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

CACGP
In Cooperation with IAMAS Commission on Atmospheric Chemistry and Global Pollution

A Note from the IGAC Co-chairs: Sandro Fuzzi, Phil Rasch and Shaw Liu

Last month the IGAC scientific steering committee convened for our annual SSC meeting. The meeting provided an opportunity to review current activities and to plan for the future. It is clear there are a number of exciting opportunities on the horizon for IGAC. In this issue we describe a few areas of current activity. We include a paper describing the discussions of a recent workshop on the Upper Troposphere and Lower Stratosphere (UTLS) and a report on an IGAC workshop focusing on the aerosol Indirect Effect (IE) on clouds and climate.

In the last few years a number of opportunities have arisen to interact with our sister organization SPARC. We have begun discussions between IGBP/IGAC and WCRP/SPARC on how we might collaborate further. It is obvious that there are many areas of common interest and overlap, and these provide opportunities for synergistic activities. It is noteworthy that the UTLS workshop was jointly organized with SPARC, and the Indirect Effect workshop covered a topic receiving increasing attention by SPARC. With this focus on planning, we also want to solicit the IGAC community on potential future directions for the organization. We are always interested in well thought-out proposals that IGAC should be considering.

One of the special responsibilities of IGAC is to foster research opportunities that are multidisciplinary and difficult to do at the national level. The recent evolution of a number of scientific projects to involve modelers, in-situ measurements and remote sensing provides a specific example of this kind of opportunity. Models provide a tool that represents our understanding of the physical system. As such, their flaws reflect our lack of knowledge but, despite these flaws, they provide useful tools for planning and interpretation of measurements. Models provide an unparalleled opportunity for connecting and interpreting in-situ measurements with remote sensing, and can offer a guide to what kinds of measurements are most useful for understanding the atmosphere within the Earth system. With models one can also examine the effect of chemistry on the climate system. We encourage the whole IGAC community to think about problems that involve this synergy between models and measurements.

We also encourage consideration of novel ways of combining tasks to extract new information. One example of this might be a program designed to improve our understanding of cloud transport and chemical processing that combines the expertise of the meteorological community and the atmospheric chemistry community. There have been a number of field experiments over the years in each community that have made valuable contributions to understanding clouds and their interaction with their environment. Examples of landmark international field experiments with an emphasis on clouds and meteorology were GATE (GARP Atlantic Tropical Experiment) and TOGA/COARE (the Tropical Ocean and Global Atmosphere Program/Coupled Ocean Atmosphere Response Experiment). The Indian Ocean Experiment (INDOEX) and IGAC's Aerosol Characterization Experiments (ACE-1 & -2, ACE-Asia) provide similar examples of landmark programs with a chemistry/climate focus.

In each of these field experiments certain limiting factors impaired the ability to understand how clouds and their environment interact. For example, the use of "mixing lines" (also know as tracer/tracer correlations) that have proven so useful in understand stratosphere troposphere interactions in the atmospheric chemistry/transport community, have also been used in the meteorological community. It is easy to postulate that a synergy would arise if both communities were to contribute to a single field program. The atmospheric chemistry and remote sensing communities could suggest species and provide tools (not just instruments) that would be more optimal than those historically used by the meteorological community, and the meteorological community could provide information, measurements, and models of cloud transport processes that could be significantly more sophisticated and realistic than those used by chemical modelers.

New important challenges are ahead of us, both in our own field of expertise and as part of the wider IGBP-ESSP ensemble, to foster understanding of the functioning of the Earth System. We hope IGAC will help the atmospheric chemistry community rise to these challenges.

SCIENCE FEATURES

Processes governing the chemical composition of the extra-tropical UTLS

A report from the joint SPARC-IGAC Workshop
18-20 May 2005
Max Planck Institut für Chemie
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I. Introduction and background

The links between atmospheric chemistry and climate are receiving increasing attention on several fronts. One region where the two are tightly coupled is the Upper Troposphere/Lower Stratosphere (UTLS), which spans the altitude range from ~8-16km (depending on latitude). Transport in this region and, in particular, exchange between the troposphere and stratosphere occurs through a combination of processes including, in the tropics, cumulus convection, and, in the extratropics, synoptic-scale weather systems, together with the large-scale Brewer-Dobson circulation. It is recognized that net exchange from troposphere to stratosphere in the tropics and from stratosphere to troposphere in the extratropics is under large-scale dynamical control (Holton et al 1995). However, the net exchange alone does not determine many important aspects of chemical distributions in the UTLS region. Recent observational and modeling studies have further revealed important complexities in UTLS dynamical processes and chemistry, the interplay between the two, and consequences for chemical distributions in the UTLS. In particular these studies have raised questions about the best definition of the boundary between the troposphere and stratosphere, i.e. the tropopause. This applies both to the tropics and to the extratropics. The processes and scientific questions in the two regions are rather different and confining attention to one or the other has some advantages. The subject of this report is the extratropical UTLS, i.e. poleward of the subtropical jets. Many important aspects of the tropical UTLS are discussed in recent papers by Folkins (2005), Folkins and Martin (2005), Gettelman et al (2004) and Küpper et al (2004) and references therein.

Based on this new information, a more sophisticated picture is being put together of the factors controlling UTLS chemistry and the climate feedbacks. Perturbations to the distributions of trace gases such as O_3, H_2O, and aerosols in this region can lead to direct forcing of climate. Indirect effects through, for example, changing cirrus following new particle production or contrail formation from aircraft emissions can also impact the radiative balance in this region. In turn, climate change, through changing temperatures and transport patterns, has the potential to effect the chemical composition of the extra-tropical UTLS and thus the composition of the troposphere and stratosphere. Transport of ozone from the stratosphere to the troposphere may change in response to ozone recovery and greenhouse gas impacts in the stratosphere. Also, as noted in the WMO 2003 Ozone Assessment, transport in the extra-tropics from the troposphere to the stratosphere of very short-lived halogenated species (VSLS; in particular bromine-containing compounds) and pollutants may be important for understanding current and future stratospheric ozone change.

In an effort to integrate and synthesize new findings and their implications, the IGAC Project (International

Global Atmospheric Chemistry; under IGBP and CACGP) and the SPARC Project (Stratospheric Processes and their Role in Climate; under WCRP) held a joint workshop at the Max Plank Institut für Chemie, Mainz in May 2005 to discuss processes governing the chemical composition of the Upper Troposphere and Lower Stratosphere (UTLS) in the extra-tropics. One aim of the workshop was to update our current state of knowledge following previous workshops discussing the tropopause (i.e. in Bad Tölz, Germany, 2001; Haynes and Shepherd, 2001) and chemistry-climate interactions (Giens, France, 2003; Ravishankara et al., 2004) which both included some discussion about extra-tropical UTLS composition. It was also felt that it is timely to review these issues given the upcoming WMO assessment in 2006 and given the issues raised in the previous ozone assessment (WMO, 2003). It was also noted that it is nearly 10 years since the publication of the very influential review by Holton et al (1995), which summarized the state of knowledge at that time related primarily to dynamical drivers of stratosphere-troposphere exchange (STE). Recent observations and modeling studies allow for refinement of these concepts, especially with respect to small(er)-scale dynamics and coupling to chemical composition.

II. Workshop Design & Discussion Topics

The workshop discussions were designed around four major scientific questions (below) pertinent to improving our understanding about UTLS extra-tropical chemical composition. Invited overview presentations were given on sub-themes identified within each topic and these were followed by lively discussions in plenary. Discussions were also held in breakout sessions where it was decided to combine the first two topics and discuss the roles of dynamical and chemical processes together. Rapporteurs summarized the discussions on the last day of the meeting. This report summarizes these discussions, focusing on the main highlights from the workshop.

The four framing questions for the workshop were:

1) Which dynamical and meteorological processes govern the chemical composition, especially ozone and water vapor, of the extra-tropical UTLS on seasonal and inter-annual timescales?

On a large scale, both temporally and spatially, the chemical composition of the extra-tropical UTLS is influenced by the downward transport of trace gases via the large-scale stratospheric circulation and the upward transport of trace constituents from the troposphere by dynamical processes such as frontal uplift and deep convection. Coupling of air masses between the sub-tropical UT and the extra-tropical LS may also be important. Many important details of these transport processes still need to be understood. Analyses of various datasets are now providing insights into the causes of large-scale seasonal and possibly inter-annual variability in transport processes and chemical composition. The extent to which small-scale processes (e.g. gravity wave breaking near the tropopause, turbulence in the vicinity of jet-streams, radiative processes associated with upper level clouds and condensation) play a role in governing the composition and exchange within the extra-tropical UTLS is also not well known. In addition, there is increasing evidence that deep convection or convection embedded in frontal systems could be important.

2) What is the relative importance of chemical versus dynamical processes in governing the chemical composition of the extra-tropical UTLS?

Analysis of observational datasets has shown that an extra-tropical tropopause layer (ExTL) exists in chemical composition between the stratosphere and troposphere which exhibits characteristics of both regions. The extent to which dynamical and/or chemical processes are influencing the composition of this region still remains to be quantified. A better characterization of how strongly the 3-D spatial (latitudinal, longitudinal, altitudinal) and seasonal chemical fields in this region are perturbed by exchange processes between the stratosphere and troposphere is needed in order to identify the relative importance of the chemical and dynamical processes. The impact of different processes such as (pyro-) convection and small-scale mixing on chemical composition are very uncertain and require better quantification.

3) Which chemical/physical processes are important in governing UTLS composition?

The physical conditions of the UTLS region (low T, decreasing pressure) give rise to particular conditions such that chemical reactions proceed at different rates than in the lower troposphere or the main bulk of the stratosphere. Large uncertainties still surround our knowledge about many reaction rates and pathways (e.g. VOC degradation, VSLS degradation) which could be important for the chemical composition of this region and which influence the distributions and budgets of HO_x, NO_x, BrO_x and O_3, for example. Very little is known about the aerosol budget in this region. In addition, heterogeneous reactions on ice/aerosols are also very uncertain as are the processes governing aerosol formation/ ageing, ice super-saturation and cirrus prop-

erties. Scaling up from the process scale to realistic parameterizations in global models is also an issue.

4) How do we better quantify the net exchange of ozone and other trace constituents between the stratosphere and the troposphere?

The flux of ozone from the stratosphere is an important term in the tropospheric ozone budget but global model estimates of net flux still vary by more than a factor of two (EU Chemistry-Climate report, 2004). In addition to the climate impacts, intrusions of ozone-rich stratospheric air into the troposphere can occasionally have significant implications for local regulation of allowable ground-level ozone concentrations and the achievability of established limits. Given that the flux may already have changed or may change in a future climate it is important to quantify this flux more accurately using new, better metrics. Variations in the methods used to determine the flux together with the paucity of independent estimates based on observations is contributing to these uncertainties. In particular, there is a need to define more meaningful parameters by which to quantify STE; i.e. ones which can be derived from observations and calculated in models. The concept of a chemical tropopause or exchange boundary between the stratosphere and troposphere is an important issue for defining the exchange, and the choice made for this boundary often influences the conclusions of STE studies. Advances in our knowledge about the processes governing the chemical composition of the UTLS region, refined methods to diagnose fluxes from meteorological data sets and the use of new observational datasets could lead to improved quantification of fluxes. There is also a more basic need to continue the evaluation of global model performance in the UTLS region given that these are the tools being used to integrate our current knowledge and provide predictions of future composition and climate to policy makers.

III. Discussion Summaries

Summaries of the plenary talks and breakout sessions follow. Reference is made in some cases to talks given by specific speakers *[names given in italics]*, but we note that this does not preclude the valuable contributions on the topic made by other participants. A full list of workshop participants and talks can be viewed on the workshop web page: http://www.atmosp.physics.utronto.ca/SPARC/UTLS%20IGAC/Index.htm. As many acronyms (for field projects, satellites, etc.) are used herein, an acronym list with translations is also provided at the end of the paper.

A. Chemistry and Dynamics: Indicators and Controlling Factors of UTLS Chemistry

(i) The Extra-tropical Tropopause Layer (ExTL)

While the boundary between the troposphere and stratosphere is generally considered to be defined by the thermal tropopause, this definition is not necessarily appropriate or meaningful when discussing chemical composition. The chemical and thermal tropopause are not generally coincident and, further, the chemical transition from UT to LS is not as abrupt or well-defined as the temperature transition. The workshop discussions followed the progress made in the last five years to identify and characterize the Extra-tropical Tropopause Layer (ExTL) from various in-situ and satellite observations of chemical tracers. Trace gas profiles of O_3, CO, CO_2, N_2O and H_2O, as well as scatter plots among these species, obtained from recent observations made as part of the airborne MOZAIC, CARIBIC, SPURT, STRAT/POLARIS and AIRS satellite projects, clearly reflected the existence of a transition layer in the upper troposphere/lowermost stratosphere (UT/LMS) where the chemical composition gradually changes from tropospheric (e.g. high CO, low O_3) to stratospheric (low CO, high O_3). Figure 1 shows an example of a CO-O_3 correlation in the tropopause region where mixing lines (blue) in the ExTL connect a tropospheric (green) and a stratospheric (red) trace gas reservoir. Number density distributions relative to the thermal tropopause show that the lower bound of the ExTL extends into the UT. The exact position is hard to determine, since it is neither associated with the thermal tropopause nor with a fixed value of PV. The upper bound of the ExTL (or the depth of the layer) depends to some extent on the residence time of the tracer under investigation. It is generally higher for species that have a long photochemical lifetime in the LMS (e.g. H_2O) than it is for short-lived species like CO, whose tropospheric signature is erased on a time scale of a few months due to net oxidation by OH in the LMS.

As outlined in a talk by *Rosenlof*, the chemical composition of the LMS is a function of the relative strength of several processes, such as episodic diabatic upwelling in particular in NH summer, quasi-isentropic cross tropopause transport, and diabatic downwelling from the overworld in the Brewer Dobson circulation. The first process is associated with deep overshooting convection and pyro-convection, and its bulk impact is largely unknown. In contrast, the upwelling is relatively easy to

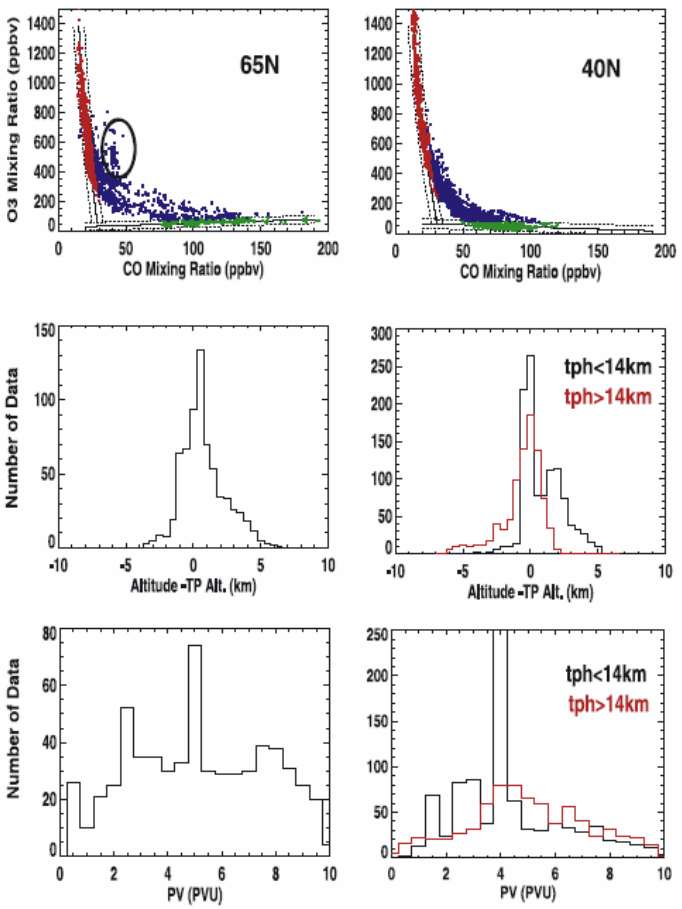


Figure 1 – CO-O_3 correlation has been used to identify the location and thickness of the extratropical transition layer (ExTL). The top panels display the relationship of stratospheric tracer O_3 and tropospheric tracer CO for the two extratropical locations sampled by the in situ measurements on board NASA research aircraft ER-2 during STRAT and POLARIS field campaigns (1995-1997). The solid lines represent the empirical stratospheric and tropospheric O_3-CO relationships, determined imperically from the data. The dash lines mark the 3σ of the respective distribution. The identified stratospheric, tropospheric, and transitional points are represented by red, green and blue. The center panels show the altitude distribution of transition points (blue) relative to the thermal tropopause. In the case of 40° N, the distributions are given as two populations, depending on whether the respective thermal tropopause height is below or above 14 km. The bottom panels show the potential vorticity distribution of the transition points. The 40°N distributions are given as two populations, similar to the center panels. (Adapted from Pan et al., 2004).

quantify via the calculation of EP fluxes[i], however there is still considerable uncertainty about the main forcing that drives the upwelling and about the observed trend of increased tropical upwelling during the last 7 years.

