CLIMATE CHEMISTRY INTERACTIONS
Report from the joint SPARC/IGAC workshop
3 - 5 April 2003 Giens, France

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Climate change is one of the most important societal concerns for the 21st century. Atmospheric chemistry plays a critical role in climate by controlling the abundances and distributions of natural and anthropogenic agents such as greenhouse gases, aerosols, and clouds, which influence incoming and/or outgoing radiation, temperature, and precipitation. Conversely, climate affects the chemical and physical processes that determine atmospheric composition through changes in temperature, water vapor abundance, short wave radiation, and other factors. These interactions between climate and physical and chemical processes are intricate, can be non-linear and often involve feedbacks, so a detailed level of understanding is needed to deal with the climate-physics-chemistry system. Elucidation of chemical processes will be key to proper societal decisions on how to influence future climate.

The atmospheric constituents – gases, aerosols, water vapor and clouds – are closely coupled through chemical processes as well as via dynamics and radiation. Interactive processes, including feedbacks, provide the interconnectivity between sources and climate, as represented in the top part of Fig. 1. This coupling influences their abundances and properties. Therefore, these interacting components have to be understood both individually and as an ensemble in order to understand and predict how each of the constituents will affect climate and climate change. In particular, good policy decisions rest on understanding how changes in these species' sources will affect climate.

An example of the effect of chemistry on climate is the influence of anthropogenic aerosols, which are potentially as important as greenhouse gases for current climate change. Aerosols scatter and absorb sunlight (the so-called direct effect), thereby altering the amount of atmospheric radiation that is absorbed in the atmosphere and at earth's surface. The direct effect depends critically on the chemical composition and mixing state of aerosols. Aerosols can also have indirect effects via interaction with clouds through their role as cloud condensation nuclei (CCN). In turn, clouds can modify aerosols, altering their optical properties, size distributions, and ability to act as CCN. These indirect effects, which are strong functions of the chemical and physical properties of the aerosols, can perturb clouds and even the hydrological cycle, two pivotal components of the climate system.

Changes in climate can also affect atmospheric chemistry significantly. For example, a change in water vapor abundance can alter the ability of the atmosphere to oxidize trace gases. A change in temperature or water vapor abundance can modify the chemical and

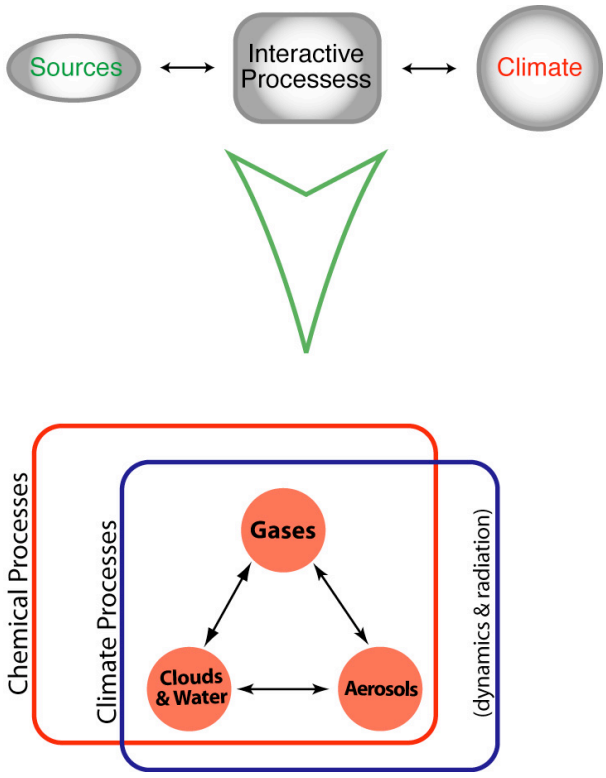

Figure 1. Schematic of the joint role of gases, clouds and water vapor, and aerosols in climate and chemical processes which allow anthropogenic and natural sources to influence climate.

physical properties of aerosols and can change the rates of chemical transformations in the atmosphere. Temperature and precipitation changes can also affect emissions from the surface. Biotic emissions will change as ecosystems shift, and atmospheric mineral dust loading may change with increased desertification or with changes to the meteorological systems that loft the dust. These interactions and feedback processes are complex and poorly understood.

