) and removal efficiencies as for year 2003 (Ohara et al., 2007).

Figure 5 shows changes in the emissions of O_3 precursors from East Asia, highlighting a rapid increase during the last decade. Chinese emissions of NOx and VOCs have contributed approximately 80% to the East Asian emissions, with NOx emissions showing accelerating growth after 2000, mainly due to the growth in power plants and transport sectors. In contrast, the Japanese emissions have been relatively small (~10% of the Chinese emissions for NOx and VOCs) and almost constant. We performed two sets of multi-year simulations to study the sensitivity to East Asian anthropogenic emissions: (1) using annually varying emissions inventories (i.e., from the REAS inventory for 1998 to 2007; for 2007, the 2006 inventory is tentatively used due to unavailability of statistical data) and (2) using a fixed emissions inventory from the year 2000.

Decadal ozone trends by chemistry transport model

Springtime O_3 trends observed at all the nine sites from 1998 to 2007 are exhibited in **Figure 6**. There are distinct differences in the O_3 levels between the sites, depending on the site's distance from the continent and latitude. The O_3 levels at the continental rim sites (i.e., RIS, TPI, SDO, OKI, ONW, YON) are substantially higher than at the open ocean sites (i.e., OGS and MNM). The six continental rim sites exhibit an O_3 level of ~50 ppbv for the year 2000. Interestingly springtime O_3 for the marine boundary layer sites located in the eastern Pacific is around 40 ppbv (Parrish et al., 2009). The difference in the two observations suggests a significant O_3 gradient in the marine boundary layer across the Pacific. It is clear that the highest O_3 levels are observed at the mountain site, HPO, with median O_3 levels exceeding 60 ppbv. At the eight surface sites (excluding HPO), the O_3 trends observed from 1998 to 2007 are relatively

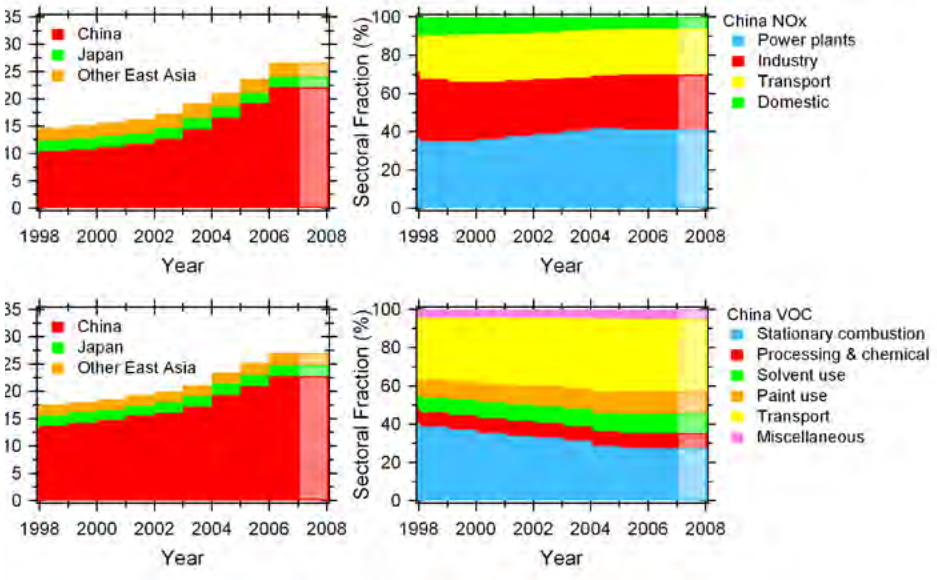

Figure 5. Decadal changes in NOx and VOC emissions from East Asia including China and Japan, estimated by the updated version of the Regional Emission inventory in Asia (REAS) (left). Sectoral distributions for China's emissions are shown (right). The emissions prescribed for the 2007 mode runs were adopted from 2006 (as indicated by shaded bars).

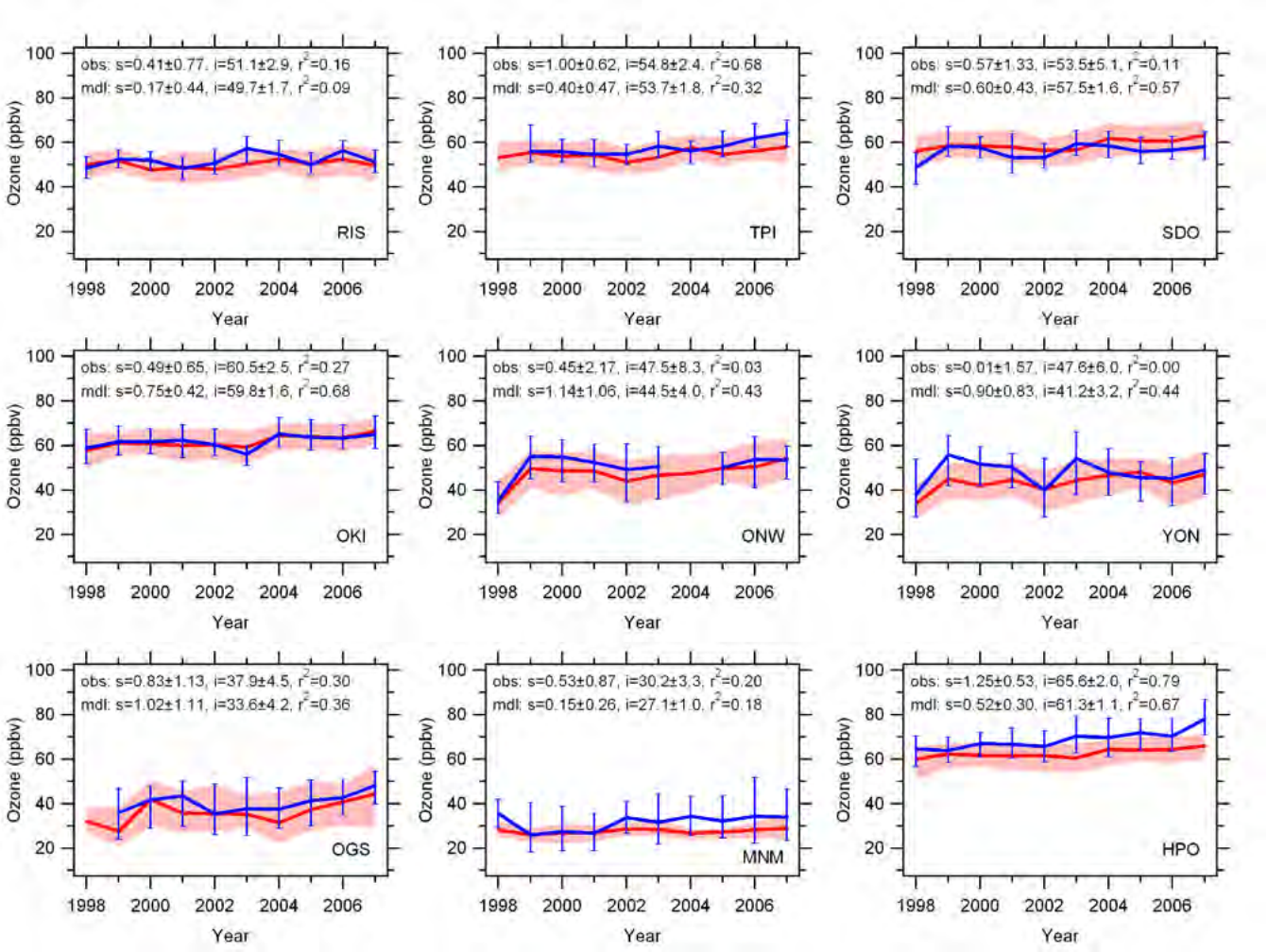

Figure 6. Trends of springtime tropospheric ozone from 1998 to 2007. Plotted are the ozone mixing ratios observed at Japanese stations (blue), and overlaid with a regional chemistry-transport model simulations with yearly-dependent anthropogenic emissions in Asia (red). Observed trends are shown by median (blue lines) and interquartile range (vertical bars), and the modeled trends by median (red lines) and interquartile range (shaded). The coefficient of determination (r^2), slopes (s, ppbv/year) and intercepts for the year 2000 (i, ppbv) with their 95% confidence limits obtained by t-test are given for observations (obs) and model (mdl) results.

small or statistically insignificant. This is likely due to a combined effect of large interannual variability driven by meteorology in the boundary layer and the relatively short time record (i.e., 10 years). In contrast, particularly after 2000, a distinct increase of O_3 is observed at HPO, showing a statistically significant (R^2=0.79) increasing trend at the median level of 1.25 ± 0.53 ppbv yr^{-1}. The model incorporating annually-dependent emissions inventory simulates the levels and the range of variability for springtime O_3 in the boundary layer quite well. The good agreement over the wide latitudinal coverage demonstrates the capability and usefulness of the model in our study. For the high altitude HPO site, the model is able to produce an increase in O_3 (0.52 ± 0.30 ppbv yr^{-1}, R^2=0.67) but this growth rate does not match the observed growth rate.

Possible contributors and factors

Since the observed O_3 increases at HPO are larger for higher concentration ozone events, we examine the modeled O_3 trends at HPO from the 50th to 95th percentiles, deconvolute impacts from emissions and meteorology, and discuss their contributions quantitatively, as shown in **Figure 7**. For the 1998–2007 period, the observed growth rates at HPO are as large as 1 to 2 ppbv yr^{-1} (12 to 22 ppbv $decade^{-1}$) at the 50th–95th percentiles. The modeled growth rates for the 1998–2007 period at HPO driven by growing East Asian emissions are 0.5 to 1 ppbv yr^{-1} (5.2 to 9.6 ppbv $decade^{-1}$) at the 50th–95th percentiles, with greater increases associated with the 95th percentile. The model shows a larger rate of increase in O_3 after 2003, in agreement with the observations. However, the modeled growth rates are substantially lower than the observations, with a noticeable discrepancy after 2003. The model simulations with constant emissions reveal

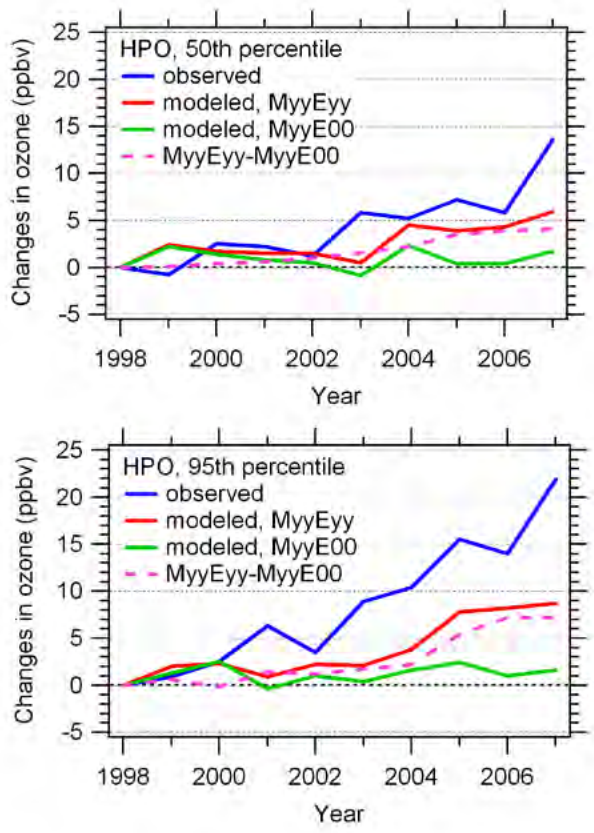

Figure 7. Decadal changes in springtime ozone at Mt. Happo at 50th (upper) and 95th (lower) percentiles in the probability distributions of ozone. MyyEyy and MyyE00 indicate model runs with yearly-dependent and constant emissions inventory (year 2000), respectively, with yearly-dependent meteorological fields. MyyEyy–MyyE00 denotes contributions from anthropogenic emissions only. All the data are normalized to those in 1998.

that transport causes interannual variability in O_3 of ~2 ppbv at the 50th–95th percentiles; however, changes in emissions are the major cause of the observed variation in O_3 at HPO after 2003. The differences between the two sets of multi-year model simulations indicate that changes in anthropogenic emissions and atmospheric transport variations contributed almost equally to the observed O_3 growth during 1998–2003, but that contributions through changes in anthropogenic emissions became dominant after 2003, amounting to 0.4–0.9 ppbv yr^{-1} (4.4–8.6 ppbv $decade^{-1}$). However, about half of the observed O_3 increase at HPO cannot be explained by the assigned anthropogenic emissions from East Asia.