Analysis of seasonal variations of trace gas measurements, presented by *Hoor* for the SPURT project, reveals the importance of three reservoirs for the understanding of the chemical composition of the ExTL (Figure 2). The seasonal cycle of CO_2 in the UT (Fig. 2, black) and the ExTL (green) is in phase, demonstrating the strong coupling between the ExTL and the UT due to frequent cross-tropopause exchange. Above the ExTL in the LMS (red) the CO_2 maximum is shifted by approximately 3-4 months indicating transport from the tropical lowermost stratosphere. This transport to extra-tropical latitudes occurs within 2-4 months and leads to mixing with photochemically aged air diabatically descending from the overworld (Rosenlof et al., 1997).

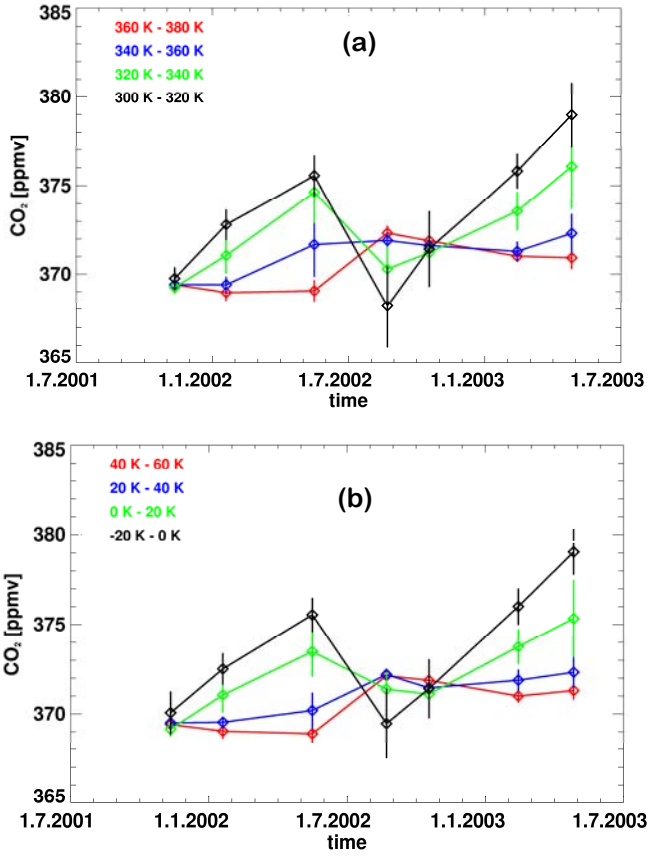


Figure 2 – Seasonal variation of CO_2 concentrations as a function of (a) distance in potential temperature relative to the tropopause (2 PVU surface) and (b) the potential temperature (Hoor et al., 2004).

[i] Eliassen-Palm flux (EP flux) is a measure of atmospheric wave propagation in the meridional plane.

(ii) Meteorological Processes

Several key meteorological processes in the troposphere contribute to the aforementioned episodic diabatic upwelling into the lowermost stratosphere. These processes include synoptic-scale transport events referred to as conveyor belts as well as smaller-scale deep convective systems. Both conveyor belts and deep convective events are associated with significant latent heat release due to condensation of water vapour and therefore they are distinct from isentropic transport. The role of these non-isentropic transport events for stratosphere-troposphere exchange (STE) has gained increased attention during the last years and hence constituted the main items of the presentations by *Stohl* and *Lawrence*.

The discussion of meteorological processes that are associated with significant transport events from the stratosphere to the troposphere (STT) or vice versa (TST) started on the synoptic scale. In his presentation on this topic, *Stohl* focussed on the Lagrangian perspective and first suggested the terminology introduced during the STACCATO project, whereby STE is regarded as the overall STT plus TST processes, and deep exchange refers to rapid transport on synoptic time scales between the boundary layer and the lower stratosphere. Deep exchange defined in this way is regarded as particularly important because it brings together stratospheric and boundary layer air masses, with strongly differing chemical compositions, and does so on a short time scale ($\leq$ 1 day, e.g. Stohl et al. 2003). The concomitant occurrence of deep STT and deep TST can lead to a vertically inverted pattern with air of stratospheric origin close to the ground and polluted boundary layer air at the tropopause (Stohl and Trickl, 1999). Particular attention was given to the role of warm conveyor belts (WCBs) that occur ahead of intense cold fronts and which transport warm and moist air from the subtropical boundary layer to the northern extra-tropical upper troposphere within 1-2 days. According to the recent WCB climatology of Eckhardt et al. (2004), boundary layer starting points frequently occur near very polluted areas (east coasts of North America and Asia). About 5% of the WCBs eventually enter the lowermost stratosphere. The processes associated with the diagnosed increase in potential vorticity of the WCB air parcels entering the stratosphere is not yet well understood. One hypothesis is that diabatic potential vorticity changes occur due to radiative processes in the WCB outflow regions, characterized by strong vertical humidity gradients and clouds. *Wirth* showed results from idealized studies on this issue which indicated that significant PV changes can occur due to radiative processes near the interface of humid upper tropospheric and dry stratospheric layers.

Other synoptic-scale processes that are relevant for the STE in the mid-latitudes (e.g. the formation of tropopause folds, Rossby wave breaking) were not discussed in detail.

Lawrence presented a concise overview on the role of deep convection for STE. He showed that observations, parameterizations and cloud resolving models (CRMs) have been used to study this process. Almost no direct observations exist for STT due to convection, but idealized model simulations – for instance with the WRF model – show that convection can trigger STT. For upward transport across the extra-tropical tropopause (TST) associated with convection there are several observational and model studies that provide clear evidence for the existence of this process, in particular during the summer months. However, the net quantitative impact of this process is still largely unknown and requires further investigation.

Analysis of observations (e.g. STERAO, EULINOX, TRACE-P) has shown that transport of pollutants by this mechanism is important at extra-tropical latitudes, especially over Asia and central North America, leading to perturbations in upper tropospheric trace constituent budgets (e.g. O_3, HOx). Interestingly, data collected by the MOZAIC program over the last three years shows significant enhancements in NOy and O_3 in the upper troposphere over North America, and these are often not correlated with CO (Petzoldt et al., 2005; Figure 3). This indicates that lightning or possibly aircraft emissions may be a principal source of NOx in the UT over continental regions, something that was also suggested by analyses of previous aircraft campaign data (NOXAR, SONEX; e.g. Jeker et al., 2000). This is in contrast to regions downwind of continents, where frontal uplift of surface pollutants may be more important. More recent campaigns have shown that trace gases, including short-lived VOCs or OVOCs, can also be transported into the lower stratosphere (e.g. MINOS, *Fischer*). However, further study is needed on the significance of these measurements and processes for lower stratospheric composition (e.g. transport of short-lived bromine containing species). Continued analysis of data collected during previous campaigns has also led to reductions in the range of estimates for the global amount of NOx from lighting emissions to 2-9 Tg N per year. The combination of cloud-resolved modeling of convection/chemistry (DeCaria et al., 2005) and anvil NOx observations suggests that on average an intra-cloud flash produces nearly as much NO as a cloud-to-ground (CG) flash (*Pickering*; Figure 4). This is very different from previous estimates which assumed that an intra-cloud flash produced only one tenth of that

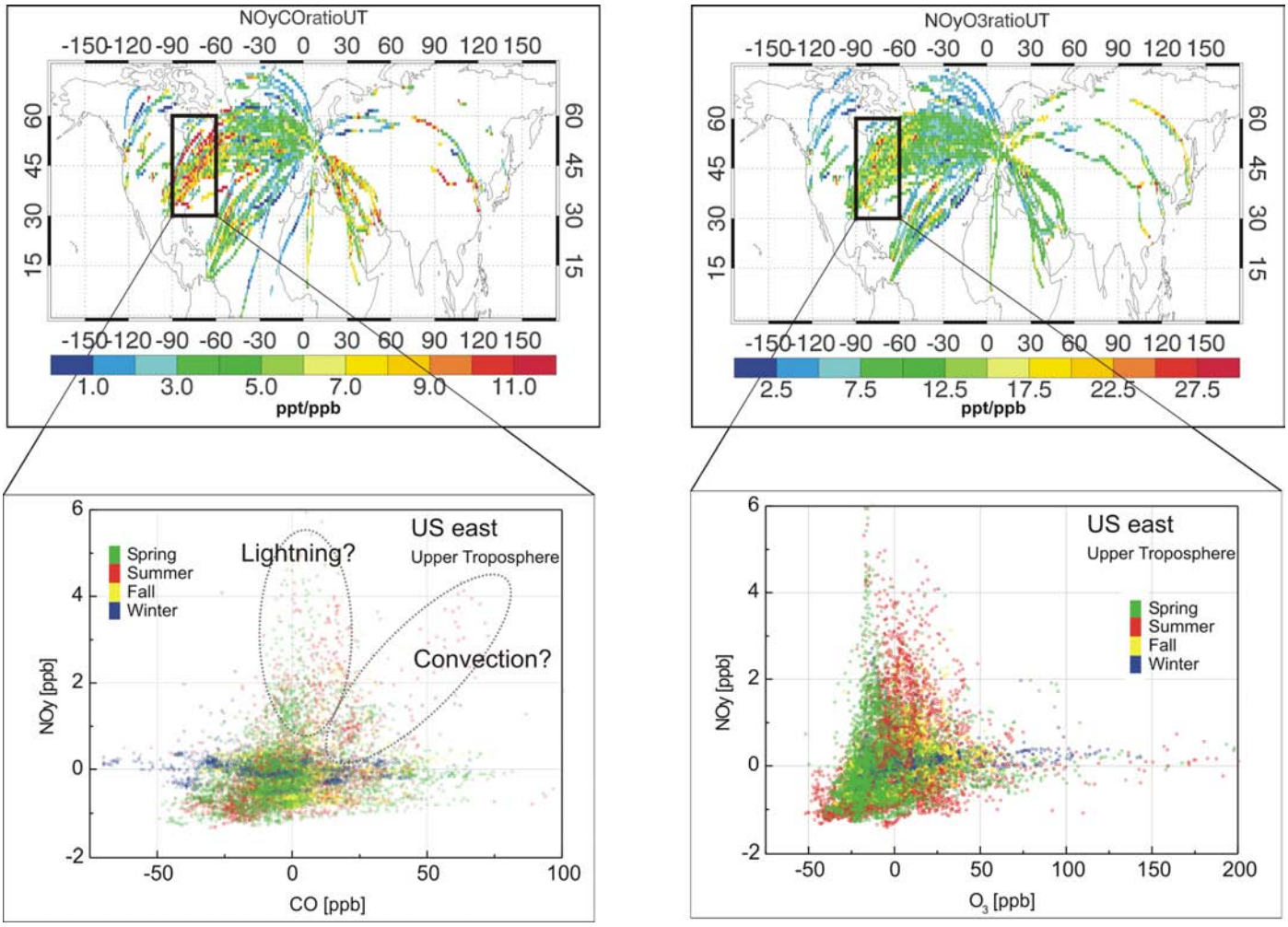


Figure 3 – Correlation of NOy against CO (left) and O_3 (right) over North America as a function of season (3 years of data, seasonal cycle removed). High NOy with low CO in summer suggests lightning influence and possibly aircraft emissions, whereas high NOy with high CO indicates the influence of convective transport of boundary layer pollution (anthropogenic emissions) into the UTLS over this region. In contrast, high O_3:CO correlations exists in UT over Asia/Siberia in certain years such as 2003. (Petzoldt et al., 2005).

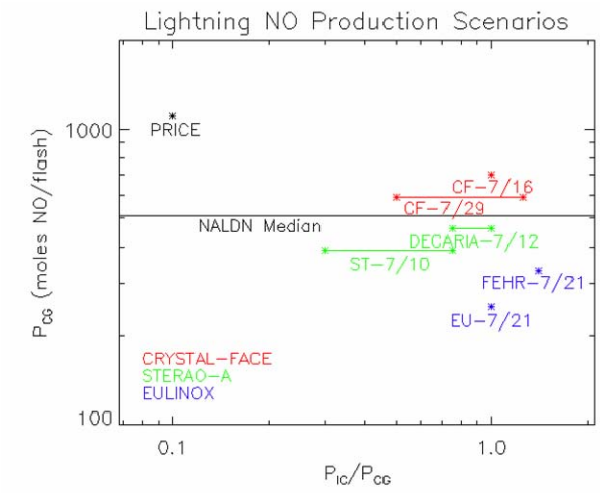


Figure 4 – From the presentation by K. Pickering, this figure shows recent estimates of the average production of NO by a cloud-to-ground (CG) flash and the ratio of average NO production via an intra-cloud (IC) flash versus that from a CG flash for various cloud/chemistry model simulations of observed storms in the northern hemisphere extra-tropics. These ratios are much higher (mean: 0.86) than previously assumed by, for example, Price et al. (1997) who used a ratio of 0.1 in their lightning parameterization in global chemical models. Also shown as a black line is the NO production rate per flash for cloud-to-ground flashes (PCG) for the median peak current for North America. This value of just over 500 moles NO/flash is much lower than was assumed by Price et al. (~1100 moles/flash).

of a CG flash (Price et al., 2007). Also, the newer estimates of the number of moles of NO per CG flash are significantly lower than previous estimates.

Recent evidence indicates that convection associated with forest fires, so-called pyro-convection, may also have a significant impact on mid-latitude UTLS composition. New modeling work presented by *Luderer* using the ATHAM model showed that the initiation of a deep pyro-convection event is very dependent on background meteorological conditions (e.g. cold frontal passage) as well as the sensible and latent heat budgets of the storm, the fire and the environment. There is a wealth of new evidence from airborne instruments (e.g. recent ICARTT campaign over N. Atlantic in 2004; MOZAIC data over Siberia in 2003; MOPITT CO satellite data – 2003/2004) of significant enhancements to the levels of trace gases such as CO during summertime periods of boreal forest burning. Whilst mainly confined to the free troposphere, certain very large pyro-convective storms can also penetrate above the tropopause, injecting material into the lower stratosphere. *Fromm* showed examples of enhanced values of aerosol (as viewed in terms of aerosol index by the POAM II satellite) several kilometers above the local tropopause in the lower stratosphere. Enhanced CO and also acetonitrile concentrations have also been observed in the LS and are attributed to forest fire emissions (e.g. Crystal-FACE, Jost et al., 2004; Livesey et al., 2004; MOZAIC, *Cammas;* Ray et al., 2004). The significance of these events – which may be occurring several times per year – is the topic of on-going research, as is their impact on stratospheric composition (aerosols, O_3) and the radiative budget. It is possible that even one large event per year may cause significant perturbations to background aerosol levels *[Fromm]* Figure 5.

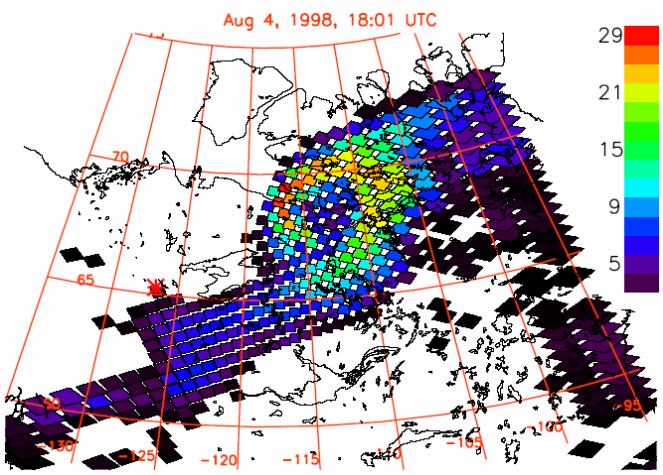
In addition to the new information emerging from field campaigns and satellites there have also been significant developments in the complexity of processes included in Cloud Resolving Models (CRMs) and mesoscale models, such as the inclusion of detailed chemical schemes including soluble species as well as aerosol and microphysical processes. However, many discrepancies still exist between different models, as shown by recent comparisons *[Barth]*. In particular, further validation is required of trace gas transport by convective systems into the LS and for this purpose new data is needed, particularly on short-lived species above convective systems. In addition, many of the mechanisms being studied/evaluated using CRM or mesoscale models are not included in global models. For example, downdrafts and gravity wave breaking at the tropopause, associated with deep convection, may be leading to STT in the extra-tropics. Embedded convection in frontal systems can be important for transporting trace gases such as CO into the UT and possibly the LS although the overall role of this mechanism still needs to be quantified and validated in models. There is also a need to compare results from CRMs with those from global models using the single column modeling approach and to continue with improvements to parameterizations of deep convective transport of tracers in global models and in particular treatments of soluble species and lightning emissions.