Currently, there is a great deal of attention being given to short-lived species (such as black carbon, or soot) because of the possibility of a "quick return" as a result of some policy action. Furthermore, these short-lived species are pollutants that need to be addressed for human health and other concerns. Therefore, as shown in Fig. 1, clear understandings of the processes that connect sources (i.e. emissions and precursors) to abundances and the processes that connect the abundances to the climate forcings are essential for an accurate prediction of the future climate and an assessment of the impact of climate change and variations on the earth system. However, because of the variability in space and time for short-lived species, even the current contributions to the climate forcings are not easily evaluated using atmospheric observations alone; modeling

calculations are required.

The upper-troposphere and lower-stratosphere (UTLS) provides a good example of coupling in the atmosphere as far as composition and climate interactions are concerned. This coupling occurs because:

- Radiative forcing by greenhouse gases such as water vapor and ozone is especially sensitive to concentration changes in the UTLS, due to large temperature contrast with the surface.
- The UTLS is the layer in which stratospheric and tropospheric air are mixed. The exchange between these atmospheric domains controls the influx of tracers into the stratosphere (including water vapor and long-lived greenhouse gases). It also controls the O_3 and NO_x flux from the stratospheric reservoir down into the troposphere.
- The influence of rapid convection within cloud structures and the large-scale vertical transport associated with convergence, the imprints of lower tropospheric events such as biomass burning and forest fires and regional air pollution episodes are also imposed on the upper troposphere.

While it is clear from these examples that the Upper Troposphere and Lower Stratosphere are physically linked, from an organizational standpoint these two regions have been, for the most part, studied by two separate groups of researchers. SPARC ("Stratospheric Processes and their Role in Climate", a project of the World Climate Research Program, WCRP) has traditionally focused on the stratosphere while IGAC (the International Global Atmospheric Chemistry" project, one of the core programs of the International Geosphere-Biosphere Program, IGBP) has focused on the troposphere. Clearly, there are mutual interactions between the stratosphere and the troposphere, and both influence climate. Further, there are common problems being faced by both research groups that could be worked on jointly, such as dealing with spatial and temporal mis-matches in observed and modeled data sets. Therefore, SPARC and IGAC have initiated a joint activity to study these overlapping areas together. Of the many important chemical and physical processes involved in climate-chemistry interactions, the joint SPARC-IGAC workshop explored the following the five general areas:

(1) Stratosphere-troposphere coupling,
(2) Lower stratospheric ozone abundances, and
(3) Chemically active greenhouse gases.
(4) Aerosols and their interactions,
(5) Water vapor and clouds,

To assess the current state of our understanding with respect to these key issues, a joint SPARC-IGAC workshop was held in Giens, France, during 3-5 April 2003. The organizing committee was comprised of A. R. Ravishankara, Shaw Liu, Ulrich Platt, Alan O'Neill, Tim Bates, Sandro Fuzzi, and Claire Granier. The specific goal of the meeting was to identify, discuss and prioritize outstanding issues related to the interactions between climate and chemistry that could be attacked jointly by the two research communities.

The workshop agenda and a list of session chairs, speakers, and session rapporteurs are listed in Table 1. (A complete list of speakers and attendees is provided as an appendix to this report.) The workshop was divided into 5 sessions, each with a speaker who summarized the issues. The talk was followed by short presentations and discussions. Many major issues related to climate and chemistry in general, and climate-chemistry interactions in particular, were discussed at the workshop. Special attention was paid to identifying regions of uncertainties. After the workshop, the rapporteurs (with help from chairs and other key participants) summarized the findings in writing. (The rapporteurs report will be available at URL: http://XXXX.) These summaries, along with the deliberations of a post-workshop discussion meeting in Boulder, CO, is the basis for this report.

Table 1. Topics at the SPARC/IGAC Workshop on Climate-Chemistry Interactions.