The observation-vs-model discrepancy suggests existence of additional sources and/or processes which are not well implemented in our model. Since a regional model is used in the present study, long-range transport from outside Asia (i.e., intercontinental transport) is a possibility. Considering that the intercontinental O_3 transport from Europe to East Asia mainly occurs in the boundary layer over the Eurasian continent (Wild and Akimoto, 2001), and that European and North American NOx emissions have declined over the recent decades, intercontinental transport to East Asia is unlikely to have any significant influence on the decadal O_3 variation at HPO. Ordóñez et al. (2007) suggested that changes in the lowermost stratospheric O_3 could have influenced the variability of O_3 levels in the lower free troposphere over Europe during the 1990's, particularly in winter and spring. However, Terao et al. (2008) showed that although tropospheric O_3 can be highly influenced by lower stratospheric O_3 over Canada and Europe, the influence is much smaller over East Asia, and suggested that pollution transport is a dominant contributor. Thus, stratospheric influence is unlikely one of the main causes of the rapidly increasing O_3 observed at HPO during the last decade.

Anthropogenic emissions estimates still have large uncertainty. The increase of Chinese NOx emissions was found to be not quantitatively consistent with the satellite-derived NO_2 column over China (Richter et al., 2005). The growth rate of the tropospheric NO_2 column was found to be larger than the estimated changes in NOx emissions since 2000, implying that the actual NOx emissions are likely larger than the emission estimates (Akimoto et al., 2006). Stavrakou et al. (2008) derived top-down anthropogenic NOx emissions using satellite NO_2 measurements, proposing a 15% increase in the summertime surface O_3 over China for the 1997–2006 period. This is substantially larger than the modeled increase (~9%) and closer to the observed increase (~20%) at HPO. Recently Zhang et al. (2009) reported rapid growth of NOx emissions in Inner Mongolia due to newly constructed power plants. These sources were not implemented in REAS. Changes in locations of emission sources, particularly large point sources at high-altitude regions, can be effective in producing O_3 in the free troposphere.

Conclusions: implications for the future

Returning to our initial question, the answer is "Yes, we can. The trend is small in the lowermost troposphere but huge in the free troposphere, with great implications for the northern hemisphere" (Tanimoto et al., 2009). Measurements made at HPO clearly revealed a rapid increase in the springtime tropospheric O_3 during the last decade. This might be because of the capability of this remote high-altitude site to effectively detect East Asian emission changes. Many of the measurement sites associated with long-term monitoring programs are located in the boundary layer, and those in the free troposphere are still very few in number. In the near future, high-frequency O_3 measurements in the free troposphere should be expanded to better elucidate the temporal evolution of tropospheric O_3. Priorities should be to increase the number of monitoring stations at mountain sites, regular deployment of research aircraft, and implementation of LIDARs as well as O_3 balloon soundings.

Our evaluation with a regional CTM points out that there are limitations in our current understanding of and ability to quantitatively predict tropospheric O_3 trends,

particularly in response to changes in emissions over the continental Asia during the last decade. Modeling efforts with improved emissions inventories and finer spatial resolution would be of great utility in the effort to better reconcile the difference between the observed and modeled tropospheric O_3 trends over western Pacific Ocean during the past decades.

Acknowledgments

We acknowledge ADORC and WDCGG for data management. This work was supported by the Global Environment Research Fund (S-7) by the Ministry of the Environment, Japan.

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The Extratropical UTLS: Observations, Concepts and Future Directions

Community workshop at the National Center for Atmospheric Research, Boulder, Colorado ▪ October 19-22, 2009

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▪ Introduction

An international community workshop focused on the extratropical upper troposphere – lower stratosphere (UTLS) was held in Boulder, Colorado in October 2009, sponsored by the US National Science Foundation and SPARC/WCRP. The UTLS has been a key research focus of SPARC and its collaborations with IGAC, and the Boulder 2009 workshop follows two previous SPARC-sponsored workshops in Bad Tölz (2000) (Haynes and Sheperd, 2001) and Mainz (2005) (Law et al., 2005). The Boulder workshop was organized in recognition of significant ongoing progress in the UTLS research community, including a wealth of new observational data focused on the extratropical UTLS (especially chemical measurements). These include data from recent research aircraft campaigns (including SPURT and START08) and in-service aircraft (MOZAIC, CARIBIC and CONTRAIL), combined with satellite, balloon and ground-based observations. In addition many global models (including models contributing to the SPARC CCMVal effort) now include a well-resolved UTLS region, so that there are renewed efforts at comparing models with observations, and improving representation of modelled physical processes in this region. Furthermore, while the UTLS has long been recognized as a key region for understanding global ozone, it is now appreciated as highly relevant for climate variability and change, so that detailed understanding and accurate modelling of the UTLS is important to a wide scientific audience. Accordingly, the Boulder workshop aimed at an updated evaluation of the state-of-the-art for observations, modelling and process understanding for the extratropical UTLS.

The Boulder workshop was attended by over 90 scientists (see Figure 1), covering four days of invited overview talks, plus numerous contributed presentations and posters. A web site for the workshop (http://www.acd.ucar.edu/utls/workshop.shtml) includes the detailed schedule, and the archived presentations

Figure 1.
Participants attending the Boulder UTLS workshop, October 2009.

of most talks and posters. The workshop included an hour at the beginning of each day to review the highlights and discussions from the previous sessions, assembled by groups of rapporteurs. Key activities included summarizing current understanding and key uncertainties regarding the UTLS, and pinpointing future research needs.

The workshop was divided into 5 interlinked sessions. It began with a session on (1) tropopause structure and dynamics, followed by (2) the chemical composition of the UTLS, (3) UTLS transport, (4) convection and microphysics in the UTLS and finally (5) long term variability and trends. The final half-day included an overall summary session, with discussion of future proposed observations and observing needs. This summary includes references to many presentations in the workshop (referenced by lead author, without a date), while references with a date refer to published work (cited at the end).

Session 1: UTLS and tropopause dynamical structure

This session focused on various aspects of dynamical behavior of the UTLS and tropopause region, including new observations and modeling studies. Much current work is focused on quantifying specific dynamical aspects of the UTLS, including the tropopause inversion layer (TIL), double tropopauses (DT) and the extratropical tropopause transition layer (ExTL), with overall goals of 1) understanding these structures in the context of synoptic meteorological variability, 2) the relationships of dynamical and chemical transitions across the tropopause, and 3) their seasonal and latitudinal behavior. In particular, the global structure of static stability, structure of the TIL, connections between TIL and ExTL, global scale occurrence of the DT and relation to dynamical intrusions, and possible relationships between DT and TIL are topics of current investigation. The emerging picture is that there are connections among dynamical structures (TIL, ExTL, DT, etc.) and links between dynamical and chemical behavior, although the responsible mechanisms continue to be identified.

One focus in this workshop was the observational characterization of the TIL, using high vertical resolution radiosondes, aircraft measurements and GPS radio occultation data. GPS data provide characterization of the global behavior of the TIL, with new results highlighting the large-scale seasonally-varying structure of the tropical TIL, and polar TIL variability linked to stratospheric warming events (Grise et al., 2010). Comparisons with vertical profiles from MOZAIC aircraft data reveal good agreement with GPS measurements, and furthermore highlight sharp discontinuities in chemical tracers (O_3, CO, and H_2O) linked to the thermal tropopause (Schmidt et al.). The high density of COSMIC GPS

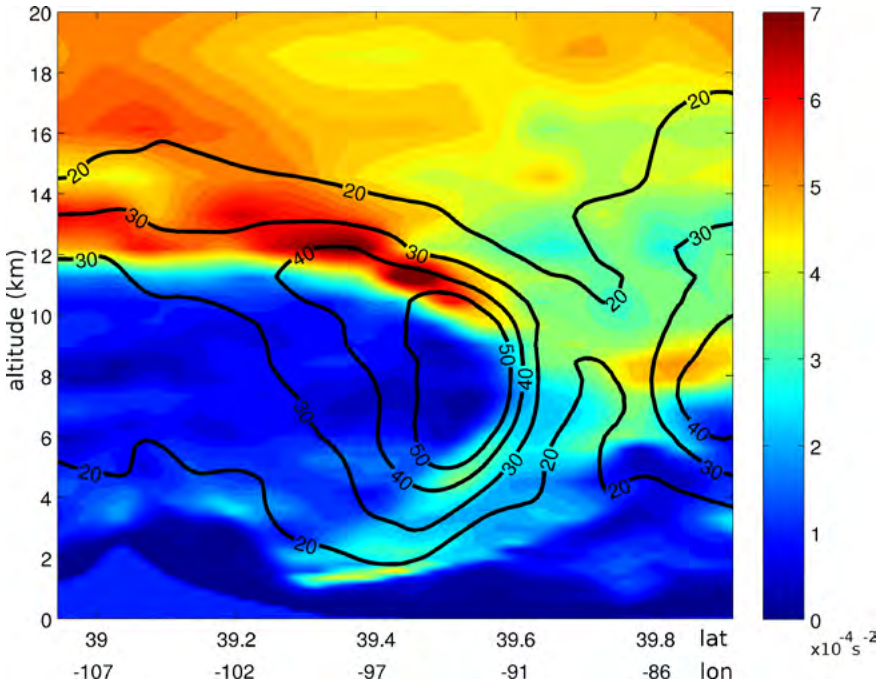

Figure 2. Cross section of static stability (Brunt Vaisala frequency squared, colors) and wind (contours, m/s), showing high resolution structure near a tropopause fold. [from Tandon et al.]

data also allows analysis of synoptic TIL behavior, revealing characteristic links to jet structure (Figure 2, Tandon et al). The strongest climatological TIL occurs during polar summer, with a high degree of symmetry between hemispheres; Randel and Wu proposed this behavior is linked to the strong radiative effects of water vapor near the tropopause, which exhibits a strong summertime polar maximum. One novel analysis of tropopause structure using a curve-fitting algorithm to define transition layer depth (Homeyer et al., 2010) categorized the midlatitude tropopause as a function of transition depth. The presence of a TIL is associated with a narrow transition layer, which dominates the observed population, while no TIL is evident in the large transition case. Other novel uses of satellite data included evaluation of the space-time structure of DT occurrence frequency using high vertical resolution HIRDLS ozone and temperature measurements (Phillips and Gille), and analysis of global UTLS gravity wave characteristics using the dense sampling of COSMIC GPS data (Wang and Alexander).