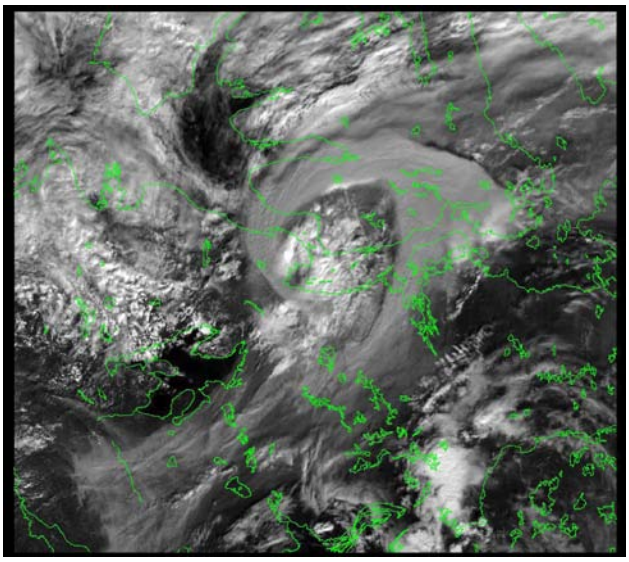
(iii) Mixing Processes

During the workshop discussions it was evident that there is a need to clarify the terminology around small-scale mixing phenomena. "Mixing" is sometimes used to mean "molecular mixing" and sometimes used to mean "stirring", i.e. deformation of material surfaces (and hence concentration fields of chemical species) by differential advection so that molecular diffusion is potentially enhanced, but without that diffusion necessarily acting to homogenize chemical concentrations. Stirring is a route to molecular mixing, but does not itself imply molecular mixing. This distinction is important because it is only molecular mixing that leads to chemical reactions (e.g. between species in previously chemically distinct airmasses).

The distinction between stirring and mixing is particularly important in the context of models. Lagrangian models can predict large gradients in chemical concentrations as a result of stirring but cannot (without significant modification from their usual form) describe the final step of molecular mixing. Eulerian models, on the other hand, assume that chemical concentration fields are constant – in other words well-mixed – within a grid box (typically 100 km x 100 km x 1 km for a global model). On the other hand, in-situ atmospheric data shows that chemical concentrations vary significantly in space – essentially down to the resolved scale of the observations: ~1 km for horizontal sections and a few tens of meters for vertical sections.

Two different types of stirring may be important to molecular-level mixing. The first is stirring via the large-scale flow, which can be resolved by global climate models and in global meteorological datasets. Here the distinction between stirring and mixing is important because the time scale for molecular mixing may be significantly larger than the time scale for stirring, as estimated by stretching rates. The second is stirring by three-dimensional turbulence arising in convective clouds, through breaking of gravity waves, and other such processes. The nature of three-dimensional turbu-





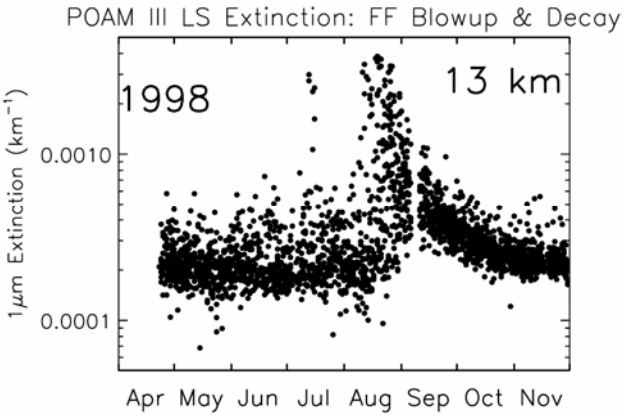


Figure 5 – Panels showing the impact of pyro-convective events (Fromm et al., 2005). (a) Earth Probe TOMS aerosol index (AI) over far northern Northwest Territories, 4 Aug 1998, a day after a pyro-convective eruption at Norman Wells (red asterisk). The very high AI values suggest a high-altitude plume. (b) A photograph of a pyro-convective cloud (Courtesy: Mr. Noriyuki Todo of Japan Airlines International Corporation). Details of the circumstances are: Flight data ID : JAL009 B747-400; Cruising Flight Level: FL340 (34,000 feet); location: N57 42.1 W125 00.0; time: 20hr 48min 27sec UTC on 27 June 2004. (c) Defense Meteorological Satellite Program (DMSP) Operational Linescan system (OLS) visible-channel reflectance image, 4 Aug 1998 at 1820 UTC, just minutes from TOMS AI map shown in (a). The croissant-shaped cloud is significantly less reflective than water-ice clouds. Infrared imagery (not shown) shows gray plume which is opaque at the UTLS level. (d) POAM III 1-micron extinction at 13 km altitude, for latitudes that vary gradually between 55 and 70N. Brief enhancement in mid-July was caused by a pyroCb described by Fromm et al. (2000). The large extinction enhancement in August followed the Norman Wells pyroCb of 3 August. Decay of the extinction can be seen over the successive three months.

lence, where stretching is dominated by the smallest eddies, is such that the time scale for molecular mixing (again for chemical species whose molecular diffusivity is similar to the viscous diffusivity) is similar to the time scale for stirring. In this case, distinguishing between stirring and mixing is not as critical.

Whiteway, Vaughan and others noted that inertia-gravity waves are likely to play a significant role in mixing in the tropopause region and above since their breaking gives rise to intermittent layers of three-dimensional turbulence (Figure 6). These waves may be generated by topography, by convection, or by synoptic-scale processes. However, the importance of convection for gravity-wave generation in the extra-tropics is not clear and furthermore the generation of inertia-gravity wave breaking by synoptic-scale systems is still poorly understood, though the fact of the generation is not in dispute. The tropopause level and above is a preferential region for breaking because of the change in static stability when going from the troposphere to the stratosphere. Wave breaking is regularly observed, such as over relatively weak topography in the U.K. mountains, and sometimes results from interactions between short wavelength gravity waves (perhaps directly generated by topography) and longer wavelength inertia-gravity waves (generated by synoptic scale processes). The resulting turbulent layers may be greater than 1 km in depth and hence imply substantial vertical transport.

One way of assessing the quantitative aspects of mixing is direct observations of the mixing processes themselves *[Whiteway]*. Another is to try to infer the characteristics of mixing from the observed structure of chemical concentrations fields, and determining which model representation of mixing gives the best fit to observations *[Legras]*. An interesting conclusion that comes out of this approach is that the strength of mixing processes is highly variable, as might be expected from the intermittency of three-dimensional turbulence and the likely association with particular geographic features such as topography. A related approach was used in the incorporation of mixing into the CLAMS model (a Lagrangian model with adaptive generation/destruction of parcels) where it is possible to optimize the mixing formulation to give best agreement with chemical observations *[Konopka]*.

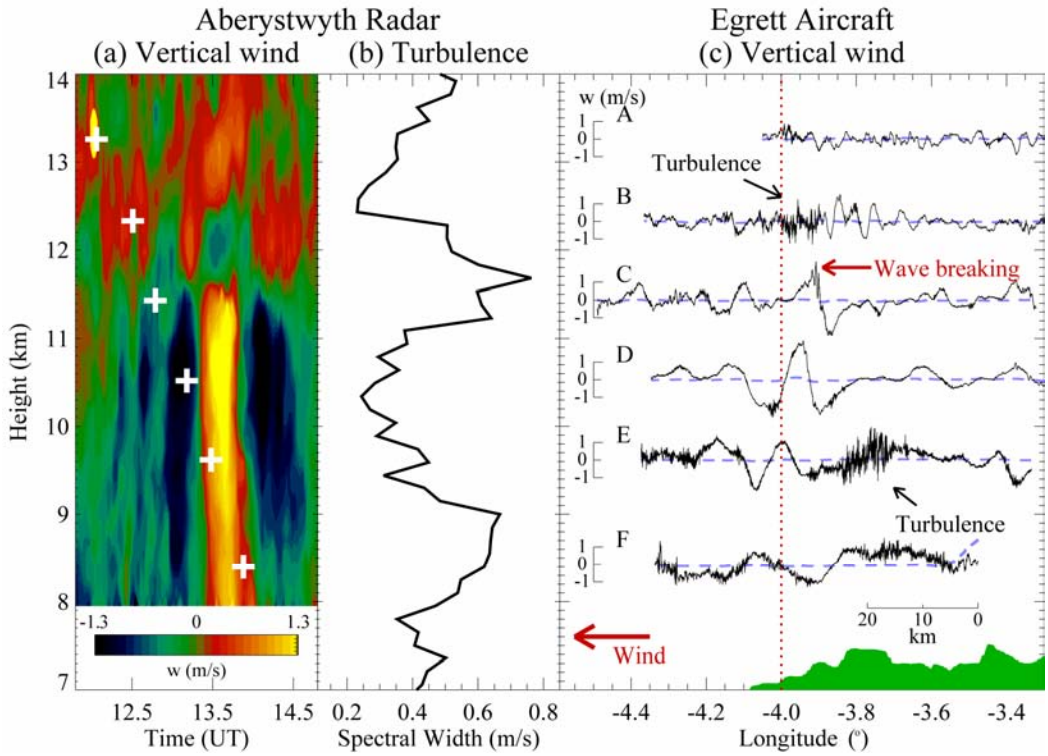


Figure 6 – (a) Measurements of vertical wind by the Aberystwyth VHF radar. (b) The spectral width of the radar signal averaged between 12:30 and 01:00 UTC. (c) Vertical wind measured on the Egrett. Each flight leg is placed at its height relative to the vertical scale in (a). The topographic height below the Egrett track is shown in green at the bottom with the same relative vertical scale as in (a). The coast of Wales is at 4.1° longitude; the position of the Aberystwyth radar is indicated by the vertical dotted line at 4.0° longitude. Crosses in (a) indicate the time and height when the Egrett passed directly above the radar. The turbulent layer between 11-12 km is estimated to have an internal turbulent diffusivity of about 2 m^2/s (*Whiteway, 2003*).

We know that global models with horizontal resolution of 100km or greater and satellite observations with resolutions of tens of kilometers cannot represent observed chemical concentration variations on scales of 1km or less. However, a more important question is whether the neglect of these variations leads to systematic large-scale errors in chemical predictions. This has been investigated in three different ways: (i) The implications of changing model resolution have been explored (Esler et al. 2004). (ii) The chemical implications of smoothing in-situ observations to give spatial resolutions typical of global models have been investigated (Crowther et al. 2002, Esler et al. 2004). (iii) The effect of mixing between

different boxes in Lagrangian calculations has been explored (Esler et al. 2001). Here the strongest effects are seen when the different boxes have very different initial chemical concentrations. In the UTLS context, this occurs when mixing air that originated in the boundary layer with air with the characteristics of the lower stratosphere. The extent to which this actually happens is not clear *[Vaughan]*. Approach (i) is the most straightforward to interpret with respect to implications for global-scale models and suggests that at current resolutions models may be making errors of up to 15% in key chemical quantities such as ozone production efficiency.

At present there are, as noted above, clearly several limitations to the representation of mixing in models. With Lagrangian models the difficulty is how to represent mixing effects without losing the essential simplicity of the Lagrangian approach. With Eulerian models the difficulty is how to reduce mixing to avoid unrealistic smoothing of important chemical contrasts (such as the tropopause itself). It is clear that mixing is, in reality, intermittent, but whether or not the details of that intermittency are important for large-scale chemical distributions or whether they must simply be taken into account to interpret individual observations remains to be determined.

B. In-situ Chemical and Microphysical Processes

The large- and small-scale dynamical processes discussed in the previous section alter the extra-tropical UTLS chemical composition by moving and mixing airmasses between the troposphere and stratosphere. In-situ chemical and microphysical processes in this thermodynamically and chemically unique region further alter its composition. Here we discuss several key species of particular importance to chemistry/climate interactions, controlling processes, and what steps are needed to better constrain them. Discussions below are based on presentations given by *Carslaw, Crowley, Dorf, Gettelman, Murphy, Peter, Ravishankara, Singh, Thornton, and von Glasow.*

(iv) Photochemistry

Upper tropospheric HO_x and NO_x. An accurate knowledge of the abundances of HO_x and NO_x in the upper troposphere is critical, since photochemical production of O_3 is controlled by the reactions of NO with HO_2 and RO_2. Recent observations in the field and laboratory have yielded insights to some important controlling processes. Observations from many tropospheric aircraft flights indicate that models tend to overestimate HO_x. These data were generally obtained at lower altitudes and at a higher ambient humidity than earlier observations that exhibited a discrepancy in the opposite direction. Observations also indicate that models tend to underestimate the HO_2/OH ratio at high levels of NO by large amounts (Figure 7). Recent laboratory observation show that, at high NO concentrations, the production of a few percent yield of HNO_3 by the NO+HO_2 reaction may alter the HO_2/OH ratio to be more consistent with observations (Butkovskaya et al., 2005). Finally, laboratory data have shown that acetone photolysis may be a less efficient source of HO_x than was previously believed (Blitz et al., 2004). Future approaches for constraining controlling processes on UT HO_x and NO_x include: i) Efforts to validate measurements of HO_x precursors via simultaneous observations by different instruments as well as budget studies; ii) Determining the level of agreement between modelled and measured OH and HO_2 if, in the models, only sources from $O(^1D)+H_2O$ and CH_4 oxidation as a function of NO or NO_x are considered (e.g., how important are non-water and no-methane sources of HO_x?), such as is done by Olson et al., 2004; and 3) Comparing observations of NO, HNO_3, and CO with CCM and CTM output in order to better quantify the efficiency of production of NO by lightning *[Singh, Salawitch]*.

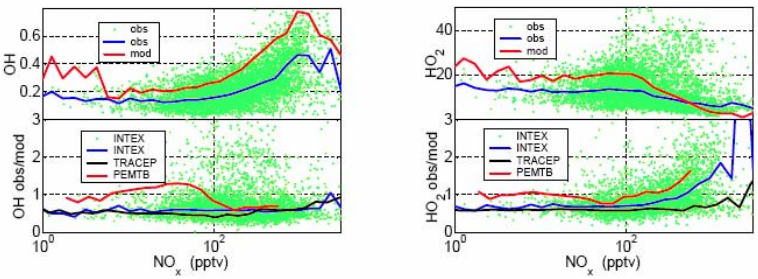


Figure 7 – Comparison of measured and modelled HO_x, as a function of NO_x, for data collected during INTEX, TRACE-P, and PEM Tropics B. (Presented by *Singh*; Courtesy Bill Brune, private communication).

Chlorine Activation. Recent aircraft data show that levels of ClO between 30 and 40 pptv are quite commonly observed at high latitudes in the northern hemisphere for stratospheric airmasses within several kilometres of the tropopause (Thornton et al., 2003). Levels of ClO between 20 and 30 pptv are also observed in the extra-tropical, UTLS region (Figure 8). These observations suggest that Cl activation on sub-visible cirrus, or on cold sulphate aerosols, might be responsible for a significant component of observed depletion of lower stratospheric ozone (Solomon et al., 1997; Breg man et al., 2002), in contrast to earlier studies in dry, particle-poor regions of the extra-tropical UTLS (Smith

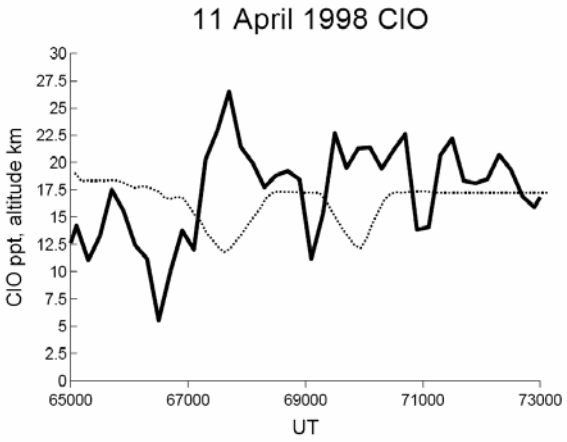


Figure 8 – ClO (solid) and altitude (dotted) for portion of 11 April 1998 WB-57F flight over central United States. Time is UT seconds. ClO is averaged for 120 sec (Courtesy Brett Thornton).

et al., 2001). The global significance of these regions of activated ClO is unclear. The observations of high ClO tend to occur in a spatially non-homogeneous manner. This could be due to variations in available chlorine along the flight track, which is difficult to assess without accurate, precise, high temporal resolution measurements of HCl, a surrogate for Cl_y. On the other hand, the patchiness could be related to the sporadic character of Cl activation, such as could be induced by mixing that combines particle or water-rich air with air that has high levels of Cly. It remains unclear whether formulations for Cl activation by PSCs (polar stratospheric clouds) can be applied to the heterogeneous activation of ClO on extra-tropical cirrus, given the nature of the water-rich aerosols and particles that form in the UTLS. Efforts needed to resolve these issues include simultaneous measurements of ClO and HCl in the UTLS, analysis of ice frost point temperature and cloud data from satellite data to assess global significance, and the modelling of existing ClO measurements to evaluate the heterogeneous chemistry schemes used in CTMs and CCMs. *[Thornton, plenary and breakout discussions]*.