1- Stratosphere-troposphere coupling
Main speaker: R. Rood
Rapporteurs: T. Shepherd and A. Douglass; Session Chair: A. O'Neill

2- Lower stratospheric ozone and its changes
Main speaker: J. Pyle
Rapporteurs: M. Chipperfield and P. Simon; Session Chair: U. Platt

3- Tropospheric ozone and other Chemically Active Greenhouse Gases (CAGG)
Main speaker: D. Derwent
Rapporteurs: D. Hauglustaine and I. Bey; Session Chair: S. Liu

4- Aerosols and their roles in climate
Main speaker: F. Dentener
Rapporteurs: K. Carslaw and P. Quinn; Session Chair: T. Bates

5- Water vapor and clouds
Main speaker: U. Lohmann
Rapporteurs: C. Mari and K. Rosenlof; Session Chair: T. Peter

1. Stratosphere-troposphere coupling

Key Points:

The classical picture of stratospheric transport, in which material enters the stratosphere in the tropics, is transported poleward and downward, and finally exits the stratosphere at middle and high latitudes, was proposed to explain observations of stratospheric water vapor and ozone over 50 years ago. This conceptual model has since been refined but not

drastically altered. The mean-meridional stratospheric circulation, called the Brewer-Dobson circulation is controlled by stratospheric wave forcing (quantified in the so-called Eliassen-Palm flux divergence), sometimes coined the "extratropical pump", with the circulation at any level being controlled by the wave forcing *above* that level. However, the wave forcing can be difficult to compute accurately, and it is common to diagnose the mean circulation from the calculations of the zonally averaged diabatic heating. It is possible to estimate the net mass flux across a given surface of constant potential temperature (an isentropic surface) from the diabatic heating (for example, the 380K potential temperature surface, which is nearly coincident with the tropical tropopause and which marks the upper boundary of the lowermost stratosphere). On the other hand, transport along isentropic surfaces such as the isentropic transport of material between the upper tropical troposphere and the lowermost stratosphere is more difficult to quantify – especially for the net transport of a given species that results from two-way mixing. Observations show that the composition of the lowermost stratosphere varies with season, and suggest a seasonal dependence in the balance between the downward transport of air of stratospheric character and the horizontal transport of air of upper tropospheric character. For any time period the integrated mass flux to the troposphere at middle and high latitudes is the sum of (1) the mass flux across the 380K potential temperature surface, (2) the net mass transported between the upper tropical troposphere and the lowermost stratosphere, and (3) the mass decrease (increase) of the lowermost stratosphere, as indicated in Fig. 2. The first quantity is straightforward to compute, but the last two quantities are sensitive to small-scale processes, including synoptic-scale disturbances and convection.

For long-lived species such as N_2O and CH_4, the net mass flux from the stratosphere to the troposphere is independent of the details of motion near the tropopause. The annual total diabatic and isentropic flux of ozone to the troposphere can be similarly constrained, and is equivalent to the flux of ozone crossing the 380K surface at middle and high latitudes. This is true because the mass flux from the upper tropical troposphere into the lowermost stratosphere contains very little ozone. The net ozone flux has been estimated in several ways, including using direct fluxes from models. The estimated net ozone flux into the troposphere varies widely between models and most models' fluxes are higher than estimates made using observations, such as when observations of O_3 and a long-lived stratospheric tracer such as N_2O are combined with information from stratospheric models. This estimate relies on the model to calculate the loss rate of N_2O. The correlation between simultaneous measurements of HCl and ozone has also been proposed as a marker for tropospheric ozone of stratospheric origin. Alternately, the correlation between ozone and other stratospheric species with no tropospheric sinks may be used to estimate the stratosphere to upper troposphere flux of ozone. These downward fluxes of stratospheric constituents could change in response to changes in stratosphere climate through changes in stratospheric dynamics. For species of mostly tropospheric origin (*e.g.*, H_2O or short-lived halocarbons) the net mass flux is not sufficient to determine the flux of the species because of chemical transformations and spatial inhomogeneity, so it is necessary to quantify the mixing in the vicinity of the tropopause.