Recent modeling work has focused on the evaluation of the ExTL in comprehensive global models (e.g. the CCMVal assessment, see summary below), in addition to more idealized studies focused on specific process understanding. Wang and Polvani have used idealized baroclinic wave cycle studies to simulate the conditions of DT formation, finding that DT occurrence is favored when the initial conditions include a strong TIL. However, their idealized model results produce DT for strong cyclonic circulations and cut-off lows, rather than for tropospheric intrusions from low latitudes, as often observed. Understanding dynamical and radiative contributions to TIL formation and maintenance continues to be an active topic. Birner used a global circulation model to quantify forcing of the global tropopause from the stratospheric residual mean circulation and radiative equilibrium calculations. The relationship between the TIL and the chemical mixing layer was examined by Kunz et al., (2009) using SPURT observations of UTLS ozone and water vapor and Fixed Dynamical Heating (FDH) calculations. These results demonstrated that enhanced water vapor associated with the mixing layer was a primary cause of high stability above the tropopause (strong TIL), while ozone had relatively minor influence. This finding is consistent with an evolving understanding of a key role for water vapor near the tropopause for formation and maintenance of the TIL.

Summary of CCMVal assessment of the extratropical UTLS

A comprehensive assessment of coupled Chemistry-Climate Models (CCMs) has recently been completed by the CCM Validation Project (CCMVal), and one focused activity involved evaluation of the UTLS behaviour in models. Results are described in detail in SPARC (2010), Gettelman et al. (2010) and Hegglin et al. (2010). Broadly, CCMs perform reasonably well in the UTLS with respect to the sharp dynamic and tracer gradients, given their coarse resolution. Models are able to simulate a TIL of reasonable magnitude and have distinct tracer gradients and tracer-tracer correlations across the tropopause. Deficiencies in detailed vertical structure arise from coarse resolution, along with possible problems with transport across the sub-tropical jet. Models simulate increases in tropopause height and increases in ozone in the UTLS in the 21st century future scenarios. The tropopause structure is strongly affected by Antarctic ozone depletion and recovery in the Southern Hemisphere, but trends also are predicted in the northern hemisphere. A consensus is that further global-scale observations with high vertical resolution are necessary to better evaluate models. Also, more complete representations of tropospheric chemistry will be valuable for evaluating detailed UTLS transport (which is often based on tropospheric tracers such as CO or shorter-lived hydrocarbons).

Session 2: structure and chemical composition of the ExTL

This session aimed to define and characterize the ExTL and to further explain its role in the chemical, physical and dynamical structure of the extratropical UTLS. Given the complex thermodynamic and chemical structure in this region, there are many different metrics used to define the ExTL. This situation is highlighted by the fact that different vertical coordinate systems, tracer-tracer correlations, and definitions of the tropopause (thermal, dynamical, chemical) are employed in different studies. The most commonly used coordinate system remains "delta Z or delta theta from tropopause", defined using the thermal tropopause, along with O_3-CO correlations. Figure 3 (from Hegglin et al., 2009) illustrates the use of these metrics to quantify the thickness of the ExTL mixing layer. It was noted that results based on other tracers can suggest different position and thickness of this ExTL related to the tropopause altitude, so that details depend on the definition of the tropopause itself (thermal or dynamical), and on the tracer-tracer correlation used. This situation can be partly understood by noting that different tracers are associated with different life times (in terms of sources and sinks in the UTLS), along with differences in transport pathways and time scales, and thus a different 'depth' of the ExTL is expected based on diagnoses with different tracers. Also, the thickness of the mixing layer is broader at high compared to low latitudes (Pisso et al.). These details emphasize the difficulty in bringing an integrated view to the broad picture of the ExTL. It is recognized that the transition layer extends on both sides of the tropopause (with a thickness of approximately 2 km on either side). Tracer structure has also been categorized by spatial location with respect to the jet position, showing systematic differences for the poleward (cyclonic) and equatorward (anticyclonic) sides of the jet (Manney et

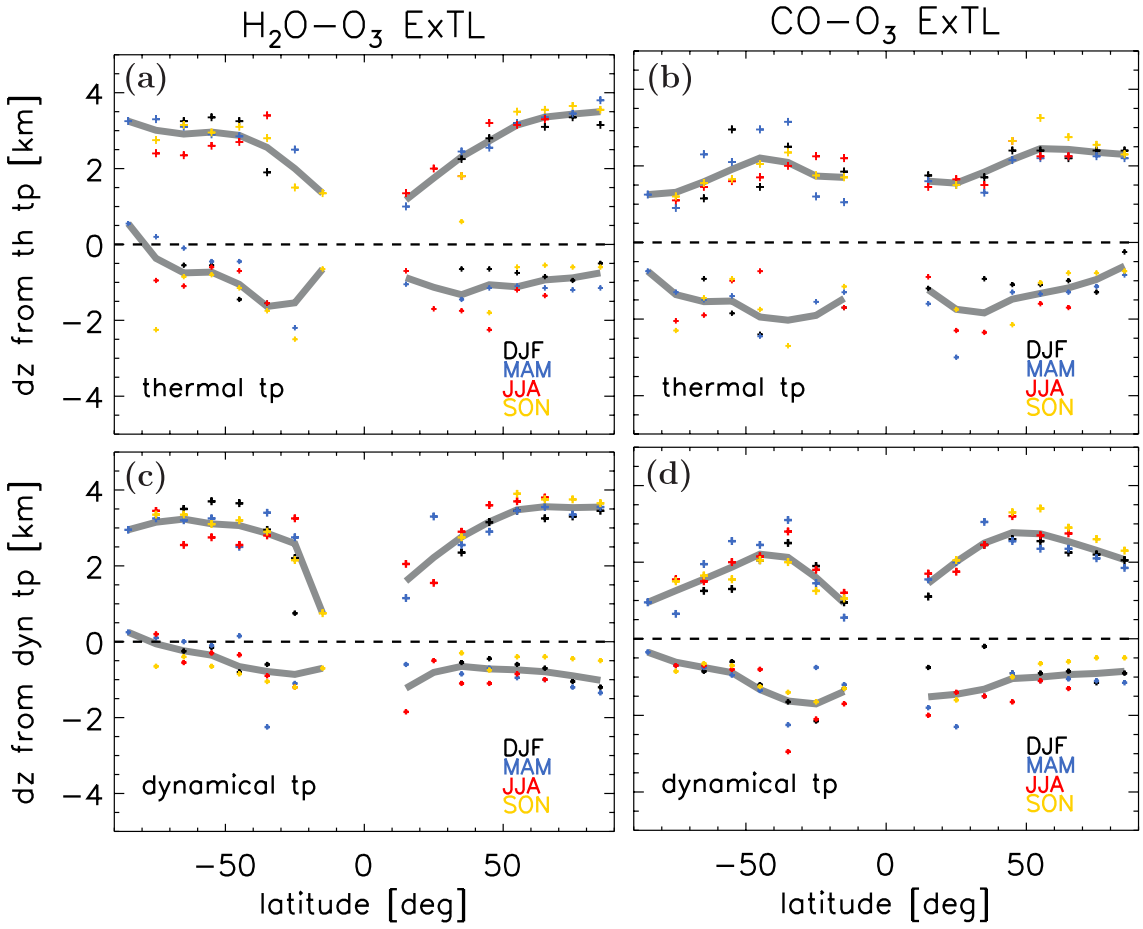

Figure 3. Depth of the chemical mixing layer calculated from ACE-FTS data. Results are shown for calculations based on (left) H_2O-O_3 and (right) CO-O_3 correlations, using vertical coordinates with respect to the thermal (top) and dynamical (PV=2) tropopause (bottom). [from Hegglin et al.]

al.), or for the east vs. west side of cyclones (Brioude et al.). A newer concept is that the chemical transition layer marks a change in transport time from the lower troposphere to the lowermost stratosphere (LMS). Another important aspect is that a strong seasonality exists above the transition layer with "young" air in summer and autumn and rapid "flushing" from late spring to summer (Mackenzie et al.). Also, there is evidence for a strong coupling from low latitudes (the TTL and the tropical lower stratosphere) to the extratropics.

Other discussions in this session focused on the use of equivalent latitude coordinates for the UTLS (Pan et al.) and new ideas on defining the tropopause in 3D models (Neu et al.). Pan et al. suggested that while equivalent latitude – theta coordinates are appropriate for describing stratospheric processes, there are limitations for application near the tropopause (because of the non-conservation of PV in this region). Moreover, equivalent latitude coordinates are typically derived from meteorological analyses that have limited resolution compared to other data (aircraft or balloon). Neu et al. pointed out the problem that thermal, dynamical, or ozone tropopause definitions cannot simply be used for comparison with numerical simulations. It was proposed to use an "E90" tracer (an artificial tracer with a 90-day time scale) which defines the tropopause as a mixing barrier to better identify the tropopause height in CCMs. This tracer leads to good comparisons of ozone in the extratropics

Session 3: UTLS transport and stratosphere-troposphere exchange

The workshop devoted a day to discussion of UTLS transport and Stratosphere-Troposphere Exchange (STE). Key discussions centered around several themes: (1) coupled chemical-dynamical structure of the extratropical tropopause, (2) aircraft observations of tracer correlations and mixing regions around intrusions and jet structures, and (3) the role of gravity waves and small scale processes (including deep convection).

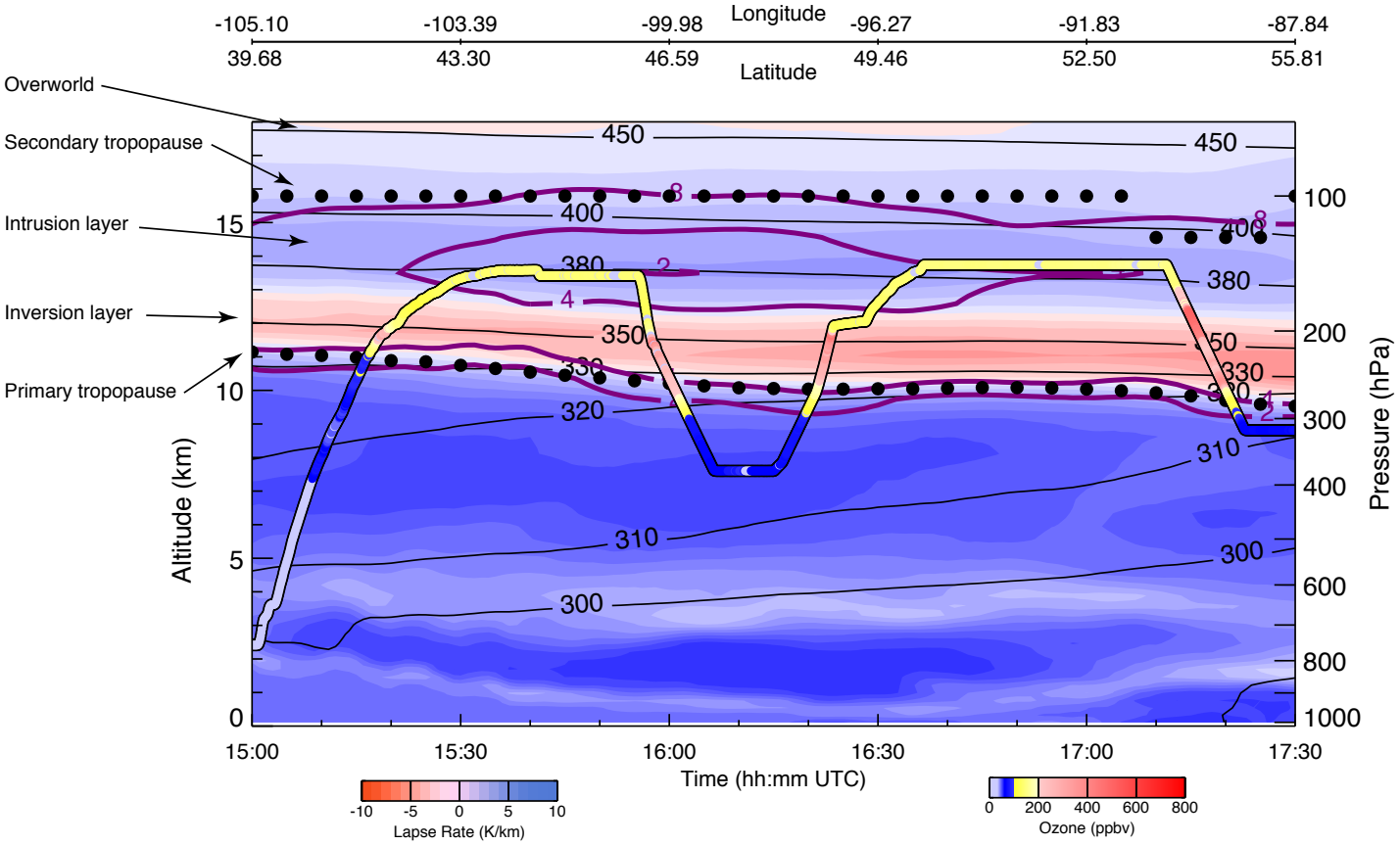

Figure 4. Vertical cross section along track from START08 flight 01, showing static stability, in situ ozone, PV (purple contours), and theta (black contours). The stable layer in the lower stratosphere is sandwiched between the troposphere and a tropospheric intrusion from the tropics.