Bromine and Iodine. Measurements of total column BrO by the GOME instrument reveal abundances that are more than a factor of two higher than found in typical models (Figure 9). The first issue raised by these observations is the need to define the relative contribution of tropospheric BrO and stratospheric BrO to this discrepancy. Results to date are not consistent. Ground-based measurements of the variation with solar zenith angle of differential slant column BrO suggest most of the discrepancy is caused by a global, ubiquitous, 2 to 3 pptv level of background BrO in the free troposphere (e.g., Müller et al., 2002). On the other hand, ground-based measurements of diffuse and direct solar radiation indicate an upper limit for tropospheric BrO of 0.9 pptv at 45°S, with mean values of ~0.2 pptv (Schofield et al., 2004). This suggests the discrepancy between measured and modelled column BrO might be the result of significantly higher levels of bromine in the LS than are commonly found in models. If BrO really is ~2-3pptv throughout the troposphere as suggested by the former study, then the BrO+HO_2 cycle could represent an important sink for O_3 (von Glasow et al., 2004), the hydrolysis of $BrONO_2$ could be an efficient route for production of HNO_3 (Lary, 2004), and BrO could be a significant oxidant for DMS (and perhaps other species) in the marine boundary layer (Boucher et al., 2003). If the "excess" bromine is in fact in the LS, this bromine could be supplied by the decomposition products of very

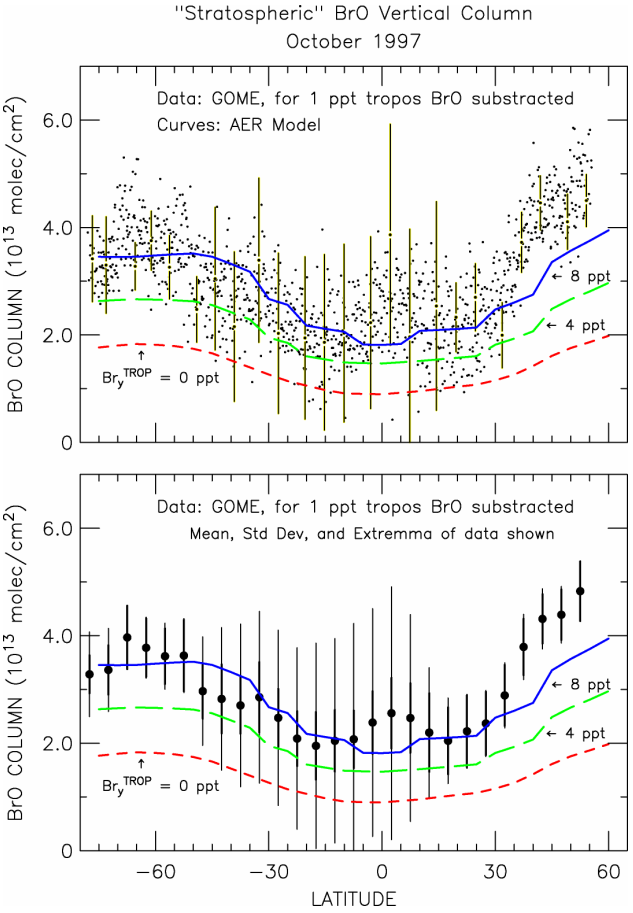


Figure 9 – Comparison of estimated stratospheric column BrO from GOME, October 1997, assuming a uniform 1 ppt distribution of BrO in the troposphere (close to the upper limit of 0.9 reported by Schofield *et al.*, JGR, 2004) compared to stratospheric column BrO from the AER 2D model, for the WMO 2003 baseline Br_y scenario (labeled Br_y^{TROP} =0) and for model simulations assuming non zero levels of Bry at the tropopause (Br_y^{TROP}) equal to 4 ppt and 8 ppt, respectively. *Top panel*: raw GOME data. *Bottom panel*: mean, standard deviation (thick error bars), and extremma (thin error bars) of GOME data in 5° wide latitude bins. After Salawitch *et al.*, GRL, 2005.

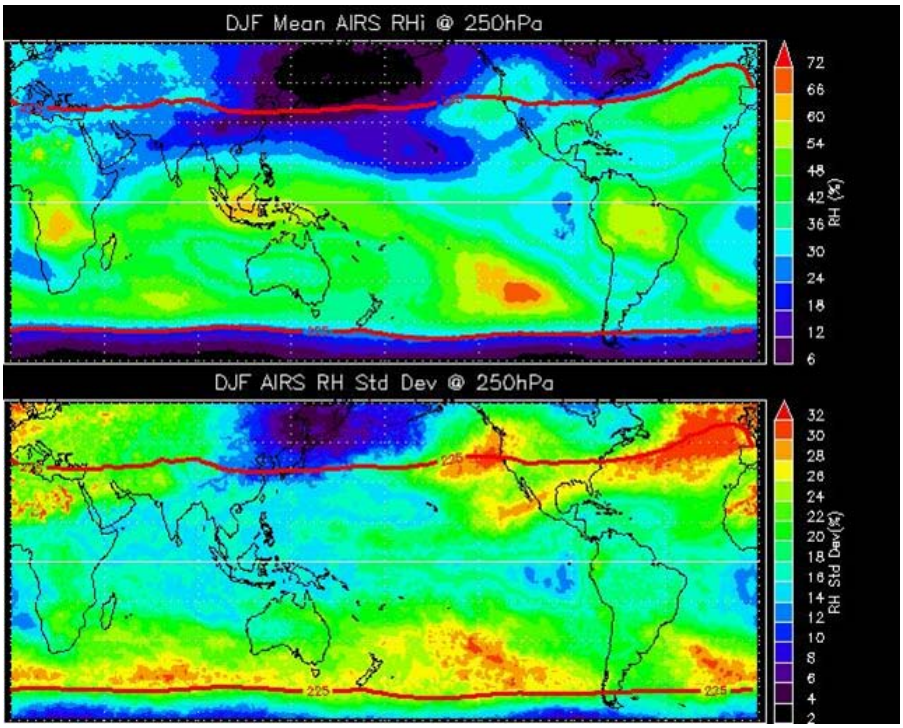


Figure 10 – From the presentation by *A. Gettelman*. Relative humidity over ice (RHI) from the Atmospheric Infrared Sounder (AIRS) at 250hPa averaged for December-February (top) and standard deviation of daily RHI from AIRS at 250hPa for an average of December-February 2002-2005 (bottom). The thick red line marks the thermal tropopause at 225hPa, corresponding to the AIRS layer pictured. The extratropical stratosphere poleward of the tropopause is very dry. The tropopause marks the boundary of high humidity regions, and the upper troposphere has high humidities, particularly in convective regions. The daily variance of RHI maximizes around the tropopause at this level, and is highest in the North Atlantic and North East Pacific, mostly equator-ward of the thermal tropopause. High variance is also found along the tropopause in the southern hemisphere. Variations do not imply transport, but fluctuations between tropospheric and stratospheric air at this level.

short lived (VSL) bromocarbons and could have important consequences for our understanding of ozone trends (WMO, 2003). The substantial organic content of many aerosol particles just above the tropopause suggests there is injection of tropospheric particles into the stratosphere, and the presence of Br on these particles provides the possibility of cross-tropopause transport of bromine, in both directions, by aerosols (Murphy and Thomson, 2000). Also, the presence of iodine on aerosols may explain the lack of stratospheric IO (e.g., via aerosol uptake of I_y species). Resolution of these issues require accurate and precise measurements of BrO in the UTLS region (i.e. that have sensitivity as low as 0.5 pptv); the simultaneous measurement of a suite of organics and inorganic decomposition products; and, laboratory measurements of heterogeneous chemical reactions of inorganic bromine species and the kinetics of the organic decomposition products of VSL bromocarbons *[Salawitch, Dorf, von Glasow, Murphy, and plenary and breakout discussions]*.

(v) Photochemistry

Water abundance and super-saturation. An accurate knowledge of the abundance of H_2O and ambient temperature is crucial for understanding cirrus cloud formation, estimating radiative forcing, and accurately retrieving aerosols and trace chemical species from satellites. Ice super-saturation has been frequently detected in clear air and inside cirrus clouds, predominantly in the UT (Jensen et al., 2001; Haag et al., 2003). Satellite observations point to a high variability of relative humidity in the ExTL in regions of major storm tracks (Figure 10). These are regions of significant dynamical perturbations, likely coinciding with enhanced mixing of tropospheric H_2O across the tropopause. This picture is corroborated by a few case studies of cross-tropopause tracer transport. Mixing ratios of H_2O well above stratospheric background levels are observed, reaching far into the lowermost stratosphere, especially in summer *[Schiller, breakout discussions]*. Supersaturation and the nucleation of the ice phase appears to be confined to a vertically narrow layer (up to 1 km thick at mid-latitudes and more extended polewards) above the tropopause (Pan et al., 2000). In situ processes affecting H_2O amount and cloud formation/frequency near the ExTL do not seem to influence the observed trends in mid-latitude stratospheric H_2O *[Gettleman, breakout discussions]*. The quantification of the different microphysical and dynamical sinks and sources of H_2O is still very uncertain. It remains to be determined how often cirrus formation takes place in ice-supersaturated regions.

Aerosol transport and composition. Aerosol precursor gases and primary aerosol particles are injected into the UTLS by rapid vertical transport processes such as warm conveyor belts and deep convection (including pyro-convection), thereby influencing the aerosol budget and high cloud occurrence around the ExTL *[Carslaw]*. Besides organics, many UT particles contain both sulfate and carbon and a large fraction contain insoluble inclusions such as mineral dust and soot

(Murphy et al., 1998; Kojima et al., 2004). A small number of such particles may act as efficient heterogeneous ice nuclei, affecting cirrus formation by freezing at lower supersaturations than for liquid particles. The influence of aerosols originating in the troposphere on the highly variable and non-uniform UTLS particle composition is seen in measurements at up to 5 km above the tropopause *[Murphy]*. This challenges the conventional wisdom that those aerosol particles in this region are entirely composed of H_2SO_4 and H_2O. It remains unclear to what degree vertical transport affects the UTLS aerosol, how lofted aerosols are modified by interacting with gases and hydrometeors in convective clouds, and how these aerosols in turn modify the evolution of deep convective clouds and the formation of cirrus. A global, speciated mass budget of the UTLS aerosols including sources and sinks is lacking, and therefore it is currently not possible to accurately validate recently developed global aerosol models.

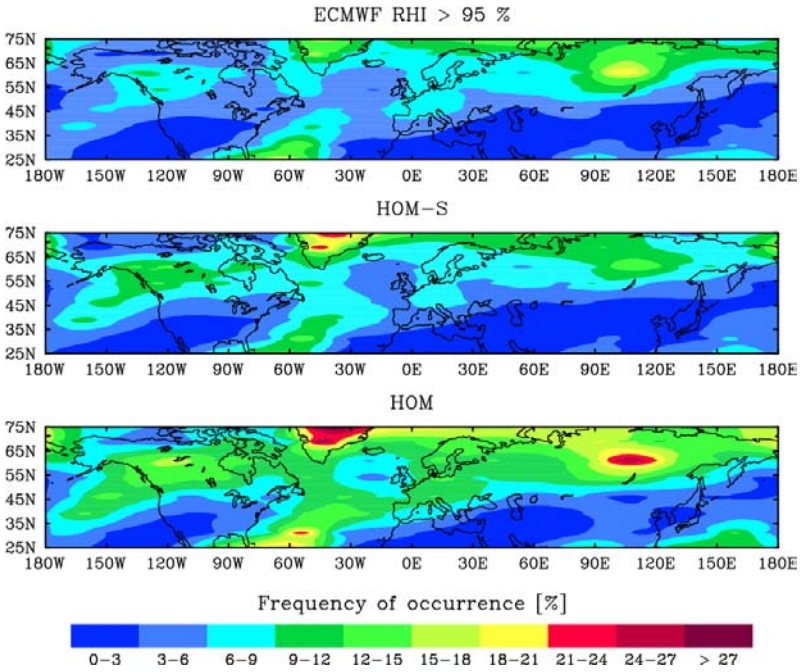


Figure 11 – Calculated frequencies of cirrus cloud occurrence during fall 2000 based on meteorological fields taken from the ECMWF model in T511/L60 resolution (Haag and Kärcher, 2004). The regions in which the relative humidity over ice (RHI) exceeds 95% are evaluated along synoptic trajectories driven by the ECMWF winds and are used as a measure for cirrus cloud cover (top, ECMWF). The forecast model uses a thermodynamically-based cloud scheme and forms cirrus at ice saturation. The other panels show results from explicit calculations of aerosol and cirrus cloud microphysics along the trajectories. This approach takes into account that cirrus form at significant supersaturations via homogeneous freezing and consider kinetic effects during growth and evaporation of ice crystals. The microphysical simulations use the synoptic temperatures (middle, HOM-S) and synoptic temperatures with superimposed small-scale temperature oscillations (bottom, HOM) caused by parameterized gravity waves. The occurrence frequency is lower in case HOM-S than in HOM, because average sizes of ice crystals are larger and their sedimentation speeds are faster in HOM-S, decreasing average cloud lifetimes.

Ice formation from aerosols. Ice cloud formation and characteristics may be changing due to two influences: a change in the abundance and properties of ice-nucleating aerosols (i.e. the aerosol indirect effect) and changes in the small-scale dynamical forcing patterns (Kärcher and Ström, 2003). The relative importance of these two is not well known. The dependence of the number of ice crystals on the updraft speed in a rising air parcel is much stronger than in liquid clouds, making cloud formation processes more susceptible to small dynamical changes than in the mid- to lower-troposphere. Frequent observations of high ice supersaturation in conjunction with high ice crystal number densities suggest a global-scale predominance of homogeneous freezing in the UTLS (DeMott et al., 2003; Gayet et al., 2004; Hoyle et al., 2005). Homogeneous freezing is sensitive to changes in the variability of vertical air motion on spatial and temporal scales unresolved by global models (Figure 11). The organic aerosol fraction does not seem to contribute significantly to ice formation (Cziczo et al., 2004; *Peter*), but a few heterogeneous ice nuclei could modify cirrus development and high cloud cover if they cause ice formation at lower supersaturations than required for homogeneous freezing (Figure 12). Changes in dynamical forcing could easily mask changes in cloud properties induced by ice nuclei, and these two influences are difficult to separate in measurements. Discriminating between natural and anthropogenic causes of cirrus changes in a future climate requires that mesoscale temperature fluctuations to be understood and that their sources (typically gravity waves) be accurately parameterized in global models. It is furthermore important to know to what extent ice nuclei modify radiatively important cirrus cloud properties.

Gas uptake in cirrus clouds. Uptake of chemically active trace gases by cirrus ice crystals could possibly lead to vertical redistribution or even irreversible removal of the gas from UT air masses, potentially altering the ozone budget there *[Crowley]*. Molecules re-

siding at the surfaces of ice crystals might alter ice particle growth rates by modifying the supersaturation over individual crystal facets (Gao et al., 2004). Cubic ice may alter ice crystal nucleation and growth, possibly over a wider range of temperatures than previously thought (Murray et al., 2005). A number of field measurements indicate there is substantial uptake of HNO_3 in low temperature cirrus clouds, in one case even in concert with enhanced in-cloud supersaturation. According to recent laboratory measurements, equilibrium uptake models frequently used in the past to calculate the uptake of HNO_3 on ice are inapplicable at the low HNO_3 partial pressures typical for the ExTL (Ullerstam et al., 2005). Perhaps more important, atmospheric ice is not in equilibrium. Both laboratory studies examining HNO_3 and HCl uptake and theoretical work suggest that growth and evaporation of ice may strongly affect the amount of species taken up (Kärcher and Basko, 2004). Growth models for small ice crystals that are valid for UTLS conditions and which are capable of accounting for habit changes and surface pollution are not available. It is unclear to what extent non-equilibrium processes connected to ice growth in cirrus conditions affect trace gas uptake and heterogeneous reaction rates.