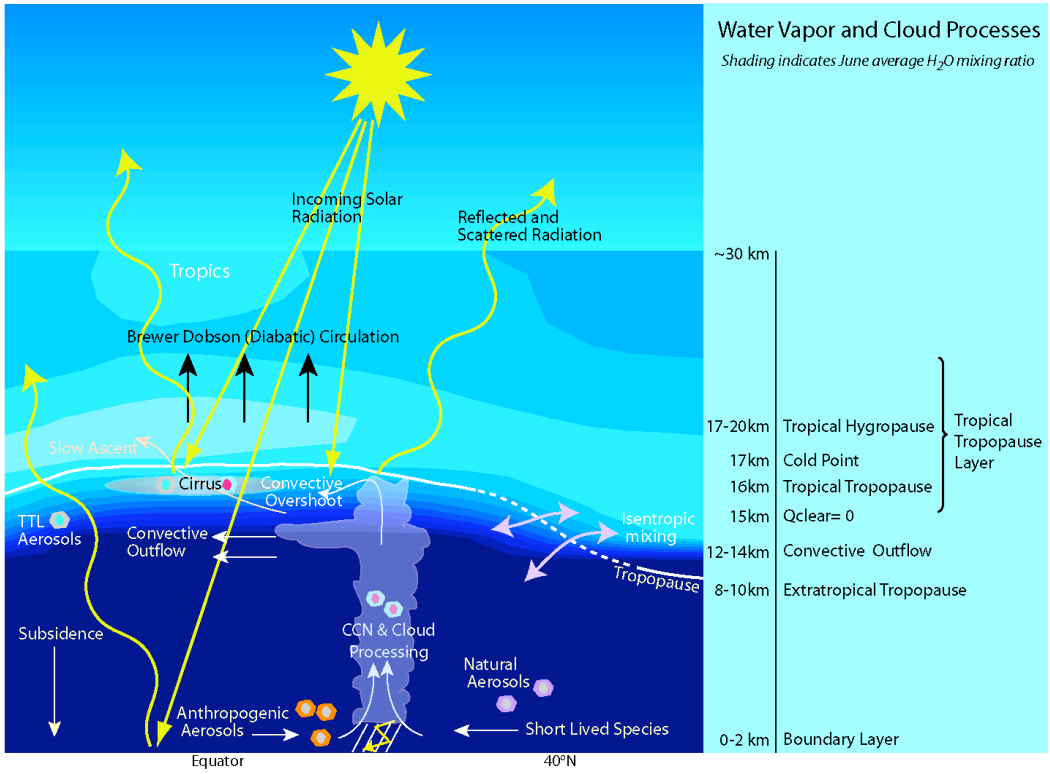

Figure 2. Water vapor distribution, cloud processes, and transport processes that influence the abundance of water vapor, clouds, and chemical constituents in the atmosphere.

Recent data indicate that the stratosphere and troposphere are coupled by more dynamically complex mechanisms than are given by the traditional model of large-scale circulation driven exchange. Waves generated in the troposphere propagate into the stratosphere where they can exert a force, and the circulation induced by the wave forcing extends downward into the troposphere. There has been considerable recent interest in the apparent coupling in the variability of the stratosphere and troposphere through the Northern and Southern Annular Modes (NAM and SAM), sometimes called the Arctic and Antarctic Oscillation (AO and AAO). The extreme states of this mode of variability correspond to strong and weak polar vortices. Observational and modeling evidence suggest a statistical connection through which the troposphere is influenced by the stratosphere, but specific mechanisms that would produce such a connection remain elusive. It is likely that the statistical connection is due to modulation of tropospheric wave propagation into the stratosphere. Because the impact is easier to see in the stratosphere, this coupling may have diagnostic and forecast utility for understanding tropospheric modes of variability.

In the tropics the upwelling branch of the Brewer-Dobson circulation passes through the cold tropical tropopause, and air entering the stratosphere is severely dehydrated. The extent of dehydration may depend upon factors other than the relationship between ice saturation vapor pressure and tropopause temperatures. In particular, for small-scale cloud processes, including convective transport, the fate of lofted ice may be important. Additionally, dehydration in the lower stratosphere after parcels have been affected by convection may be significant. If stratospheric water abundance is linked to small-scale cloud processes, then stratospheric water may be linked to tropospheric aerosols as well.

Recently there has been much interest in the upper part of the tropical troposphere, known as the "tropical tropopause layer" (TTL), which is a transition region between troposphere and stratosphere. The TTL extends from the level of main convective outflow in the tropics (10-14km) to the cold point (18-20km). This is a region that takes on both stratospheric and tropospheric characteristics, and any climate change induced alterations of this region could potentially impact stratospheric composition, as it is essentially the source region for stratospheric air. Ozone profiles (from the SHADOZ ozone sondes) in the TTL show that the cold point is not typically coincident with the "chemical" tropopause (i.e., the altitude where ozone begins to increase significantly), except where deep convection reaches up to the tropopause. The data suggest that horizontal transport of air from the lowermost stratosphere into the upper tropical troposphere is a source of ozone to the TTL, as well as possible photochemical ozone production.