The broad-scale structure of the extratropical tropopause and its behavior as a transport barrier are fundamental aspects of the interaction of baroclinic eddies with the background jet in the troposphere, and this behavior can be simulated in simple dynamical models (Haynes et al., 2001; Greenslade and Haynes, 2008). Theories for the existence, height and maintenance of the extratropical tropopause are mature, but not fully unified, and the importance of small-scale processes (gravity waves and convection) is still unclear. More recent work has highlighted the possible importance of moist processes (Frierson, 2008).

A substantial amount of effort has focused on analysis of UTLS chemical observations from recent field campaigns, with efforts to isolate transport pathways and identify mixing processes. Observations from flights during START08 (Pan et al.) were used to highlight the detailed structure of tropospheric intrusions, associated with the existence of air with tropospheric characteristics above the extratropical tropopause (Figure 4); this behavior is linked to double tropopauses, potential vorticity gradients and ozone lamina. Back trajectory calculations for the air in the intrusion layer show that this air mass was transported poleward above the subtropical jet (Bowman et al.). These features are reasonably well simulated in chemical transport models nudged with high vertical resolution assimilated winds (Stone et al.). Tilmes et al. (2010) have developed a new seasonal climatology of tracer observations from many research aircraft campaigns (Figure 5), and further separate observations according to tropics, subtropics and polar latitudes (based on tropopause altitude). Such data provide critical comparisons for chemical structure of the UTLS in numerical models.

The use of tracer-tracer correlations from in-situ data and simulations is effective at identifying air mass origins and transport pathways, and Vogel et al. and Konopka and Pan have extended this concept using Lagrangian (CLaMS) model calculations to quantify the origins of air and the degree of mixing within various regions (focusing on both tropospheric and stratospheric intrusions). Konopka and Pan furthermore quantify the time history of mixing in their calculations, highlighting regions where the

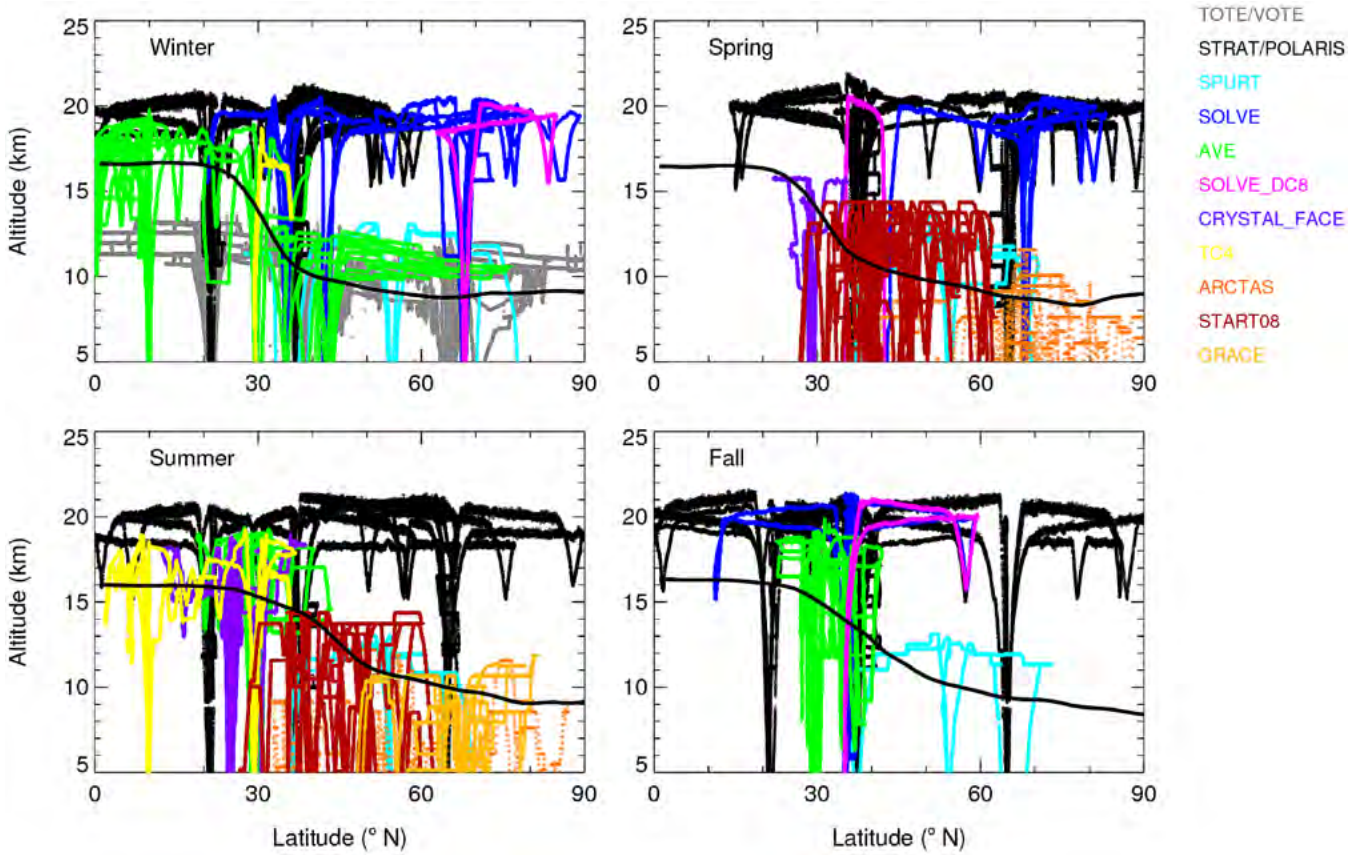

Figure 5. Seasonal UTLS sampling over the Northern Hemisphere from numerous research aircraft campaigns. [from Tilmes et al.]

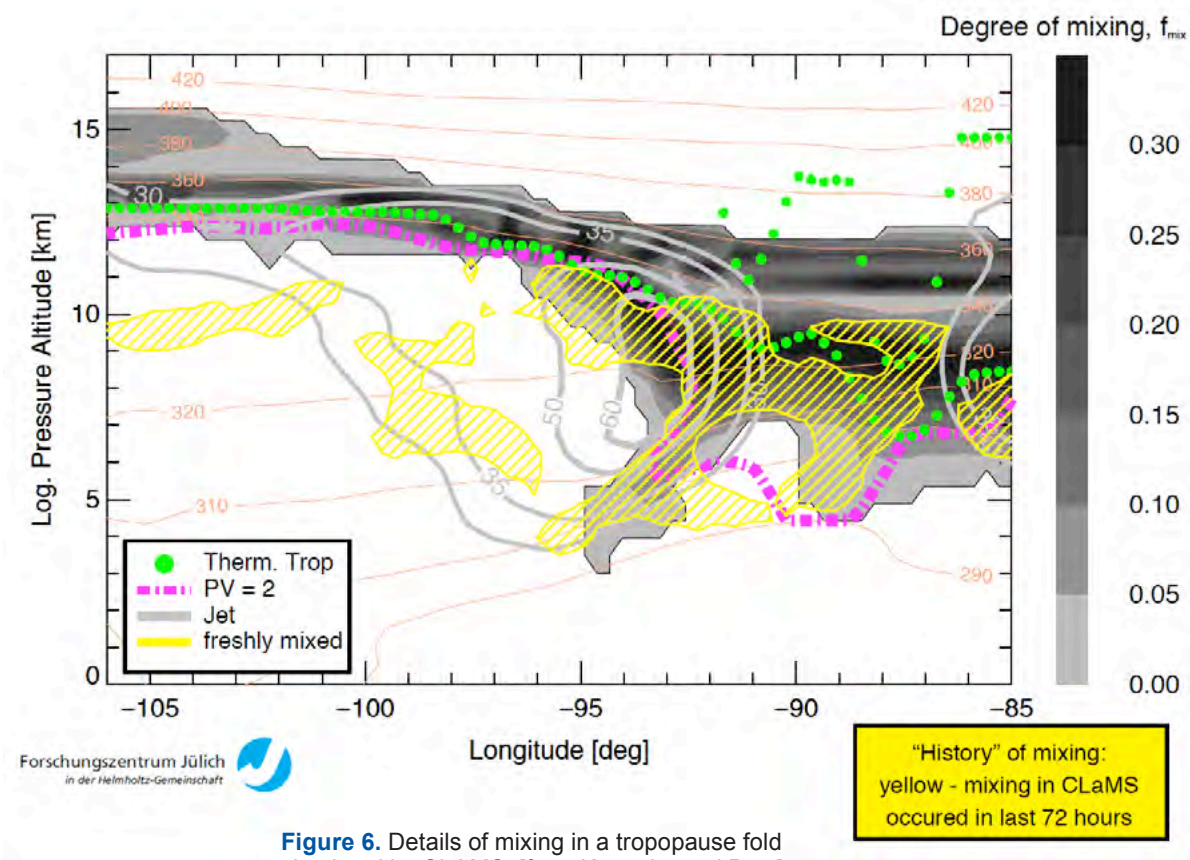

Figure 6. Details of mixing in a tropopause fold simulated by CLAMS. [from Kanopka and Pan.]