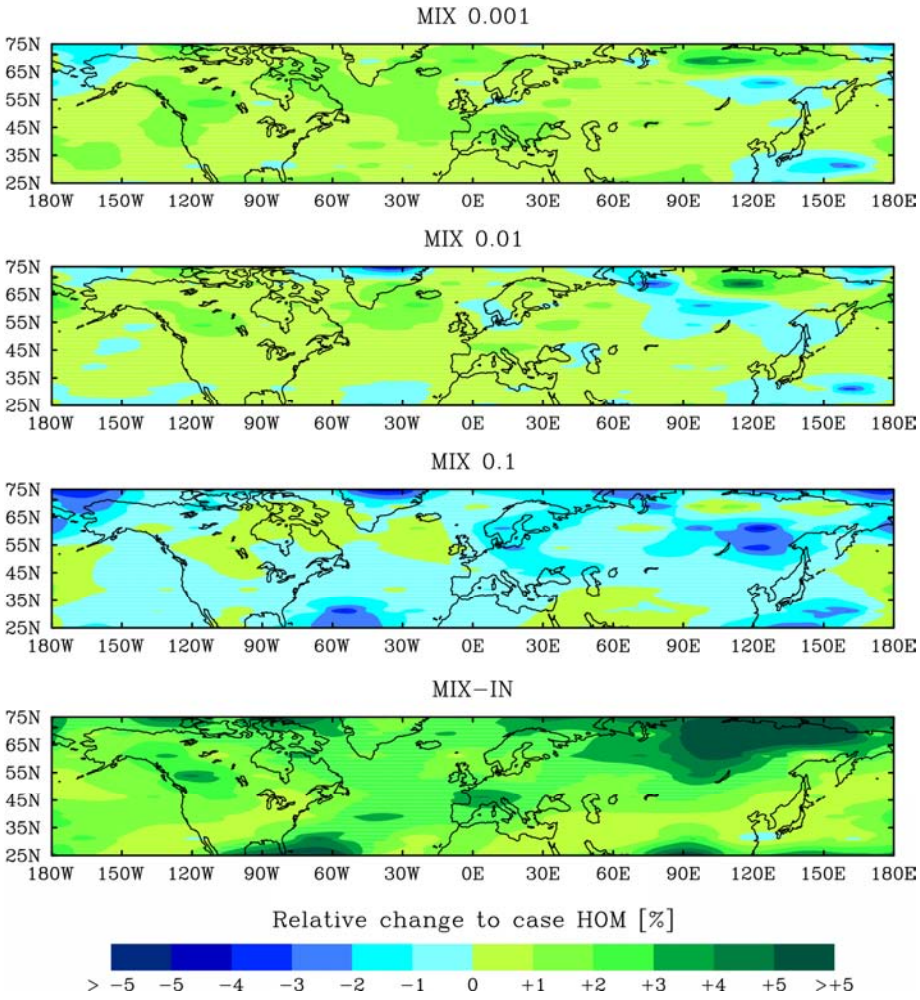


Figure 12 – Changes of the frequency of cirrus cloud occurrence relative to case HOM shown in Fig.Ya caused by additions of heterogeneous ice nuclei (IN) forming ice at 130% RHI. Total IN concentrations are *x* cm^{-3} in the cases MIX *x* (first 3 panels). The case MIX-IN (bottom) assumes 0.01 cm^{-3} extremely efficient IN that nucleate ice at 105% RHI. Field measurements suggest that background IN concentrations do not exceed 0.01 cm^{-3}, but higher values may occur locally. The cloud occurrence is a nonlinear function of ice nucleation thresholds and IN concentrations (for details see Haag and Kärcher, 2004). Changes in gravity wave properties also strongly modify the cloud occurrence (not shown).

C. Quantifying Net Exchange of Trace Constituents

Quantifying the global stratosphere-troposphere exchange (STE) of atmospheric species is a prerequisite for identifying the roles of different dynamical and photochemical processes in controlling this flux. In particular, the net flux of ozone (from stratosphere to troposphere) and of water as well as aerosols (from troposphere to stratosphere) are critical elements in the stratosphere-troposphere coupling and thus in the overall chemistry-climate coupling. As has been discussed, the area of transition from the troposphere to stratosphere is a region of partial mixing, small-scale dynamical processes, and unusual chemistry since it combines the characteristics of both the stratosphere and the troposphere. Key questions now being asked include: How important is O_3 STE to the tropospheric O_3 budget and the overall tropospheric oxidative capacity (i.e., OH)? How will climate change alter the flux of H_2O into the stratosphere? Do chemical processes in the tropopause transition region alter the STE of key species like O_3 and aerosols? How important are the large-scale, planetary disturbances vs. the small-scale dynamical processes in controlling this STE? What dynamical-chemical measurements would be needed to detect a significant change in STE? Answers to the above

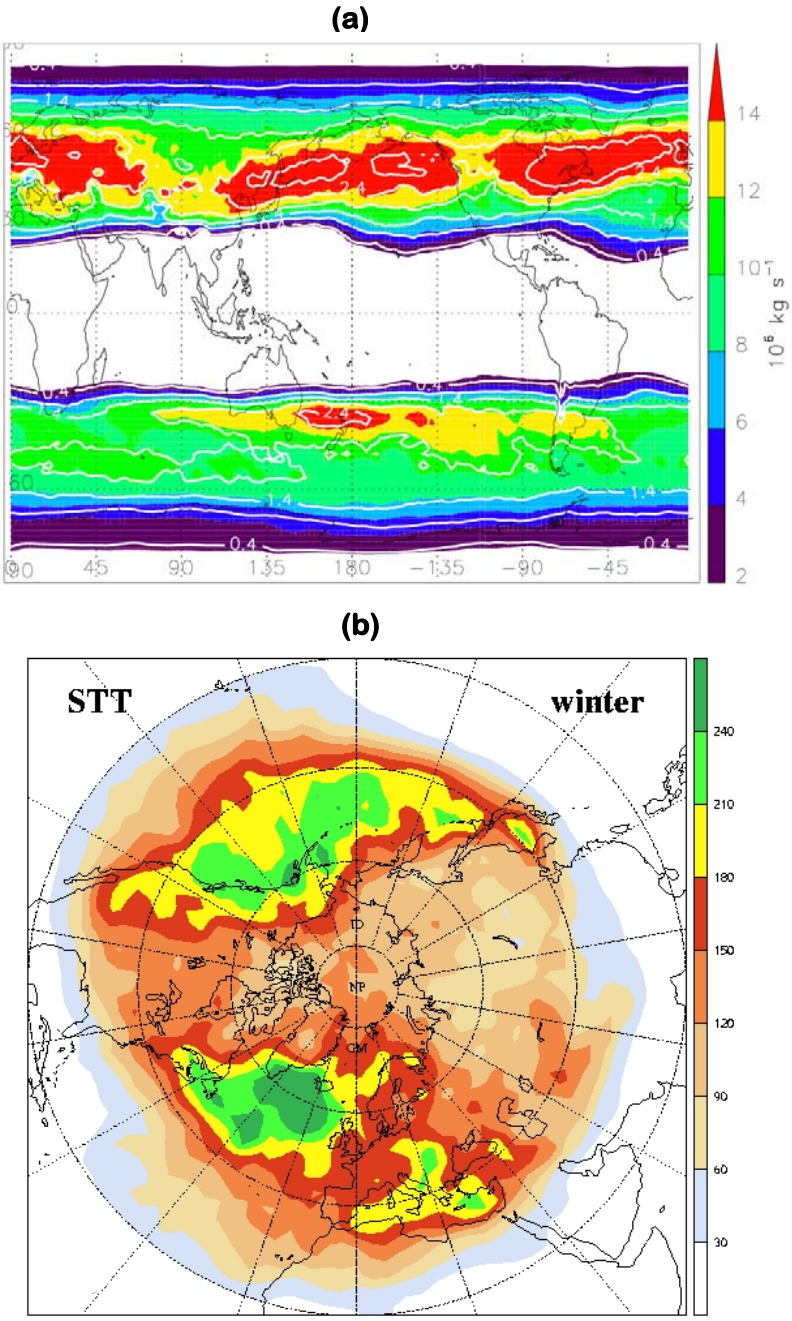


Figure 13 – Examples of Stratosphere flux by Eulerian and Lagrangian models. (a) Five-year mean extratropical diabatic flux of mass (color shading) and ozone (white contours) from the Goddard model. The ozone flux contour interval is 0.5 kg/s beginning at 0.4 kg/s [adapted from Olsen et al., 2004]. (b) 15 year climatology of STT mass flux for the Northern Hemisphere based on Lagrangian calculation using ECMWF meteorological fields. (Adapted from Sprenger and Wernli, 2003)

questions form the knowledge base required for estimating the chemical feedback in a changing climate.

Over the last decade, significant progress has been made in quantifying STE flux on both global and regional scales, and over both annual and synoptic times. Studies have ranged from high-resolution process studies to global integrations. In terms of the global pattern and magnitude of STE there is increasing convergence from the knowledge base of a decade ago, but complete agreement has not yet been reached. An example is given in Figure 13, where mass flux calculations using two different models (one Eulerian and one Lagrangian) show similarity in the preferred location of the net diabatic flux (Fig. 13a; Olsen et al. 2004) and the downward flux (STT, Fig. 13b; Sprenger and Wernli 2003) . These two quantities are comparable since STT is the dominant component of the net flux in the extratropics. The knowledge base is such that it is possible to generate maps of the O_3 STE on regional and monthly scales and to produce the now classic latitude-by-month plot of zonal mean O_3 STE to match the similar O_3 vertical column plots, as shown in Figure 14 (Hsu et al., 2005). With increasing model resolution and the use of analyzed meteorological fields, global CTMs are beginning to be able to reproduce the spatial and temporal variability observed in trace gas distributions, and they have become a useful tool for case studies of STE events. Examples from several field campaigns and intensive modeling studies have shown that in some cases we can model the fine, filamentary structure of ozone folds at the tropopause. Nonetheless, this remains a difficult task, as shown in Figure 15 (Wild et al. 2003), due to the fact that current CTMs still lack the full resolution of the observed structures. *[Prather, Olsen, Pan, Gettelman, Stohl, Law]*

Ten years ago, an important observational constraint to the calculated stratospheric ozone flux was given by the relationship of ozone with N_2O [Murphy and Fahey, 1994]. Tracer correlations in the lowermost stratosphere have proven useful in deriving global, annual mean fluxes of many constituents between the stratosphere and troposphere and in understanding the age of stratospheric air (i.e. time since it last was in the troposphere). Recently, a new observational study has shown the potential of O_3-HCl correlations to be a more accurate tracer relationship for constraining the amount of UT ozone that is of stratospheric origin (Marcy et al., 2004). *[Fahey]*

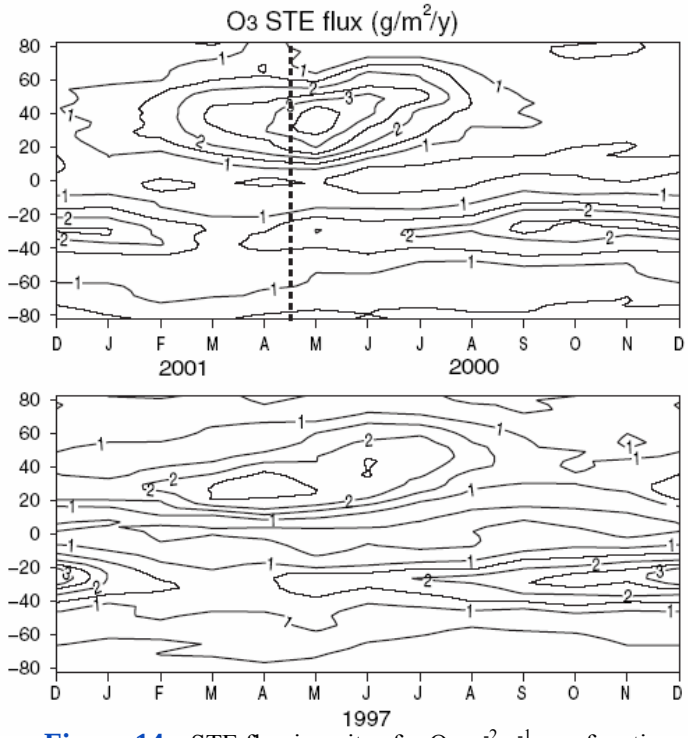
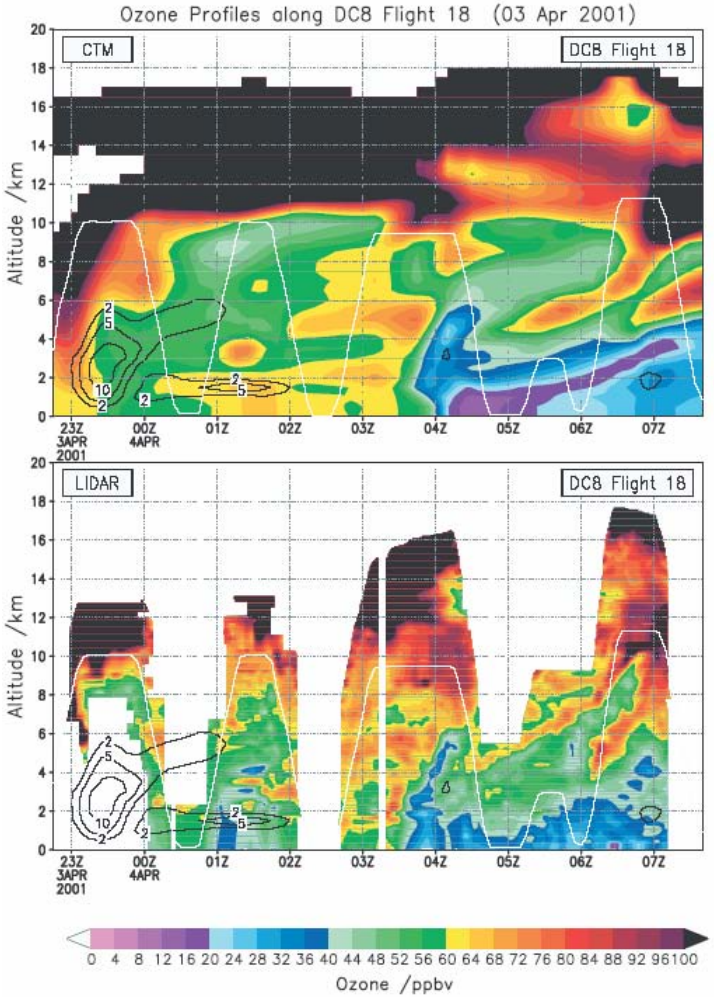


Figure 14 – STE flux in units of g-O_3 m^{-2} y^{-1} as a function of latitude and month for (top) year 2000/2001 and (bottom) year 1997 as calculated using EMWF pieced forecast met fields and the UCI CTM. (Adapted from Hsu et al., 2005).



Troposphere-to-stratosphere transport of water vapor and aerosols across the extratropical tropopause is an important yet not fully investigated aspect of STE. While evidence of "fresh" tropospheric air can be readily seen in the tropopause region where tracer correlations identify a mixed strat-trop chemical regime, it is not clear from models or measurements how large the flux of this fresh material is and whether it influences the middle stratosphere. Volcanic eruptions provide a test of our ability to model the reverse flux in this region, such as for simulations of Mt. Pinatubo aerosols mixing across the extratropical tropopause from the lower stratosphere and thus contributing to the UT aerosol burden. *[Penner]*

Despite recent progress, the community has yet to digest and incorporate this new knowledge into current applications. For example, the STE terms in the tropospheric ozone budgets among major models still differ by a factor of 2 to 3. This raises the important question: How can our improved knowledge of the extratropical UTLS actually be implemented to improve the models? *[Prather]*

One key issue is what metric to use to calibrate the performance of global models in calculating STE flux. The use of newly-established tracer-tracer correlations across the tropopause is one option, although the theory of tracer relationships within the troposphere is incomplete. Many intensive field studies (e.g., from TRACE-P to MOZAIC) clearly demonstrate that O_3-H_2O, CO-O_3, or HCl-O_3 correlations can be used to define purely stratospheric, purely tropospheric, and mixed air masses. What is uncertain is whether a CTM simulation that reproduces these correlations necessarily implies the correct STE. New generations of satellite data provide the opportunity of using tracer-tracer correlations on a global scale and with spatial resolutions comparable with that of global models. AIRS (on the Aqua satellite) O_3- H_2O correlations and MLS (on the Aura satellite) O_3-CO correlations are two

Figure 15 – Comparison of FRSGC/UCI chemistry-transport model and DC-8 LIDAR ozone profiles for Flight 18 from Hong Kong to Hawaii on 3 Apr 2001 showing stratospheric O_3 intrusions. The color scale highlights O_3 abundances less than 100 ppb, with100-500 ppb shown as black, and greater than 500 ppb masked (white). The flight track of the DC-8 is shown in white, and black contours indicate approximate cloud optical extinction (per km) specified from the met fields. (Adapted from Wild et al., 2003).

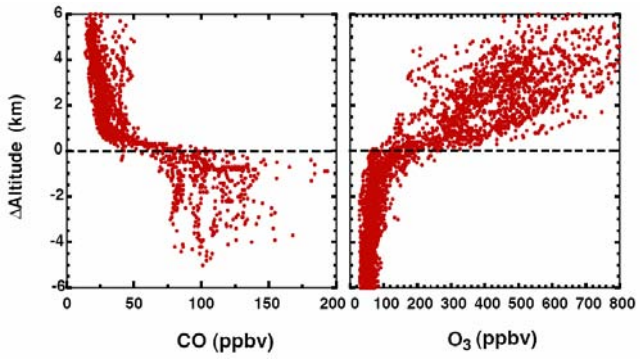


Figure 16 – Chemical discontinuity across the tropopause. The CO and O3 data are from ER-2 measurements during POLARIS campaign near Fairbanks, Alaska, April-August 1997. When altitude relative to the thermal tropopause is used as the vertical coordinate, the tracer profiles show abrupt change near the tropopause. (Adapted from Pan et al., 2004).

examples of such data sets. These datasets, however, often represent spatial averages over small-scale features. It is important to compare the satellite data with in-situ datasets like MOZAIC to understand the limitations of the data due to spatial averaging. *[Law, Prather, Pan]*

A confounding factor in determining stratosphere-troposphere exchange in models is that observations of chemical discontinuities show that transport barriers appear to exist across the tropopause (Figure 16) and the choice of the precise transport boundary may make a significant difference in the calculated flux. Models, on the other hand, often produce much smoother chemical transitions, in part due to numerical diffusion within the models. A key question is whether the calculated flux depends on the choice of boundary, which would imply that chemical transformations in the transition zone are important. *[Pan, Prather, Gettelman]*

Further, defining a correct location for the "boundary" between the stratosphere and troposphere can be ambiguous because of the presence of the extratropical tropopause layer (ExTL) which has a mix of stratospheric and tropospheric chemical characteristics. Should we determine a new way of defining STE flux with consideration of this transitional layer? Would accurate simulation of the ExTL change the STE flux? This is only important if there are chemical sources/sinks in this layer, because in the absence of chemical processes, the ozone flux is conserved across the ExTL *[Law, Prather]*.