Large scale temperature observations of the UTLS, such as are available from the current assimilated observations, may underestimate the amplitude of temperature fluctuations in this region. Small-scale variations in temperature produced by presumed ubiquitous gravity waves in the TTL appear to be necessary for detailed cloud models using homogeneous nucleation to reproduce individual cirrus observations. Thus small-scale temperature variations may impact the dehydration of air entering the stratosphere. These small-scale variations are absent from most analysis systems (*e.g.*, ECMWF). Unfortunately, there are not good observational constraints on the amplitudes of the gravity waves which cause these temperature perturbations.

Constituents that are short-lived in the troposphere (and their products) may be a significant source of chlorine and bromine species in the lower and lowermost stratosphere. In models the convective transport in the tropics controls the distribution of some species in the upper tropical troposphere. Lack of measurements of these short-lived species (and their products) in the TTL and lowermost stratosphere, an inability to validate convective transport in models, and uncertainty in the horizontal exchange between the upper troposphere and lower stratosphere all contribute to the uncertainty in the impact of these compounds on the stratosphere.

Many of the processes that produce coupling between the stratosphere and troposphere are at spatial and temporal scales that are not resolved in climate models. Future research must include a focused effort to represent such processes realistically in global models, making sure that their representation responds appropriately to external perturbations.

Outstanding Issues

Improve understanding of dynamical coupling - A key issue is prediction of the effect of extra-tropical tropospheric dynamical changes on stratospheric planetary wave forcing through changes in tropospheric forcing of planetary waves or in their propagation characteristics. This is important for stratospheric ozone abundance, which is controlled in part by the wave-driven Brewer-Dobson circulation, and for overall stratosphere-to-troposphere mass flux. Current climate model predictions of changes in these dynamical structures due to greenhouse gas increases do not even agree on the sign of the effect [*Austin et al*., 2003]. Another issue is the possible effect of stratospheric dynamical changes on the troposphere, such as via the aforementioned annular modes/stratosphere connection. In both cases, there is a question of the robustness of the effects, a need to identify physical mechanisms, a concern about the dependence of the modeled effects on model details, and the lack of sufficient statistics to draw firm conclusions.

Quantify tropical stratosphere-troposphere exchange through improvements in knowledge of the TTL and dehydration- An important question is how well the TTL must be resolved for tropospheric chemistry applications. We currently do not have a complete observed climatology of basic quantities in the TTL (i.e. H_2O, O_3, heating rates, and common tracers like N_2O or CO). More observations, both from satellites and in-situ aircraft, are necessary. Presumably it is important to represent the speed of transport of air through the TTL correctly, as this would affect the amount of chemical "aging" in the troposphere (which is important for short-lived species). This transport is likely to be dependent on model details. We need to address questions such as:

- Exactly how is air dehydrated in the vicinity of the tropical tropopause before entering the stratosphere?
- What is the relative importance of cirrus clouds (with slow ascent) versus convective activity in controlling dehydration?

Finally, we broadly understand many of the scales of variability of UTLS water vapor and how this is linked to temperature and chemical variations – for example via the annual cycle, the effects of ENSO or the QBO, and long term increases in methane. However, there are unexplained interannual variations in stratospheric water vapor, usually termed 'trends' though they are not linear or monotonic. These variations are difficult to explain because there are significant uncertainties in our long-term records of UTLS water vapor. More observations for monitoring UTLS water vapor on climate scales are needed.

Evaluate the extra-tropical stratosphere to troposphere flux- The current model range for the O_3 flux to the troposphere is too high, given the observational constraints. We therefore need to use the following data-based methods to evaluate the ozone flux:

- Develop metrics to reduce uncertainty in model predictions, such as is seen in their wide range for O_3 flux. This is important for estimates of chemical climate change, because errors in O_3 flux to the tropopause will affect the budget of

tropospheric ozone and compromise tropospheric chemical climate change experiments.