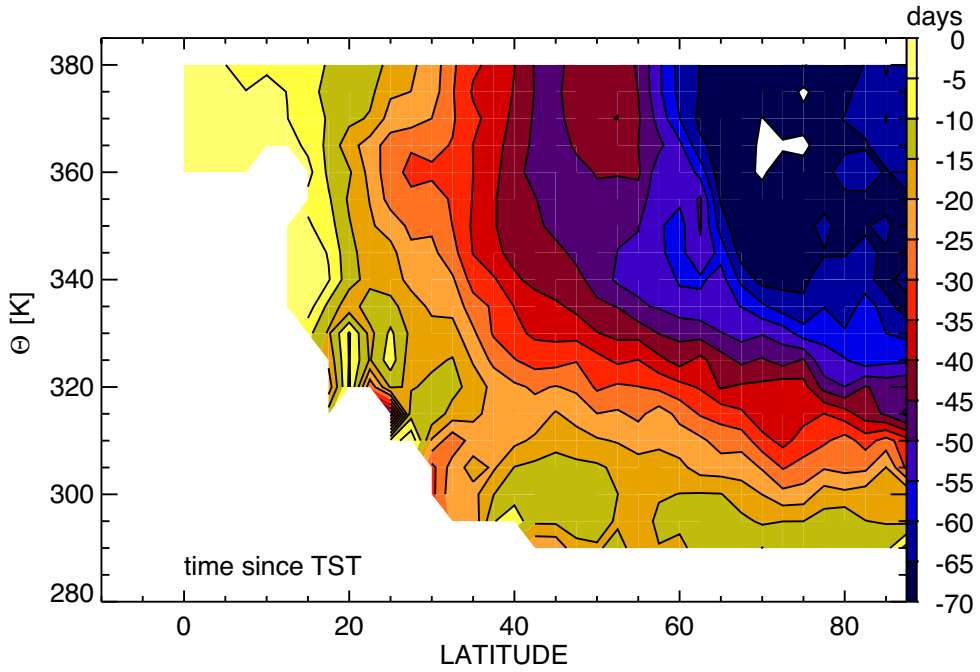

Figure 7. Trajectory estimates of average transport time (in hours) from the tropopause into the lower stratosphere. [from Hoor and Wernli]

mixing was relatively fresh (< 72 hours) or aged (Figure 6). Hoor and Wernli used a large ensemble of trajectories to quantify the distributions of transit times (and minimum temperatures) for parcels crossing the tropopause into the extratropical stratosphere (Figure 7), and these results can explain some of the observed differences in the mixing-layer structure of CO vs. water vapor. Trajectory studies by James and Legras (including diffusive effects) demonstrated the importance of two-way transport between the TTL and the extratropical lower stratosphere.

There were also discussions of several innovative observational data sets and analysis techniques. The use of tracers with multiple lifetimes with sensitivity to different altitudes (such as N_2O and CFCs) is effective for estimating transport pathways in the stratospheric overworld and 'lifetimes' of air in the UTLS between 1 month and up to several years (Ray et al.). Tarasick et al. demonstrated the use of radar observations at high latitudes to identify the tropopause and the frequent occurrence of stratospheric intrusions (associated with ozone transport to the upper troposphere). Mullendore et al. also highlighted the novel use of radar reflectivity as a proxy for convective detrainment in the UTLS. The important role of the lower levels of the polar vortex (the so-called sub-vortex) for transport to the midlatitude UTLS during late winter and spring was demonstrated using MLS and ACE-FTS satellite observations by Santee et al. MLS data were also used to document the occurrence of smoke plumes in the lower stratosphere, which originated with the large Australian wildfires during February 2009 (Massie). These remarkable features are observed at altitudes up to ~20 km, and transport to the stratosphere may be explained as a combination of upward transport within frontal systems combined with in-situ radiative heating of the smoke in the UTLS.

Results were presented from several novel high resolution modeling experiments focused on the UTLS. Miyazaki et al. analyzed tropopause structure in a GCM with 300 m vertical resolution (and ~50 km horizontal resolution), demonstrating that strong tracer and PV gradients near the tropopause result from the combined effects of transport, mixing and radiative effects, and they further quantified the importance of resolved small scales (gravity waves) for the mixing in this region. Mizuta and Yoshimura used simulations from an ultra-high horizontal resolution (20 km) global model to investigate the sensitivity of tropopause structure and STE to model resolution. A novel simulation of thunderstorm effects on the UTLS during the North American monsoon, using an extremely high resolution (4 km) WRF-chem model (Barth et al.), focused on the importance of convection on upper tropospheric ozone (including direct convective transport of ozone and ozone precursors, plus lightning-generated NOx).

The importance of gravity waves and deep convection on the extratropical UTLS were topics of several presentations. These processes may become dominant during 'mixing events' associated with intrusions, folds and fronts, when the concept of the tropopause as a transport barrier breaks down. Several presentations focused on gravity waves, generated

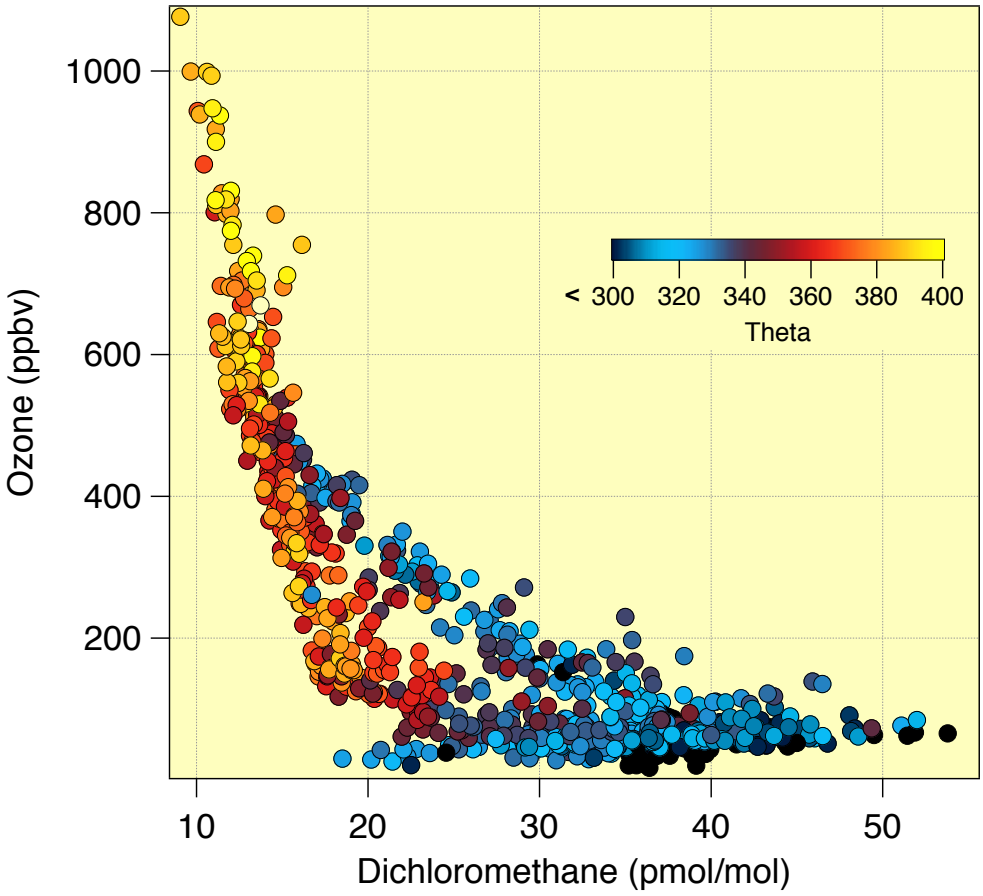

Figure 8. Scatter plot of ozone vs. dichloromethane from whole air sample measurements in START08, color coded by potential temperature of the observations. [from Atlas et al.]

from topography, convection or midlatitude baroclinic adjustment processes. UTLS gravity waves observed during the START08 experiment (associated with jet-front adjustment processes) show reasonable agreement with simulations from a 5-km WRF simulation (Meng et al.). Also, aircraft observations of UTLS mountain waves (Moustaoui et al.) show complex effects on tracers (CO and ozone), which can be understood in the context of the coupling of large and small-scale waves. There were discussions that large scale features ('stirring') are well reproduced in current forecast systems (at 25 km resolution or so), and even to some extent in nudged global simulations at 100km. However, several presentations highlighted that the mixing of chemical constituents down to the molecular level (where chemistry operates) is not being well represented, and this topic is not well understood.

Session 4: Chemical and microphysical distributions

A key theme of this session was the important role of tracers with different lifetimes for deriving quantitative information on the structure of the tropopause region. Correlations of species with different photochemical lifetimes (such as various non-methane hydrocarbons, NMHC's) provides quantative information on the temporal range of tropospheric influence and mixing into the lowermost stratosphere. Examples were presented using correlations of NMHC's during START 08 (Figure 8, Atlas et al.) focusing on the transport and mixing related to tropospheric as well as stratospheric intrusions. The potential of these shorter lived compounds in a more climatological context was demonstrated using acetone from regular in-flight measurements during CARIBIC, which showed an important tropospheric influence extending deep into the lowermost stratosphere from summer (June) to late autumn (Figure 9, Zahn et al.). Details of large-scale transport in the tropopause region were also

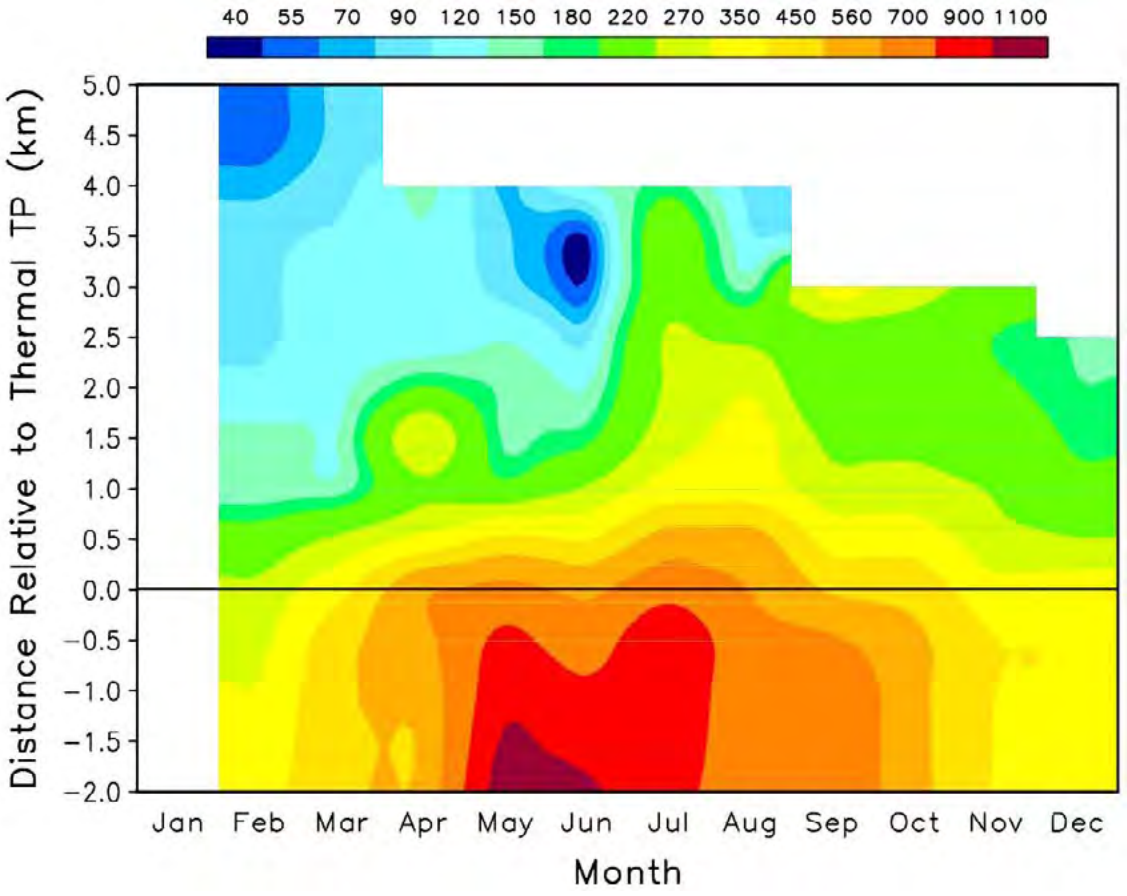

Figure 9. Monthly climatology of acetone mixing ratio vs. height in tropopause coordinates from CARIBIC measurements. [from Zahn et al.]