Over the past 25 years, there have been significant long-term declines in mid-latitude LS ozone levels, and this is an important factor in changing the STT flux of ozone. Both dynamics and chemistry likely contribute to this long-term ozone depletion. The possible importance of VSL species to enhancing the chemical loss of O_3 due to Cl_y and Br_y species was discussed. More observations are needed to quantify the significance of VSLS-related long-term ozone depletion, as well as the relative contribution of chemical and dynamical forcings to the observed long-term changes in ozone *[Logan, Salawitch]*

New satellite data provide an exciting opportunity for validating and constraining models in the UTLS region. In particular, the AIRS instrument on Aqua and TES, MLS instruments on Aura all provide global ozone field in the UTLS region. Figure 17 gives an example of an AIRS ozone cross-section, illustrating the dynamical variability of ozone in the tropopause region and the potential of using satellite data for characterizing the impact of STE on UT ozone. We have only begun to explore the use of these datasets for characterizing and quantifying the integrated effect of STE. *[Pan, Gettelman]*

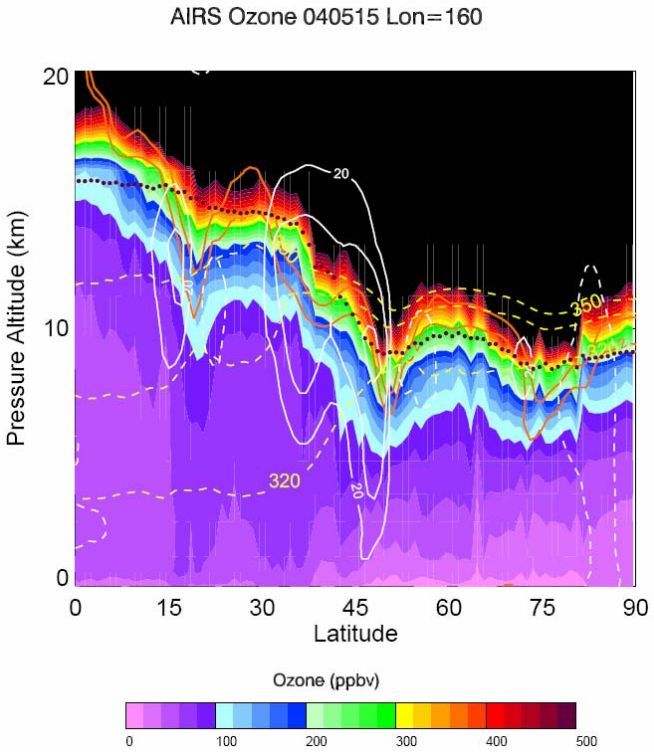


Figure 17 – A cross section of ozone data from the AIRS satellite instrument. Data are 1°x1° binned averages. The white contours represent the zonal wind, highlighting the subtropical jet and polar jet locations. The light yellow dash contours are potential temperature. Orange contours are 2 and 4 PVU potential vorticity. These meteorological fields are from 1°x1° degree and 26 level NCEP GFA data.

D. Concluding Remarks

While perhaps more questions than answers emerge from the above discussions the convergence of knowledge at the workshop was very useful in helping better define what is known regarding processes controlling the composition of the extra-tropical UTLS. Just as important, the workshop helped to identify the remaining outstanding questions.

It is clear that consistent use of well-defined terminology (c.f. STE=STT+TST) is imperative, so that disparate studies can be integrated for a larger-scale picture. In this regard there is especially a need to better understand the newly-identified ExTL. Given the complex thermodynamic and chemical state in this region, what metrics should be used to define the ExTL? What is its special role in the chemical, physical and dynamical state of the extra-tropical UTLS?

In some cases focused measurement campaigns would allow us to clarify which processes are significant to the extra-tropical UTLS region and therefore warrant more extensive study. For example, targeted measurements of aerosol composition in the northern hemisphere UTLS, in conjunction with satellite data analysis, could help determine how pyro-convection is influencing the chemical and optical properties of particles in this region. In-situ measurements could also be used to investigate, for example, the effects of short-lived bromine containing species transported to the UTLS.

Laboratory studies of reaction rates and heterogeneous ice cloud formation processes under conditions appropriate for the mid-latitude UTLS region are needed for more accurate model representation. Focused studies are also needed to understand how the coupling of dynamical processes over a range of scales control the chemical mixing and microphysical cloud formations in the extra-tropical UTLS (i.e.: How "mixed" is the air in this region?) and to improve our modeling capability in this region (i.e.: What processes are essential to include in order to represent the chemical and micro-physical state of this region?). While the importance of deep convection in this region is now recognized, the measurements needed to quantify its effect on a global scale remain to be identified.

Finally, there is a need to incorporate existing knowledge into models in order to assess regional and global-scale impacts on, for example, cirrus cloud formation. In particular, while some key species and processes are starting to be included CTMs and CRMs there is still the need to determine appropriate parameterizations for GCMs and CCMs. While models' predictions of STE across the extra-tropical tropopause have recently improved, large uncertainties in flux calculations still exist. New metrics must be found for validating these models against observations.

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Acronyms

AIRS – the Atmospheric Infrared Sounder on the Aqua satellite (http://www-airs.jpl.nasa.gov/)
Aura – One of NASA's EOS (Earth Observation System) satellites (http://eosdatainfo.gsfc.nasa.gov/eosdata/aura/mls/mls.html)
CARIBIC – Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container (http://www.caribic-atmospheric.com/)
CCM – chemistry-climate model (http://www.pa.op.dlr.de/workshops/CCMVal2005/)
CLAMS – Chemical Lagrangian Model of the Stratosphere
Crystal-FACE – The cirrus Regional Study of Tropical Anvils and Cirrus Layers – Florida Area Cirrus Experiment (http://cloud1.arc.nasa.gov/crystalface/)
EULINOX – The European Lightning Nitrogen Oxides Project (http://www.pa.op.dlr.de/eulinox/)
ExTL – Extratropical Tropopause Layer
GOME – instrument on the ERS-2 satellite for global monitoring of ozone (http://earth.esa.int/ers/gome/)
ICARTT – International Consortium for Atmospheric Research on Transport and Transformation (http://www.al.noaa.gov/ICARTT/)
MINOS – Mediterranean Intensive Oxidant Study
MLS – Microwave Limb Sounder on the Aura satellite (http://mls.jpl.nasa.gov/)
MOPITT – Measurements of Pollution in the Troposphere (http://terra.nasa.gov/About/MOPITT/about_mopitt.html) instrument on the TERRA satellite (http://terra.nasa.gov/About/)
MOZAIC – Measurement of Ozone and Water vapour by Airbus In-service Aircraft (http://www.aero.obs-mip.fr/mozaic/)
NOXAR – Measurements of Nitrogen Oxides and Ozone Along Air Routes

(http://www.iac.ethz.ch/en/research/chemie/tpeter/Noxar.html)
POAM II – Polar Ozone and Aerosol Measurements (http://wvms.nrl.navy.mil/POAM/poam.html)
POLARIS – field study; Photochemistry of Ozone Loss in the Arctic Region in Summer
PV – potential vorticity
SONEX – SASS Ozone and Nitrogen Oxide Experiment
STRAT – Stratospheric Tracers of Atmos. Transport (http://cloud1.arc.nasa.gov/strat/strat.html)
SPURT – SPURenstofftransport in der Tropopausenregion (Tracegas transport in the tropopause region; http://www.meteor.uni-frankfurt.de/spurt/)
STACCATO - Stratosphere-Troposphere exchange in a Changing Climate on Atmospheric Transport and Oxidation Capacity
STE – stratosphere/troposphere exchange
STERAO – Stratosphere-Troposphere Experiment - Radiation, Aerosols and Ozone (http://chill.colostate.edu/sterao.html)
STT – stratosphere-to-troposphere transport
TES – Tropospheric Emission Spectrometer instrument on the (http://tes.jpl.nasa.gov/), on the Aura satellite
TRACE-P –TRAnsport & Chemical Evolution over the Pacific field campaign (http://code916.gsfc.nasa.gov/Missions/TRACEP/)
TST – troposphere-to-stratosphere transport
TTL – Tropical Tropopause Layer
(O)VOCs – (oxygenated) volatile organic compounds
VSLS – very short-lived species: e.g., lifetime with respect to photochemical removal <~0.5 year
WCBs – warm conveyor belts
WMO – World Meteorological Organization

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Participants in the SPARC/IGAC Workshop on "Processes governing the chemical composition of the extra-tropical UTLS", taking a mid-day break from the workshop's very active discussions.

The Indirect Effects of Aerosols on Climate: *A report from the Specialty Conference co-sponsored by IGAC, NOAA, NASA and ACCENT*

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I. Introduction and background

A conference on The Indirect Effects of Aerosols on Climate was co-convened by IGAC, NASA, NOAA, and ACCENT in an attempt to address the state of knowledge of the indirect effects of aerosols, describe the current limitations of our understanding, and suggest areas where future research would be most useful. Toward this end a total of 22 invited papers and 39 posters were presented in the four subtopics:

1. Small-Scale Observations and Modeling
2. Large-Scale Observations and Modeling
3. Meteorological Effects and Constraints
4. Ice Clouds

These sessions were organized by Organizers Daniel Murphy (NOAA Aeronomy Laboratory, USA), Bruce Wielicki (NASA, USA), Ulrike Lohmann, (formerly of Dalhousie University; currently at ETH Zürich, Switzerland), and Tom Choularton (Univ. of Manchester, U.K.).

Some key points made by the speakers in each session as well as the important questions they posed for consideration are documented below. Two publications [Lohmann et al., 2005; McFiggans et al., 2005] stemming from break-out discussions at this conference have been submitted for peer-reviewed journal publication and are synopsized in the following two articles.

II. Workshop discussions

One common thread that ran through many presentations at the workshop was the number of complicated interactions between the ways aerosols affect clouds and the responses of the clouds themselves (Figure 1). Lohmann and Feichter [2005] outlined six general means by which the indirect effect acts on climate. The first three have been known for some time whereas the latter three, which acknowledge our dynamic understanding of the interaction between particles and clouds, are less familiar in the context of discussing aerosol indirect effects:

1) The First Indirect Aerosol Effect: In clouds with fixed water amount, more particles lead to more numerous but smaller cloud drops that reflect more solar radiation. Also known as the cloud albedo or Twomey effect.
2) The Second Indirect Aerosol Effect: Smaller cloud drops can lower precipitation efficiency and prolong

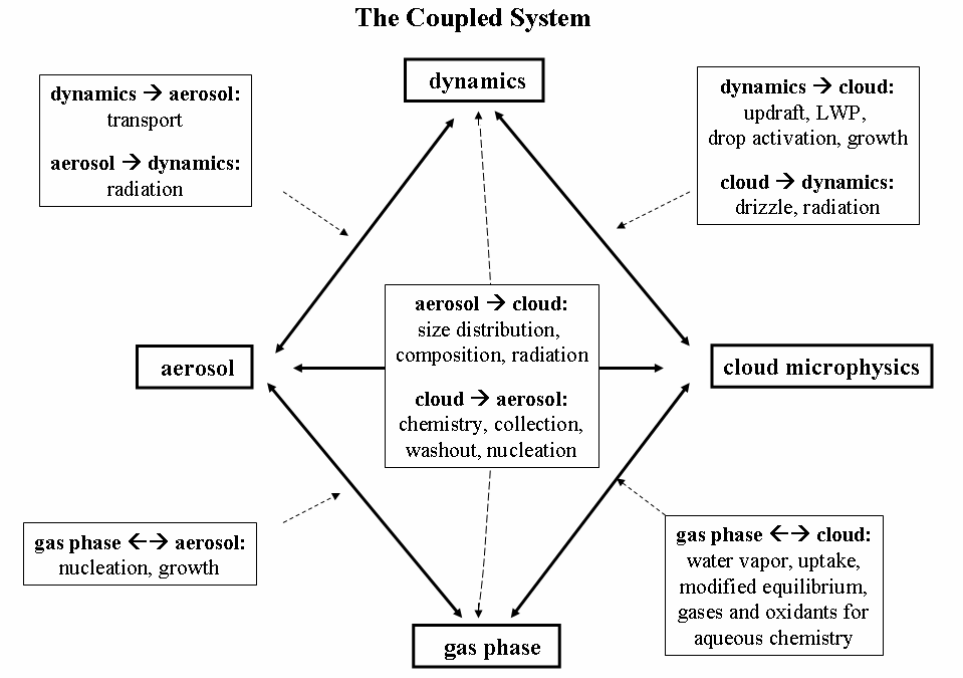

Figure 1 – The coupled system of aerosols, dynamics, microphysics, and the gas phase. Courtesy *G. Feingold*.

cloud lifetime. Also known as the cloud lifetime or Albrecht effect.
3) The Semi-Direct Effect: Absorption by particles such as soot leads to cloud evaporation. Also known as cloud burning or the Hansen effect.
4) Glaciation Effects: An increase in ice nuclei (IN) number lead to an increase in precipitation efficiency.
5) Thermodynamic Effects: In convective clouds, smaller cloud droplets lead to a delay in homogeneous freezing and latent heat release.
6) Surface Energy Budget: Increased aerosol and cloud optical thickness decreases net surface radiation.

In practice these effects are often linked. V. Ramaswamy noted in his opening remarks that although it may be "… useful to compartmentalize the different facets … it may be more profitable to think of the TOTALITY of the aerosol effects, including even the direct forcing", as in conditions of very high humidity the direct/indirect aerosol effect boundary is not distinct, particularly if partially soluble aerosol is present. In some cases, multiple indirect effects may be present simultaneously, and there may be feedbacks between aerosol-cloud interactions and the atmospheric thermodynamics/meteorology.

These complex interactions can even change the sign of aerosol-cloud effects beyond the negative forcing given by the Twomey effect. Graham Feingold, Joyce Penner, and Keith Shine all noted from both their own work and that of others that the sensitivity of cloud fraction to black carbon can change sign depending on whether the black carbon is below or above the cloud. Andy Ackerman and Graham Feingold noted that drizzle formation is linked to entrainment and this link can weaken, or even reverse, the second indirect effect. The second indirect effect is also strongly modulated by the number of giant cloud condensation nuclei (CCN), regardless of the characteristics of the size distribution at smaller sizes.

In Alexander Khain's numerical model, increased aerosol always delayed precipitation but the total amount of precipitation could either decrease or increase depending on whether single clouds or cloud systems were considered in the analysis (Figure 2). The sign of the effect of aerosols on the total precipitation could change depending on whether continental or maritime conditions were considered although a delay in precipitation for both cases was likewise predicted. Tom Choularton presented model results on the sensitivity of deep convection, specifically Hector thunderstorms, to changes in aerosol. Although there were changes in the modeled evolution of the system in response to the number of particles, the functional form was complicated and non-monotonic.

Regarding the second indirect effect, Andy Ackerman's review showed that despite evidence that ship tracks contain smaller cloud drops recent studies have failed to find evidence of the simplistic prediction that smaller drops should increase liquid water content by reducing drizzle. In addition, Greg Ayers showed that in Australia the strongest evidence for aerosol-induced changes in cloud properties have been in clouds that had not been expected to produce precipitation regardless – i.e. with or without anthropogenic particles present.

1. Small-Scale Observations and Modeling

Key Points:

Although it has been known for several decades that aerosols and clouds are interrelated [e.g. Squires, 1966] our understanding of interactions beyond the first indirect effect remains, in many areas, poor. The inherent complexity of aerosol/cloud interactions must be reflected in

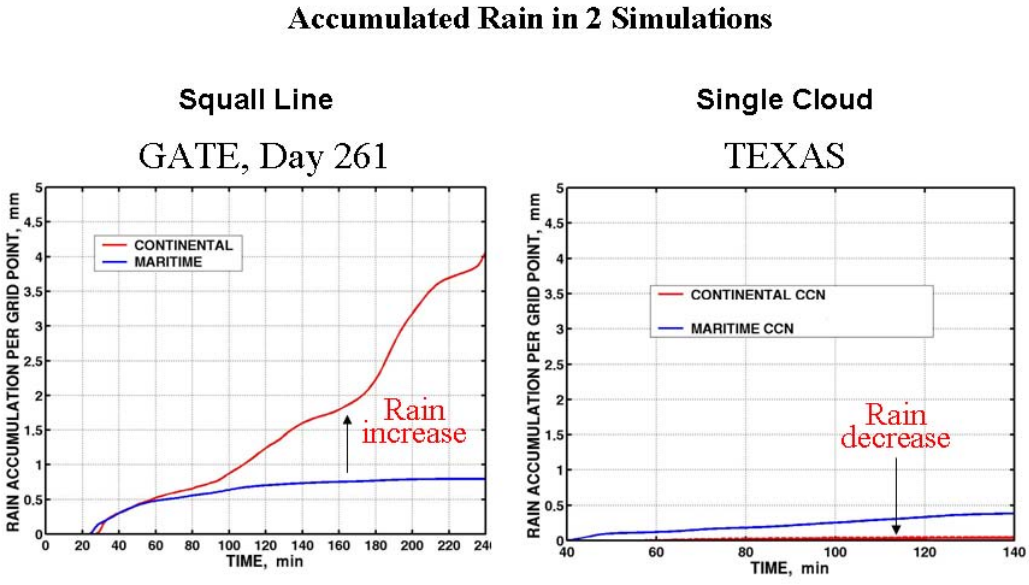

Figure 2 – Two different cases of the change in precipitation in a regional cloud model on changing from maritime (few) to continental (many) aerosols. Courtesy *A. Khain.*

models if they are to accurately predict future indirect effects. Our understanding is further complicated by the fact that it is difficult to determine the natural baseline of either aerosols or clouds.

Analyses show that the majority of atmospheric particles are internal mixtures of sulfates, often neutralized to some extent by ammonia, and organics. A myriad of other particle types, such as sea salt, biomass burning, soot, and mineral dust, are also present in the atmosphere. The organic fraction of particles, in particular, remains an area of large uncertainty both in its nature and its role in aerosol/cloud interactions. This has resulted in difficulty in treating aerosols correctly in models due to the very different physical properties of species that could be found in the particle phase. Closure experiments between aerosols and cloud condensation nuclei (CCN) have highlighted the uncertainties due to organic species in particles.

One caution is that although we are beginning to understand some aspects of aerosol/cloud interactions we should not allow public pronouncements to move too far ahead of evidence and understanding or the scientific credibility of the research community is endangered.

Outstanding Issues:

• What is the role of giant CCN in precipitation? What are their sources and composition?
• A lack of sufficient knowledge about the organic component of atmospheric aerosols continues to limit our ability to understand and model aerosol/cloud interactions. Further research on water activity and accommodation coefficients at relevant temperatures is required.
• The vast majority of our observations of aerosol properties have occurred in the Northern Hemisphere, where there is a heavier anthropogenic influence. The composition of the anthropogenic and natural aerosol mix, as well as the thermodynamic conditions, may differ significantly in anthropogenically influenced Southern hemisphere regions.

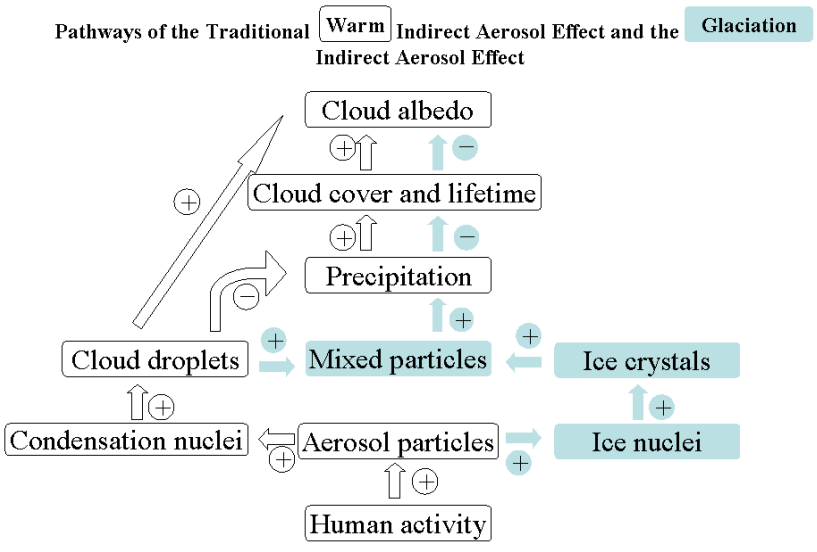

Figure 3 – The relative importance and sign of traditional warm and glacial indirect aerosol effects. Modified from Lohmann [2002]. Courtesy *U. Lohmann*.

2. Large-Scale Observations and Modeling

Key Points:

Satellite instruments, such as MISR, TOMS, AVHRR, and MODIS, have been utilized to provide information on the first indirect effect and to constrain the second indirect effect in certain cases, such as for ship tracks, smoke plumes, and contrails. These results have been validated through data comparisons, for example to AERONET sites. Although cloud screening remains a central issue for retrievals, satellites are our only tool to provide a global perthat appropriate locations and observation schemes are required to observe the different indirect effects, and multiple techniques (platforms and instruments) are required to understand biases.

Models can be used to estimate the indirect effect and to study the main sources of uncertainty in aerosol cloud-interactions. To do this they must incorporate both warm, mixed, and ice clouds (Figure 3). As models move to more physically based parameterizations with predictive capability they need more information about both aerosols and the atmospheric conditions. Needed parameters include aerosol number, size distribution and composition, and probability distribution functions of updraft velocity. Joyce Penner showed global model estimates of indirect forcing that found that indirect forcing became less negative when the mean aerosol diameter was either increased or decreased. If the model base case (Figure 4) is near the maximum response then incorporating more local detail will most likely reduce the global estimate. Perhaps not surprisingly, different parameterizations of cloud droplet number have a significant effect on global estimates of the indirect effect. Especially in the case of continental outflow over oceans, aerosol concentrations may be correlated with cloudiness because of the

water vapor content of the air, making it difficult to distinguish aerosol indirect effects from differences due simply to airmass water content.

The possible impact of changes in solar radiation and cosmic rays are poorly understood. Feedbacks between solar irradiance fluctuations and climate forcing are possible but the mechanisms are not known. Impacts of cosmic rays are also possible and theories exist as to how the ionizing property of the cosmic ray could influence cloud formation. However, published correlations between low cloud cover and cosmic ray intensity have not held up after comparison with the longer data sets now available.

Outstanding Issues:

• Further validation of remotely sensed aerosol indirect effects are needed. This validation can only be accomplished with coordinated studies using satellites, in situ and surface observations, and models.
• More studies are needed where the observations are sufficient to decouple the underlying causes of observed correlations between aerosols and clouds.
• The sources of discrepancies between models' representation of the indirect effect need to be understood. A model comparison study is underway to sharpen our understanding of the reasons for different estimates of the radiative forcing due to aerosol indirect effects.

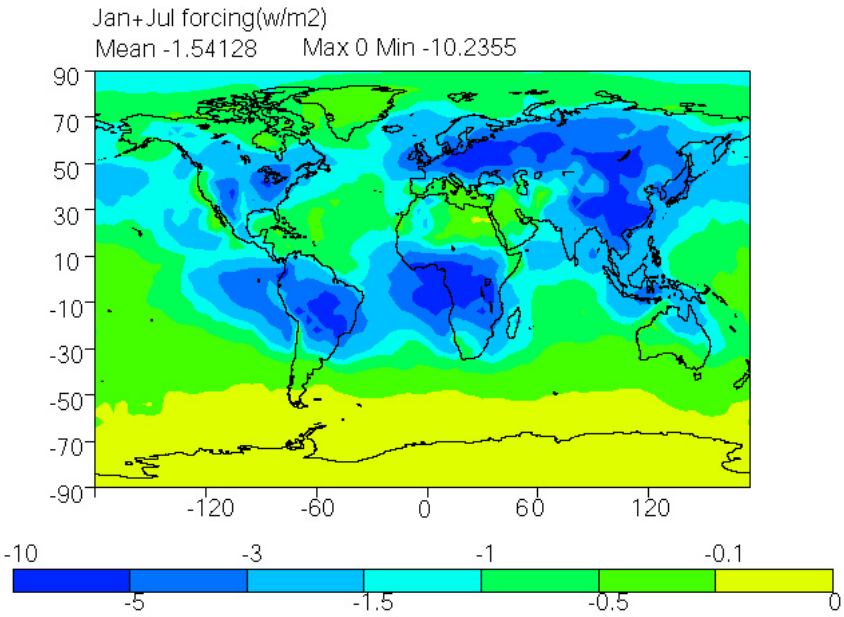

Figure 4 – Spatial distribution of the radiative forcing from the indirect effect in one GCM. Modified from Chen and Penner [2005]. *Courtesy J. Penner*.

3. Meteorological Effects and Constraints

Key Points:

Changes to either the droplet number concentration (the first indirect effect) or introducing absorbing particles starts a cascade of responses in a cloud system. These changes are strongly coupled and it may not be possible to decouple the 'cloud lifetime effect', for example, from the response of the cloud system. The response of the cloud system can furthermore be very different than the response of an individual cloud. Some examples follow. In marine stratocumulus individual cloud cells are constantly being replaced and this alters the response of the system to aerosols. As discussed by A. Ackermann, entrainment at the top of clouds is important enough that the humidity of the air immediately overlying the cloud has been shown to control the sign of the eventual response of the clouds to aerosols. The suppression of drizzle does not always increase the liquid water content of the cloud system because of dynamical feedbacks to drizzle in the boundary layer.

The semi-direct effect is probably highly dependent on the altitude and distribution of absorbing aerosols. It is also very different for different cloud regions, such as marine stratus or tradewind cumulus. This variation means that the temperature response to radiative forcing is highly variable, or perhaps even ill-defined, for absorbing aerosol.

Amazonia is very likely an excellent test area for studying the indirect effect. There is more than an order of magnitude difference in aerosol mass from wet to dry season, the transition between the two occurs in a period of weeks, and biomass burning aerosol dominates during large periods of the year. If effects are not apparent under these circumstances then they may be impossible to detect under other conditions.

Outstanding Issues:

• Does it make sense to separate the second indirect effect from cloud feedbacks?
• Do we know the vertical and regional distribution of absorbing aerosol well enough to estimate and constrain the semi-direct effect?
• Do GCMs have an adequate representation of cloud properties to capture the boundary layer dynamical responses seen in cloud-resolving models?
•Do GCMs have adequate representation of cloud properties to model the effects of absorbing aerosols?

• Global models are reasonably well constrained if the sulfate/greenhouse ratio remains constant. As this ratio changes in the future models will need to rely more on physically based predictions.

4. Ice Clouds

Key Points:

It is known that the ice phase plays an important role in global precipitation. Furthermore, satellite instrumentation shows that thin cirrus ice clouds cover approximately one third of the globe and can thus exert a considerable radiative effect. In recent years, field studies, model capability, and remote sensing techniques have increased our understanding of the interaction of aerosols with ice clouds. The ice phase is formed by two distinct mechanisms: 1) homogenous freezing when aqueous particles reach temperatures below –38° C and saturations near liquid water or 2) heterogeneous freezing at higher temperatures and/or lower saturations if chemically distinct inclusions known as ice nuclei are present.

Laboratory and field studies show that our understanding of homogeneous freezing by the ubiquitous atmospheric sulfate aerosol is sound. Some uncertainties remain, specifically the impact of surface tension and mass accommodation coefficient, particle phase transformations, and the effect of organic content, but these are currently being investigated. The indirect effect attributed to homogeneous freezing has been extensively modeled and is generally believed to have a weak response to possible perturbations.

The role of heterogeneous freezing is less well known. Cirrus clouds, especially thin sub-visible cirrus, are probably highly sensitive to IN and thus anthropogenic changes. In most, but not all cases, an increase in IN reduces the number of ice crystals and thus cloud albedo. As illustrated in Figure 5, IN have been shown to have a very specific chemical composition when compared to the average aerosol. Specifically, mineral dust and industrial particles are enhanced in the IN population. The overall impacts on IN from anthropogenic sources remains uncertain. Effects from periodic events, such as dust storms, also remain unresolved. Ice multiplication from the breakup of crystals also remains a complex and largely unknown topic.

Satellites have proven increasingly capable of elucidating ice cloud properties. Cirrus coverage values have been provided by remote sensing and the current generation of instrumentation has the potential to define cirrus parameters from space such as size, shape, scattering, and ice water path. Continued inter-comparison campaigns using surface, airborne, and satellite platforms, such as during the Central Equatorial Pacific Experiment (CEPEX), are required to validate these data.

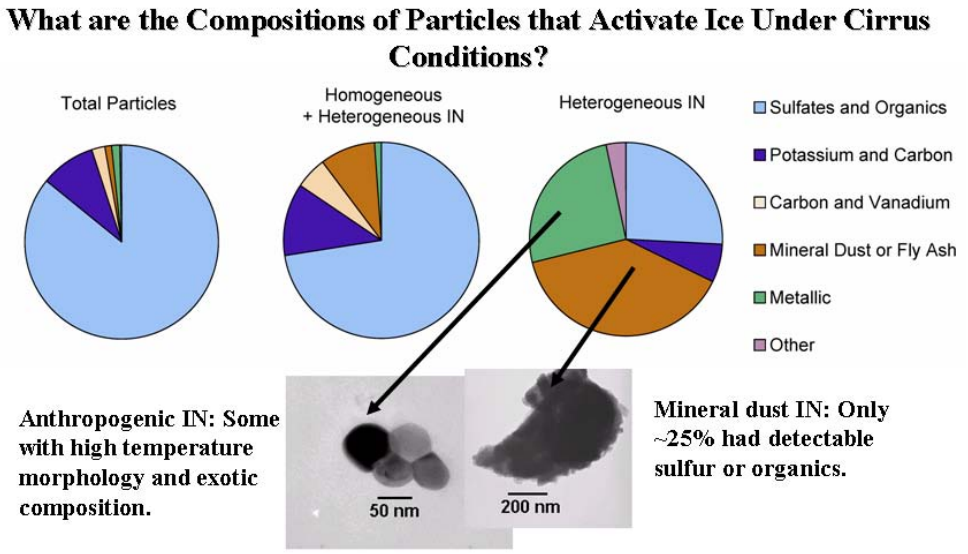

Figure 5 – Chemical composition of atmospheric aerosols and of the subset that activate freezing. Modified from DeMott et al. [2003]. *Courtesy P. DeMott.*

Outstanding Issues:

•What are the nuclei for ice cloud formation and how well do we understand anthropogenic changes in IN?

•What observations are best suited to provide an understanding of cirrus cloud changes? (long term remote sensing? *in situ*?)

•What types of models are required? What are the model limits? Can we define the best situations in which to compare models and observations of ice clouds?

•Can these observations lead to a better definition of the radiative budget from ice clouds (a key uncertainty)?

III. Concluding remarks

An apt concluding remark for the IGAC Specialty Conference on Indirect Effects of Aerosols on Climate is

paraphrased from Graham Feingold's paper: 'Can we identify the effects with big leverage?' An ongoing challenge to each participant of the conference is to seek to determine the most important topics to address in each of our areas of specialty. It is generally acknowledged that of all the indirect effects our knowledge of the first is the most comprehensive. This is due in part to this being the least complex of the indirect effects but also to this being the subject of a large commitment of resources – both observations and modeling.

Some 'big levers' that can be culled from the conference:

1) There can be complicated dynamical responses to almost any change in cloud properties. These significantly change the magnitude and sometimes even the sign of indirect effects beyond the change in cloud droplet number.

2) Small-scale models seem to consistently show that aerosol indirect effects are closely coupled to entrainment at the cloud top and changes in surface energy fluxes. What observations are needed to test the effects of entrainment and surface fluxes?

3) What are the organic species found in atmospheric particles and how do they effect aerosol/cloud interactions?

4) Attempts to 'break' models are invaluable in showing the right answers are obtained for the right reasons.

5) Estimation and constraint of the overall indirect effect can only be accomplished by a combination of satellites, aircraft, surface sites, and models. Well chosen locations for comparison and integration of these data sets are critical.

6) GCM inter-comparisons are needed to understand the reasons why estimates of the indirect effect differ.

7) An improved understanding of the vertical distribution of absorbing aerosol and detailed treatment in models is required if we are to understand the semi-direct effect.

Acknowledgements

We would like to thank all of the conference speakers, poster presenters and attendees. Special thanks to Cathy Burgdorf, Suzie Milano-Schoser, and Kathy Thompson for their work on conference organization.

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Workshop highlight:

"Can we improve model estimates of the anthropogenic indirect aerosol effect?"

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Summarized from:

Lohmann, U., Feichter, J., Kinne, S., and Quass, J., Approaches for constraining global climate models of the anthropogenic indirect aerosol effect, *submitted to Bulletin of the American Meteorological Society,* 2005.