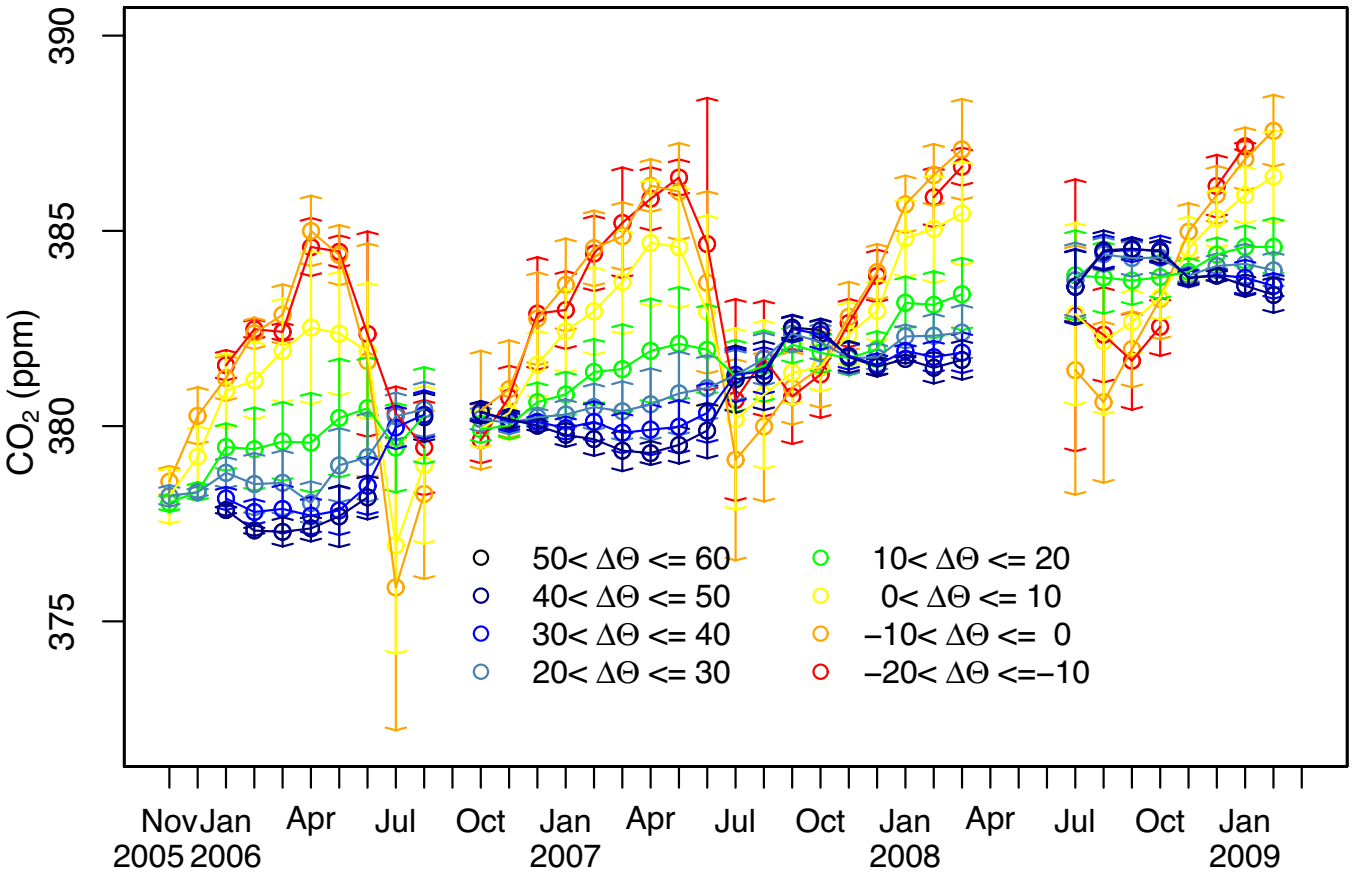

Figure 10. Time series of CO_2 at different levels with respect to the tropopause from CONTRAIL measurements. [from Sawa et al.]. Note the strong seasonal cycle that varies as a function of distance from the tropopause.

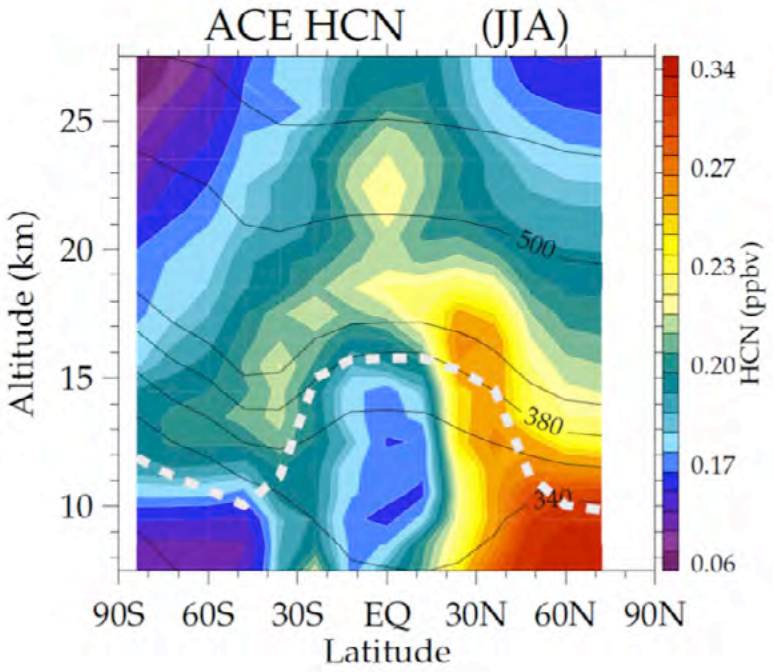

Figure 11. Meridional cross section of HCN mixing ratio from ACE-FTS satellite observations during NH summer, showing transport into the stratosphere through the Asian monsoon. [from Park et al.]

quantified using a long record of CO_2 observations from the Japanese CONTRAIL project (Figure 10, Sawa et al.), focusing on propagation of the seasonal CO_2 cycle as a function of distance to the tropopause. The propagation of the CO_2 seasonal signal highlights the importance of subtropical / extratropical coupling at the location of the subtropical tropopause.Several presentations highlighted the importance of the Asian monsoon anticyclone for transport of water vapor and other species into the LMS during NH summer. A key point is that this transport pathway can bypass the tropical tropopause, and the air in this region has distinct source regions from the deeper tropics (namely highly polluted air originating over Asia, India and Indonesia). Evidence for the importance of this transport pathway comes from recent satellite observations of CO and HCN from MLS and ACE-FTS (Figure 11, Park et al). There have been relatively few in situ observations over this region (especially for trace constituents), but some exploratory observations of water vapor and ozone over Kunming (25°N, 102°E) were presented by Bian et al., showing novel and complex behavior. While the Asian monsoon shows clear influence on the lower stratosphere, it is as yet unclear to what extent monsoon circulations over other continents play similar important roles.

One further theme for this session focused on the behavior of UTLS clouds and water vapor. High resolution balloon water vapor observations in middle and high latitudes often reveal complex laminated structure, linked to vortex dynamics or transport from the TTL (Khaykin et al.) These observations highlight that an ExTL definition on the basis of water vapor may be confused by polar or subtropical processes, rather than by exchange across the extratropical tropopause. In addition there is evidence that cirrus clouds above the extratropical tropopause may also play a role in the water vapor budget and variability (Dessler, 2009). New aircraft remote sensing measurements from the CRISTA-NF instrument demonstrate high vertical and horizontal resolution measurements of clouds and trace gases in the UTLS (Reise et al.), pointing to new possibilities for untangling mechanisms and sources of variability on synoptic scales.

Session 5: Long-term variability and trends

The final session focused on long-term variability and trends in dynamical and chemical behavior of the UTLS. An overview of long-term measurements from the MOZAIC program (Thouret et al) highlighted the availability of data from over 32,000 flights (beginning in 1994), with novel applications including quantifying quasi-global UTLS climatologies of ozone, water vapor and CO (Figure 12), and sampling regions previously void of data (such as over Africa and China). While the long-term MOZAIC measurements are useful for studying some aspects of interannual variability (such as ENSO effects), their application to long-term trends is limited by sampling variability. Long-term changes in UTLS ozone have been examined based on aircraft measurements from the GASP program (1975-1979) and MOZAIC (1994-2001), combined with ozonesonde measurements (Staehelin et al.). There are substantial uncertainties in comparing data from different aircraft instruments (over different decades) when searching for relatively small ozone changes. Such uncertainties were confirmed for these data by comparisons to ozonesonde measurements, so that decadal trends cannot be confidently estimated from these aircraft data at present. A recent increase in stratospheric aerosols has been documented from lidar observations by Hoffman et al (2009), and postulated to be linked to growing sulfur emissions over China. Solomon et al. documented a similar increase using SAGE satellite observations (which ended in 2005), and presented modeling results that highlight the importance of transport to the stratosphere through the Asian monsoon circulation.

Satellite data suggest a decrease in tropical lower stratospheric ozone over the last several decades, and similar behavior is found in many CCMval model simulations. This modeled behavior was explored in detail by Lamarque and Solomon, who demonstrated the key mechanism was an increase in tropical upwelling, linked primarily to increasing greenhouse gases (and not to ozone depleting chemicals). It was noted that past changes in tropical upwelling may be reflected in low vs. high latitude temperature variations and trends in the stratosphere, as discussed using historical MSU and SSU data (Young et al.). A novel diagnostic for long-term circulation changes in the extratopical lower stratosphere based on N_2O-ozone correlations was presented by Boenisch et al., who noted that this region may be especially sensitive to the so-called 'lower branch' of the Brewer-Dobson circulation.

A topic of further interest was changes in the global tropopause height and possible widening of the tropics in observations and model simulations. A number of different metrics for measuring tropical width were discussed by Davis and Rosenlof, who noted their sensitivity and differences based on different observational (reanalysis) data sets. Anel et al. explored the use of PV gradients to quantify tropical widening. Gettelman et al. extended this theme by quantifying tropical width in past and future global model simulations from CCMVal, noting that for many quantities trends are difficult to quantify in the face of large natural variability.

Workshop summary: Lessons learned and way forward

The workshop focused much discussion on several key ExUTLS concepts and processes: the definition of the extratropical tropopause, the double tropopause, the TIL and the ExTL. It also focused on the role of the jet streams in transport and mixing,

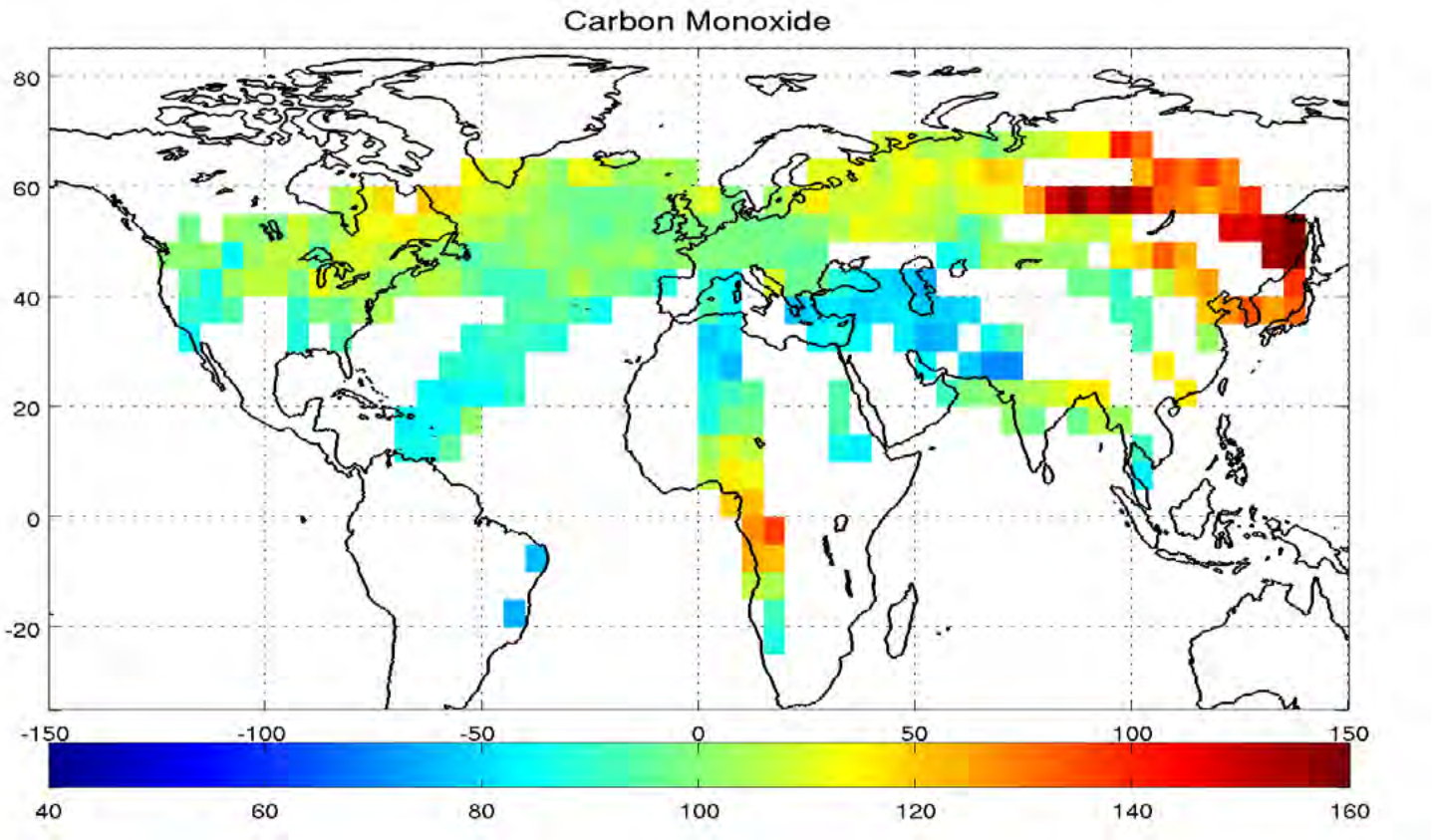

Figure 12. Climatology of NH summer carbon monoxide mixing ratio (ppbv) near 10 km from MOZAIC measurements. [from Thouret et al.]

observations of chemical tracers and 'mixed' air, and trace gas budgets that can delineate transport pathways. The emerging trend is that these elements not only need to be described individually, but it is also key to understand their mutual connections and interactions. The relative roles of different processes are not well understood.

ExTL: Since the last major UTLS workshops (Bad Tolz, 2000 and Mainz, 2005), much progress has been made in the observational characteristics of the ExTL. The synoptic scale structure of the ExTL was targeted by research flights in the SPURT and START08 field campaigns (Engel et al, 2006; Pan et al., 2010). Trace gas budgets and transport pathways were quantified. Satellite data analyses provided global statistical behavior of the ExTL (e.g. Hegglin et al. 2009). The statistical behavior using different tracers has shown variable ExTL depth (CARIBIC, Zahn et al.), reflecting the underlying dynamical structure and the time scale involved in the ExTL formation.

TIL: Significant progress has been made in characterizing the global spatial temporal structure of the TIL using global GPS satellite data (Grise et al., 2010). Increasing evidence supports the significance of water vapor radiative effect in the formation of the TIL (Kunz et al., Randel and Wu). This particuar issue connects the formation of the TIL with the existence of the ExTL. In particular, enhaced water vapor in the vicinity of the tropopause, associated with the chemical mixed layer, appears to be one key mechanism for formation and maintenance of the TIL. There are also indications, using ozone data, that the strength of the TIL in turn enhances the barrier effect of the tropopause and produces a stronger chemical discontinuity between UT and LS (Tandon et al.).

The definition of the extratropical tropopause has often been the origin of diverse views and approaches in quantifying STE. However, a more updated perspective may be that for cases where significant differences between the dynamical and thermal tropopauses exist, this is an indication of the lack of a sharp boundary between the troposphere and stratosphere (Pan et al., 2007). The occurrence of the double tropopause is now recognized to be often associated with transport and intrusions from the subtropics (Pan et al., 2009). The mechanisms which control this process and its seasonality are far from understood, and somewhat different perspectives are given by different studies. For example, recent observations during the START08 campaign found a significant influence from these events in the lowermost stratosphere during Spring. This is consistent with the GPS analyses showing winter/spring as the high season for the double tropopause occurrence in the NH (Randel et al., 2007). On the other hand, trajectory based studies (e.g. Berthet et al., 2007) suggest ventilation of the layer above the subtropical jet is stronger in summer than winter. A study using HIRDLS data and the equivalent length approach found similar conclusion (Gille et al.). Questions also remain whether this process is a middle world process, represented as isentropic mixing between the upper troposphere and LMS, or it is part of a stratospheric process and should be considered as part of the lower branch of the B-D circulation.

The double tropopause structure also highlights the role of the jet stream in transport. The core of the sub-tropical jet forms a barrier to meridional transport, which is more vigorous below and above the jet core, and has a strong regional and seasonal component tied to the jet. The transport above the jet core is observed in aircraft and satellite data, and is consistent with the double tropopause structure. Trace gas budgets (Hoor 2005) and time scales for transport in the LMS were quantified (Boenisch et al., 2009). During winter and spring, the LMS above the ExTL (more than ~2km above the tropopause) has a strong contribution from the overworld, and a tropospheric signature from the preceding autumn. From summer to autumn the UTLS is rapidly flushed with young tropospheric air. The deep monsoon anticyclones, particularly the Asian summer monsoon anticyclone, are also persistent UTLS features that strongly regulate transport, including strong meridional mixing on the east and west sides of the anticyclone (Konopka et al, 2009), and vertical transport deep into the stratosphere (Randel et al., 2010).

The workshop discussions further highlighted the wealth of available datasets for providing new insights into the ExUTLS processes. Aircraft data from research campaigns (e.g. SPURT and START08) provide a host of coordinated observations for studies relating the chemical behavior and meteorological/dynamical structure of the ExUTLS. The in-service flight data (MOZIAC, CARIBIC and CONTRAIL) will continue to provide a dense sampling of UTLS chemical distributions. There is increased appreciation for the high vertical resolution and accuracy of GPS temperature data. Also, satellite data continue to provide key information on large-scale chemical structure, particularly for regions where there are few aircraft measurements (e.g. the Asian monsoon anticyclone, and much of the tropics and SH).

Finally, there is continuing development and application of high resolution global and regional models that focus on simulation of the UTLS region, including fully coupled chemistry and climate effects. The integration of such models with current and planned observations promises significant progress for UTLS science over the next decade.

This article first appeared in the WCRP-SPARC newsletter
http://www.atmosp.physics.utoronto.ca/SPARC/index.html

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GEOmon - Global Earth Observation and Monitoring of the Atmosphere

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Introduction

GEOmon (Global Earth Observation and Monitoring of the Atmosphere) is an Integrated Project of the 6th European Commission Framework Programme, involving 38 participating institutes. GEOmon (geomon.eu) aims to sustain and analyse European ground-based observations of atmospheric composition, complementary to satellite measurements, in order to quantify and understand the continental-scale long-term changes. The project is an opportunity to build a future integrated pan-European atmospheric observing system focussing on systematic observations of long-lived greenhouse gases, reactive gases, aerosols, and stratospheric ozone. This will subsequently facilitate a European contribution to GEOSS (Global Earth Observation System of Systems), in keeping with the European strategy for environmental monitoring of atmospheric composition observations.

Scientific Objectives

The overarching goal of GEOmon is the integration of European ground-based, satellite and modelled atmospheric compositional data to understand continental-scale long-term changes. This is being achieved by:

- **Quantifying and understanding the ongoing changes of the atmospheric composition in Europe,** by providing and sustaining quality assured, harmonised long-term ground-based observations of the composition of the European boundary layer. The development of new near real time data products is also supported, to enable access to data within a few days of measurements.
- **Integrating satellite and ground-based observations** through the development of methodologies that unify ground-based and airborne data with satellite observations. The project additionally provides quality assured chemical composition measurements of the global free troposphere from instrumented passenger aircraft programs.
- **Quantifying global trends and uncertainties related to climate change** through the analysis of spatial and temporal distributions of greenhouse gases, aerosols, chemical pollutants and stratospheric ozone from both ground-based measurements and satellite observations. A comparison between observed trends and global models is being implemented. Ultimately, a GEOmon Data Centre has been established to provide comprehensive and easy access to existing key European atmospheric composition data and data products, to assist both the scientific and policy communities.

The GEOmon project comprises six different areas of focus:

1. Greenhouse gases and Global Warming

GEOmon investigates greenhouse gases in Europe (primarily CO_2 and CH_4), integrating atmospheric models, ground-based data and remote sensing observations to constrain European sources and climate warming impacts of these species. Through this, GEOmon supports Europe's network of ground-based high accuracy *in-situ* measurement programs for CO_2 and CH_4, including inter-comparison studies. Additionally, the development and implementation of near-real-time data processing at a number of ground-based stations speeds up data access. Ultimately, time series records from individual stations can be used together as a pan-European database to support continental-scale modelling studies.

In a parallel investigation, in-situ monitoring of the free troposphere by the CARIBIC-adapted passenger aircraft measures global distributions of CO_2 and CH_4. Support of these flights facilitates harmonisation of the observations with existing aircraft datasets, enabling 4D climatologies of these trace gases, evaluation of satellite observations and a comparison with modelled results. In addition, a new European network of solar (near-infrared) Fourier Transform Infrared spectrometers (solar FTIRs) has been established providing essential ground-truthing for satellite data and aiding the integration of satellite information into the existing European measurement systems.