The anthropogenic indirect aerosol effect cannot be deduced from observations alone because the satellite record is not long enough, while other existing long-term records do not provide the aerosol and cloud microphysical properties needed for such an assessment. Also, current satellite data are limited in such that aerosols can only be retrieved in cloud-free air so that aerosol and clouds cannot be observed simultaneously. Finally, no convincing method exists so far to distinguish natural from anthropogenic aerosols by remote sensing methods. Therefore, estimates from global climate models need to be combined with satellite data.

Three different avenues can be taken to improve and validate current climate models. The first avenue is to evaluate the model predictions of aerosol and cloud properties with available measurements. The problem here is that the spatial and temporal resolution need to match the observed resolution. Also, the initial and boundary conditions need to be known with sufficient accuracy. This approach can best be carried out with

process models and can be used to understand field data (Figure 1). An example is the ability of organic aerosols to act as CCN. In different geographical areas it was found that organics can increase the number of cloud droplets but due to different reasons (O'Dowd et al. 2004; Lohmann and Leck 2005). In the Arctic Ocean this increase in cloud droplet number arises from the ability of the organic aerosols to lower the surface tension whereas off the coasts of Ireland the cloud droplet increase is attributed to the addition of a new source of marine organics, mostly insoluble, active during phytoplankton blooms. These effects can then be taken into account in sophisticated parameterizations of the aerosol activation process as, for instance, developed by Nenes and Seinfeld (2003); Abdul-Razzak and Ghan (2005).

The second approach is the data assimilation avenue (Figure 2). Data assimilation uses both models and observations for initialization and therefore is not entirely independent from models. However, it can provide information of model deficiencies and measurement uncertainties especially in data rich areas. Assimilated climate models that require large model adjustments in certain areas or under certain conditions, offer obvious clues to poor representations of the adjusted model quantity. There is a European initiative (GEMS; Global and regional Earth-system Monitoring using Satellite and in-situ data; http://www.ecmwf.int/research/EU_projects/GEMS/) that will use follow this avenue as well as the US initiative PARAGON (the Progressive Aerosol Retrieval and Assimilation Global Observing Network; Kahn et al. 2004).

The third approach uses different statistical relationships of variables that are relevant to aerosol-cloud interactions (Figure 3). The rational here is that statistical correlations are temporally and spatially more robust than individual measurements. The correlations can be used to test the relevant process and they can help to gain a better understanding. This approach also has the advantage that it works with data from field campaigns, with satellite data as well as with long-term data from surface networks, such as the Baseline Surface Radiation Network (BSRN), the Aerosol Robotic Network (AERONET), and the European Aerosol Research Lidar Network (EARLINET).

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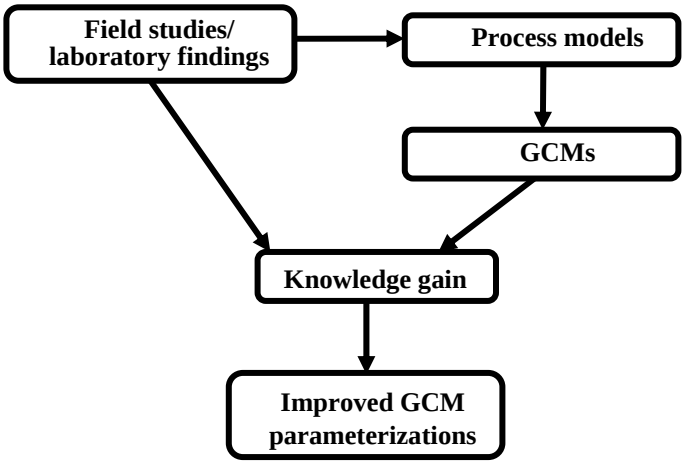

Figure 1 – Schematic of the detailed "model plus observations" avenue.

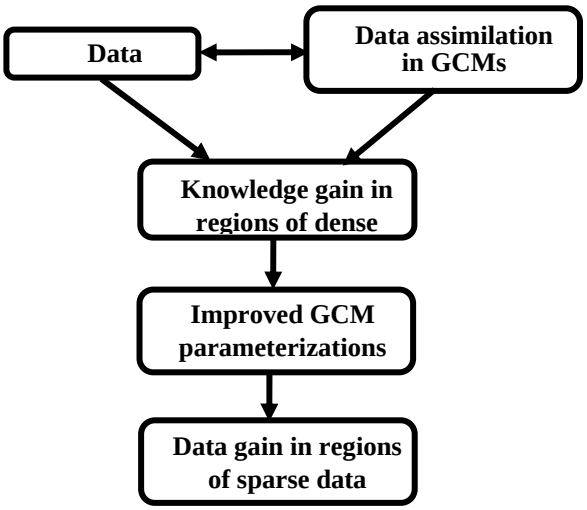

Figure 2 – Schematic of the data assimilation avenue.

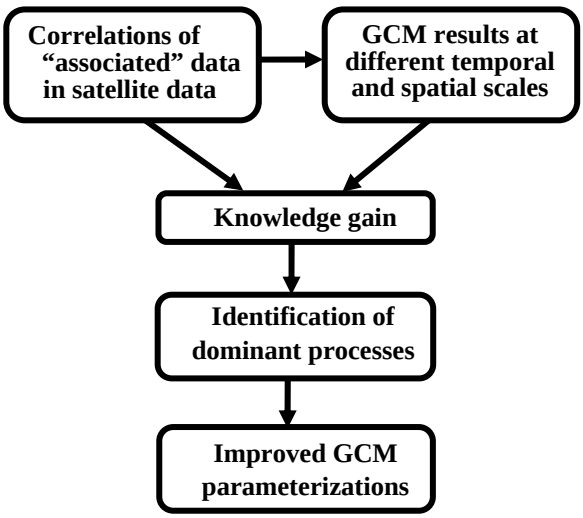

Figure 3 – Schematic of the model constraint avenue.

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Workshop highlight:
"The Indirect Effect in Warm Clouds"

Contributed by **Gordon McFiggans** (g.mcfiggans @manchester.ac.uk), *Atmospheric Sciences Group, SEAES, Univ. Manchester, Sackville St., Manchester M60 1QD, U.K.*

Summarized from:

McFiggans, G, P. Artaxo, U. Baltensperger, H. Coe, M. C. Facchini, G. Feingold, S. Fuzzi, M. Gysel1, A. Laaksonen, U. Lohmann, T. F. Mentel, D. M. Murphy, C. D. O'Dowd, J. R. Snider, and E. Weingartner, The Effect of Physical & Chemical Aerosol Properties on Warm Cloud Droplet Activation, *Atmos. Chem. Phys. Disc., 5,* 8507-8646, 2005.

The above paper resulted from discussions during the breakout session on The Effect of Physical & Chemical Aerosol Properties on Warm Cloud Droplet Activation. It is currently open for comment in Atmospheric Chemistry & Physics Discussions.

The effects of atmospheric aerosol on climate forcing may be very substantial but are quantified poorly at present; indeed, the indirect effects are credited with the greatest range of uncertainty amongst the known causes of radiative forcing. The manuscript explores the effects that the composition and properties of atmospheric aerosol can have on the activation of droplets in warm clouds, potentially influencing the magnitude of the indirect effect.

Atmospheric aerosol size distributions are highly variable and both the number of particles in a given size range and the gradient of the distribution in certain critical size ranges will determine activation behavior. Size distribution characteristics strongly interact with the dynamics to determine the number of activated droplets and these interactions are dependent on the composition distribution. The sensitivities of cloud droplet size and number to the aerosol size, composition and updraught are not equal and are interdependent, though number always appears important. Inorganic electrolyte components of atmospheric aerosols are readily hygroscopic and evidence suggests that organic components contribute only slightly to growth of aged particles. Accumulation mode particles are generally more hygroscopic than Aitken mode particles and the minimum size of particles activated to form cloud droplets is often near the border between these two particle modes. There is some trend of increasing fraction of readily hygroscopic particles, increasing growth factors and decreasing growth spread from anthropogenically influenced to remote areas. External mixtures of particle modes with respect to water uptake and growth spread within modes are nearly ubiquitous.

Measurement requirements

No single measurement technique can provide the required size segregated aerosol composition information to enable prediction of cloud activation. However, a combination of online and offline techniques can deliver highly time- and space-resolved chemical composition and mixing state information for cloud parcel model input. The measurement requirement for estimation of the aerosol effect on droplet activation is size-resolved chemical and physical property data for particles in the 40 to 200 diameter range. Given the complexity of organic components, it is unlikely that a detailed understanding of their effects on surface tension, uptake kinetics and equilibrium water content will be reliably predictable from composition measurements alone; hence direct measurements of physical properties are also required at the sizes near the cloud activation threshold. Two such useful derived properties are the hygroscopic growth factor and the CCN activation spectra. Direct surface tension measurements are also useful. A comparison of activation spectra between those measured in CCN instruments and those derived from other measured properties such as composition may provide reliable data sets for cloud model initialization and reveal limitations of the measurements. The duration of particle exposure to the maximum supersaturation in such measurements should mimic that which occurs during activation in an updraft. Flow-through cloud simulator experiments together with field-deployable aerosol and CCN instrumentation could provide important guidance on the effects of humidity preconditioning on aerosol activation, the reliability of field-deployable CCN instrumentation and the effects of complex aerosol composition on activation.

Effect clarification

Conclusions drawn concerning the effects of component properties include:

i) Limited component solubility alone is of limited effect in real atmospheric aerosol activation.

ii) The degree to which a surfactant partitions between the bulk and the surface of the droplet depends on the dominant surface active components. It should be noted that solely reducing surface tension would worsen overprediction of CCN and droplet numbers commonly found in closure studies. If surfactant films are present, they are probably more likely to be in the expanded than condensed state and are unlikely to influence hygroscopic or CCN properties greatly by mass transfer limitation. It is critically important to determine whether ordered organic films truly exist on growing cloud droplets, the significance of surface partitioning effects or whether surface tension suppression predominantly results from WSOC. Surrogates for representing atmospheric aerosol must be chosen with caution.

iii) Cloud model studies have shown that the effects of nitric acid and other soluble trace gases on droplet activation depend heavily on the kinetics of cloud formation and cannot be explained with Köhler theory alone (see dynamical considerations, below).

vi) The primitive form of the Köhler equation can circumvent requirement for unknown composition related parameters but requires laboratory-determined or predicted water activities and surface tensions.

Dynamical considerations

Köhler theory suggests that the potential effects on droplet activation of aerosol compositional complexity are very large, yet such differences are not realized when considering the dynamical system of a population of aerosol growing and competing for water vapor. Composition effects are reduced by feedbacks in the dynamical system, particularly under low aerosol loadings, though less so under polluted conditions with low water vapor supply. Kinetic limitations on droplet activation result in larger activated aerosol fractions. Natural variability in aerosol size distributions leads to increases in aerosol number that do not necessarily translate to increases in drop concentration. Coupled growth models are required to assess these effects; the greatest deviation of such predictions from equilibrium exist for the smaller particles where composition effects are most important.

Synthesizing system complexity

The complexity of aerosol behavior varies significantly between locations; how cloud activation behavior is affected by this complexity must be established to enable such considerations to be captured in large-scale models. The current difficulties in describing aerosol and aerosol-cloud interactions in global models leads to a range of predicted radiative forcing. In spite of the best efforts to explain the relationship between aerosol size/composition and drop size distributions, current climate predictive capabilities are limited by the difficulty of resolving convection and clouds in GCMs. Marine boundary layer clouds, such as stratocumulus, present a challenge because they are not well resolved in GCMs, and because of their large albedo contrast with the dark underlying ocean, changes in these clouds represent a significant climate forcing. Given the complexity of the system, it is crucial to evaluate the extent to which details of aerosol-cloud interactions are important for the overall understanding of their impact on climate. Useful questions that should be posed include:

i) is detailed aerosol size distribution and composition always required or are there adequate representations that are easier to use (e.g. how closely can the effects of composition be described simply with a hygroscopic growth factor or insoluble fraction in addition to the size distributions)?

ii) How many of the complex interactions are important, and at what spatial/temporal scales?

iii) Can we synthesise our understanding of the complexity into physically-based parameterisations that capture the essence of the process under consideration and will such parameterisations represent adequately the underlying physics when included in global scale models?

Addressing Temporal/Spatial Scales

An important issue for both observations and models is the question of scale. Current climate models do not incorporate the small temporal/spatial scales needed for adequate resolution of many processes, as is exemplified by aerosol-microphysics-chemistry processes. Predicting the impact of the aerosol indirect effect is an enormous challenge, requiring that models correctly predict the collocation and timing of aerosol and cloud events, as well as the interactions between aerosol and clouds at small scales. Observations and modeling should, as a first step, be performed at the scale appropriate to the process or interaction under investigation. Observations and model output need to be compared at similar scales, starting from the smallest scales pertinent to the process, ranging up to the regional and global scales. A methodology for consistent transfer of understanding and representation of processes from the smallest to largest scales needs to be developed. This could consist of embedding detailed models in climate models based on the principles of synthesis and parameterization outlined above or it could mean increasing the spatial and temporal resolution of the climate models to resolve the processes themselves.

It is therefore apparent that, whilst significant progress has been made in understanding the potential mechanisms by which aerosol properties may effect cloud activation, much further work must be carried out to establish the magnitude of such effects and their importance in climate prediction.

ANNOUNCEMENTS

IGAC's 9th Open Science Conference, held jointly with CACGP and WMO September 17-23, 2006 Cape Town, South Africa

http://www.Atmosphericinterfaces2006.co.za

IGAC's 9th Open Science Conference will be held jointly with CACGP (the Commission on Atmospheric Chemistry and Global Pollution) and WMO (the World Meteorological Organization).

"Atmospheric Chemistry at the Interfaces", is the theme for the conference, which will highlight the current state of knowledge of the interaction between various components of the Global System. This theme represents the common interests of the three sponsors, and focuses on the great challenges of interdisciplinary research and effective cross-disciplinary communication in times of ever increasing specialization.

1st iLEAPS Open Science Conference

January 21-26 2006

Boulder, Colorado, USA

http://www.atm.helsinki.fi/ILEAPS/boulder/

The iLEAPS Scientific Steering Committee invites you to participate in the 1st iLEAPS Open Science Conference.

The conference will highlight the relevant aspects concerning the interface between land-biosphere-atmosphere. In particular we will focus on four main topics:

- Land-atmosphere exchange of reactive and conservative compounds - key interactions and feedbacks in the earth system,
- feedbacks between land biota, aerosols and atmospheric composition in the climate system,
- feedbacks and teleconnections in the land surface-atmosphere-water-system, and
- transfer of material and energy in the soil/canopy/boundary-layer system: measurements & modeling.

Global Environmental Change: Regional Challenges
An Earth System Science Partnership
Young Scientists' Conference *November 7-8 2006*
Global Environmental Change OSC *November 9-12 2006*

Earth System Science Partnership

Beijing, China

http://www.essp.org/essp/ESSP2006/

This Conference provides the opportunity for the presentation of advances in our understanding of the physical, biogeochemical, biodiversity and human dimensions aspects of global environmental change and to highlight the ESSP approach to study of the Earth System. The International Young Scientists' Global Change Conference offers a prestigious platform for young scientists to present their research findings to leading scientists in the field. It is intended to stimulate competition, encourage excellence, reward outstanding performance and foster the development of personal and institutional networks. The conference precedes the Earth System Science Partnership's Open Science Conference, and it is expected that all the young scientists will participate in the Open Science Conference.

2nd SOLAS Open Science Conference

surface ocean solas lower atmosphere study

March 6-9, 2007

Xiamen, China

http://www.uea.ac.uk/env/solas/ss04.html

SOLAS is very pleased to announce that the next SOLAS Open Science Conference will be held in Xiamen, China, 6-9 March 2007. The Conference will be hosted by Guang Yu Shi (Inst. Atmospheric Physics) and Minhan Dai (Xiamen U.) and and will follow on from the successful meeting in Halifax in 2004.

The IGAC Scientific Steering Committee at its 20th annual SSC meeting, 11-14 October 2005. The SSC was graciously hosted by Laura Gallardo Klenner and the Centro de Modelamiento Matemático, Universidad de Chile, Santiago.

Pictured (left to right): Uli Platt (Germany), Graciela Raga (Mexico), Kathy Law (France), Phil Rasch (USA), Eric Wolff (U.K.), David Parrish (USA), Laura Gallardo Klenner (Chile), Randall Martin (Canada), John Burrows (Germany), Sandro Fuzzi (Italy), Stuart Piketh (S. Africa), Leonard Barrie (Switzerland), Kevin Noone (Sweden), David Lowe (New Zealand), Anne Thompson (USA), Sarah Doherty (USA), Makoto Koike (Japan), Achuthan Jayaraman (India), Yongfu Xu (China). Missing: Shaw Liu (China), Ilia Ilyin (Russia), Ulrike Lohmann (Switzerland)

IGACtivities Newsletter

Taipei TAIWAN

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