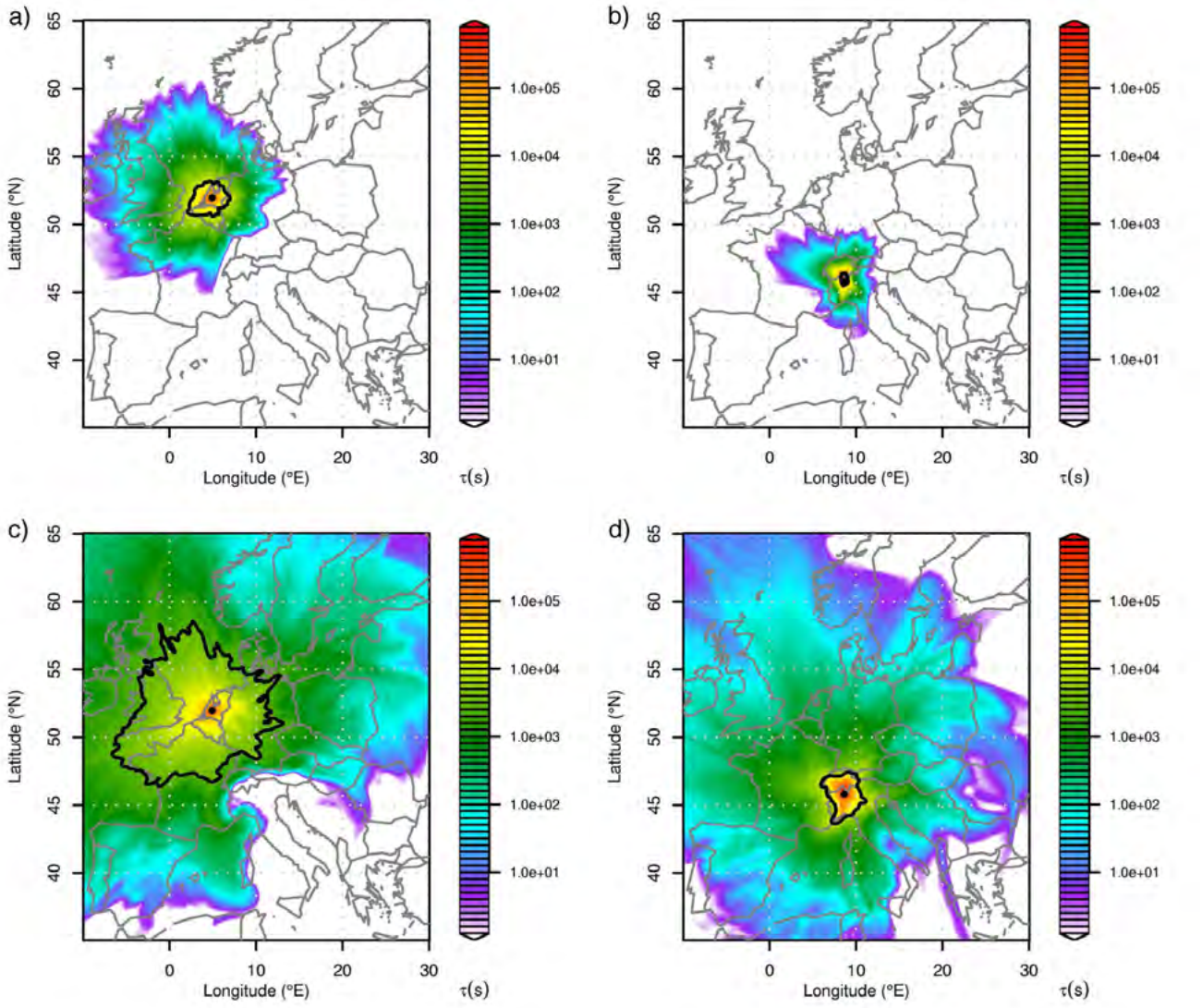

Figure 1. Total annual surface residence times (footprints) given in units seconds (color scale) and boundary of catchment area (thick black line) for the sites Cabauw (NL11, a, c) and Ispra (IT04, b, d) and two integration intervals, 12 h (a, b) and 48 h (c, d). [*from Henne et al., 2010*].

2. *Reactive gases, pollutants and climate*

GEOmon has assessed quality assured data sets of European tropospheric reactive trace gases over the time period 1996-2006 (inclusive) to be made available soon in a harmonised format via the GEOmon data centre (GEOmon.nilu.no). Data includes existing O_3, CO and NO_2 measurements from ground-based stations belonging to regional, national and European air quality networks e.g. WMO/GAW, EMEP, EEA, AURN. The characterisation of representativeness of these sites has recently been demonstrated by Henne et al. (2010) based on population (an emission proxy) and the deposition influences within each sites' catchment area. Examples of catchment areas for two sites is given i**n** Figure 1.

Using this harmonised data set over a constant time-period, trend calculation using consistent data analysis techniques has enabled the characterisation and quantification of seasonal and annual surface background European trace gas trends. This has also lead to comparative analysis with modelling studies and emissions inventories.

In addition, a network of existing ground-based remote sensing stations (e.g. FTS, DOAS, LIDAR) has been sustained and optimised in order to validate tropospheric satellite observations of trace gases, complimentary to in-situ ground based measurements.

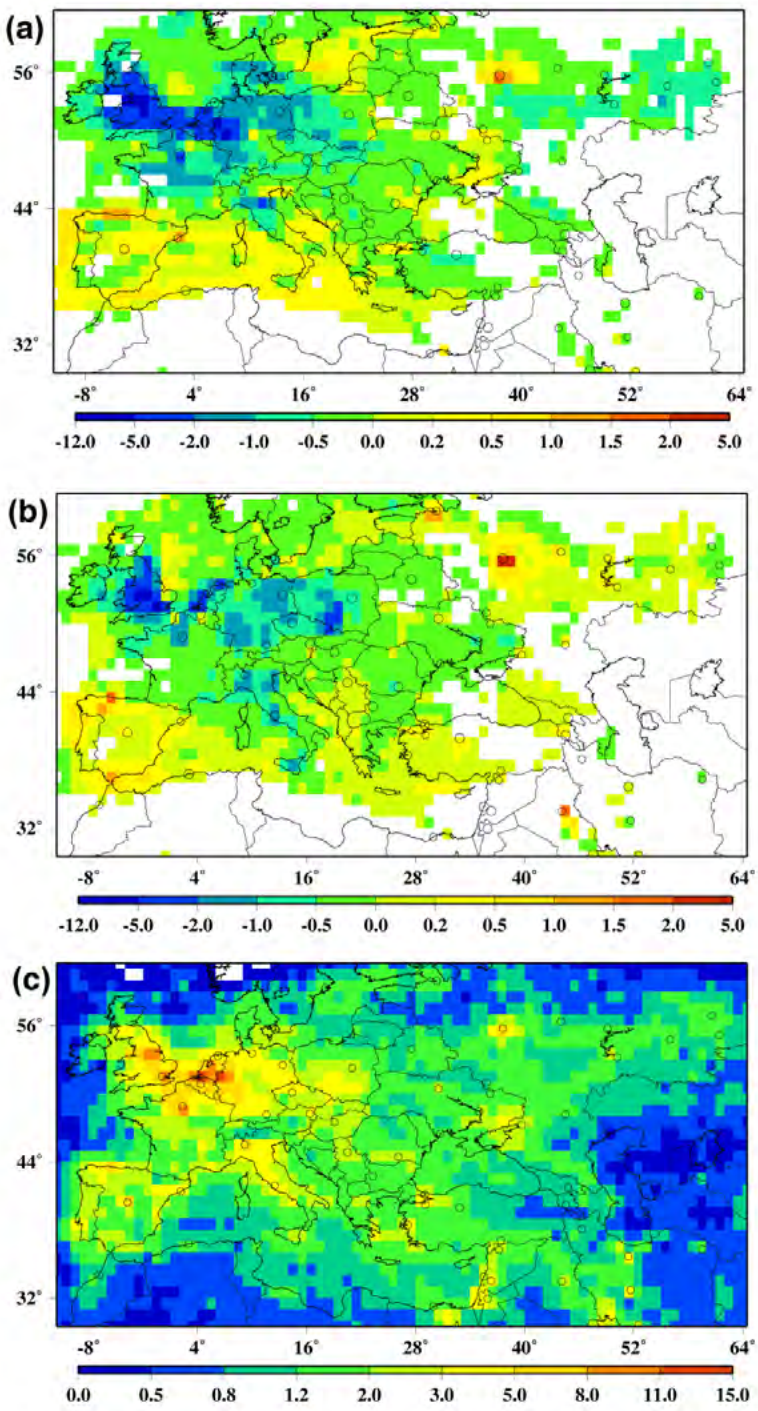

Figure 2. 1996 to 2005 linear trends (molecules × cm^{-2} × yr^{-1} × 10^{14}) from a) observed NO_2 columns in comparison to the trends in b) NO_2 columns simulated using CHIMERE model and c) magnitudes of tropospheric NO_2 columns (molecules × cm^{-2} × 10^{15}) derived from summer 2003 SCIAMACHY measurements. The trends are shown only for those grid cells for which the contribution of NO_2 from anthropogenic sources is dominating the total tropospheric NO_2 column amount [*from Konovalov et al, 2008*].

Observations from operational passenger aircraft (e.g. CARIBIC) have been evaluated and integrated into a unique high-quality data set.

3. Atmospheric aerosols and climate

A reliable and harmonised European network of aerosol observation networks has been developed, incorporating data from ground-based in situ measurements, ground-based remote sensing and complementary satellite data. This provides long-term measurements of aerosol micro-physical, chemical and optical properties linked to air quality and climate. This network is being used to assess the accuracy of, and the relationship between, ground-based and satellite aerosol measurements. Consequently, a 4D description of aerosol load over Europe based on this range of observations is being established.

4. Stratospheric ozone and climate

GEOmon supports the continuation of European stratospheric monitoring contributing to the Network for the Detection of Stratospheric Change (NDACC). Quality-assured, homogeneous ground-based measurements will be supplied to evaluate long-term trends in stratospheric O_3, NO_2, BrO and temperature. Corresponding satellite observations, together with ground-based data have been collated and integrated with modelling studies, consequently increasing our understanding of the evolution of stratospheric variables, and their link to climate changes.

5. Integration and supporting modelling studies

An overarching theme in GEOmon is the integration of the data sets described above with advanced Chemical-Transport Models. One example is given in Figure 2 which shows a comparison of trends in observed NO_2 from ground based stations with that derived from satellite measurements and those simulated from modelling studies (Konovalov et al., 2008).

Model outputs for both the troposphere and stratosphere will be compared to observations on a regional and global scale, enabling the generation of four dimensional climatologies. Additionally, long-term trend analysis of model results will be performed for comparison to observed trends, in order to identify underlying processes.

Summary

GEOmon (GEOmon.eu) has established a common data centre (GEOmon.nilu.no), providing a means to archive, process and disseminate quality controlled, harmonised atmospheric composition datasets. These include data from existing GEOSS observing systems, national and international databases of previous and current projects, as well as measurements from studies directly supported by GEOmon. The data portal comprises measurements from a range of techniques from *in-situ* ground-based and remote sensing observations, to aircraft data and numerical simulations. Using such datasets, GEOmon is conducting comparative analysis with both satellite observations and modelling studies in order to quantify and understand the long-term changes of the European atmospheric composition.

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Past issues of *IGACtivities* can be downloaded from http://www.igac.noaa.gov/newsletter/

IGAC*tivities* Newsletter

Published by IGAC Core Project Office
RESEARCH CENTER FOR ENVIRONMENTAL CHANGE
ACADEMIA SINICA
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IGAC was initiated by the Commission on Atmospheric Chemistry and Global Pollution (CACGP) and is a Core Project of the International Geosphere-Biosphere Programme (IGBP). The IGAC Seattle Core Project Office is currently supported by the National Science Foundation (NSF), National Aeronautics and Space Administration (NASA), and National Oceanic and Atmospheric Administration (NOAA). The IGAC Taipei Core Project Office is funded by Academia Sinica, Taipei. The Rome Core Project Office is supported by the Italian National Research Council and by the European Commission Network of Excellence ACCENT. Any opinions, findings, and conclusions, or recommendations expressed in this newsletter are those of the individual author(s) and do not necessarily reflect the views of the responsible funding agencies.

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