- Evaluate transport in models using the age of stratospheric air and other long-lived tracers. It is important to determine how the age of air and the stratosphere-to-troposphere flux might change with climate change. This would come about through future changes in planetary wave forcing. Models suggest that the circulation might either speed up or slow down in the future.
- Understand longitudinal variations in the stratosphere-to-troposphere flux as well as the net mass flux, as this will be important for short-lived species and for tropospheric chemistry.

Evaluate extra-tropical troposphere to stratosphere flux- This process is important for the distribution of radiatively active species (and possibly aerosols) in the lowermost stratosphere. At this point, we have neither characterized the lowermost stratosphere for "present day" conditions, nor evaluated models from this perspective. Measurements are needed to examine both seasonal and spatial variability of species in the lowermost stratosphere, using a range of tracers with a spectrum of lifetimes.

Address the issue of upscaling our knowledge- An ongoing challenge is to "upscale" information, namely to link what we learn from case studies to the representation of various processes in global models, to determine global budgets, and to understand their contribution to global change.

- Mesoscale models are crucial for validation of global models from a process point of view but are limited by the availability of the constituent data needed to initialize such models.
- Large-scale constraints can make it possible to represent small-scale processes in a global model. Two examples are (1) using a global circulation to estimate the flux out of the stratosphere (rather than counting every fold) or (2) using the boundary layer distribution of buoyancy to determine the vertical profile of convective outflow in the TTL.

An open question is whether we can treat stratospheric water in a GCM as being controlled by large-scale processes at the tropical tropopause. To do so requires that we reproduce the important effects of small scale processes through parameterizations in global models. Such parameterizations will only be arrived at through process studies and detailed in-situ observations, as well with the global-scale retrievals from satellites.

2. Lower Stratospheric Ozone

Key Points:

Ozone in the lower stratosphere (LS) plays a key role in the chemistry of both the lower stratosphere and upper troposphere. In the stratosphere, it represents a significant fraction of total ozone. In the troposphere, LS ozone represents an important source of

ozone that is realized through stratosphere-troposphere exchange events. At times this can have a significant impact on ground-level ozone and therefore on both plant and human health. In addition, ozone in the LS is coupled to Earth's climate. The coupling acts in both directions: changes in LS ozone will affect climate and climate change will affect the abundance of LS ozone. Furthermore, this coupling represents a link between atmospheric chemistry and climate change, because LS ozone is partly controlled by photochemical processes.

Gases from natural and anthropogenic (i.e. pollutant) surface sources bring reactive components to the stratosphere that affect ozone. These gases enter the stratosphere by crossing the tropical tropopause as a result of transport processes related to convection. As described in the previous section the details of this transport, which occurs in the tropical tropopause layer (TTL), are not fully understood. For long-lived pollutants (*e.g.*, CFCs), a detailed understanding is not critical. However, for short-lived source gases (*e.g.*, bromine and iodine containing gases) the timescale and geographical location of transport are critical. Therefore, we must understand the role of convection in the TTL in transporting gases to the stratosphere, as well as understanding how the details of this transport may change in the future.

Tropospheric wave driving exerts a very strong influence on stratospheric dynamics, which in turn strongly influences the distribution of LS ozone. Changes in stratospheric dynamics related to natural variability or climate change have the potential to alter LS ozone abundances. Ozone column amounts in the winter Arctic LS are particularly sensitive to such changes. Therefore, understanding future changes in the wave driving of the stratosphere are important.

Ozone in the LS is chemically long-lived and controlled by both dynamics and relatively slow chemistry (outside of the polar spring). The chemistry involves both gas-phase and heterogeneous reactions under the relatively low-temperature conditions of the LS. The losses of LS ozone that have occurred in the past two decades are expected to be reversed (*i.e.*, ozone recovery) in the coming decades as the stratospheric halogen loading declines in response to emission reductions that have come about through implementation of the Montreal Protocol on Substances that Deplete the Ozone Layer. Stratospheric cooling (resulting from increasing greenhouse gases) is expected to increase ozone in the middle stratosphere as a result of changes in the rates of key gas-phase reactions that control ozone amounts. In the LS the situation is more complicated, but it is possible that ozone will also increase there. An increase in LS ozone will reduce the UV flux in the troposphere and, hence, decrease tropospheric OH, a key tropospheric oxidant. (See IGACtivities No. 28, May 2003). Therefore, coupled chemistry/climate change studies should include the role of this potentially important process. Many of the changes in ozone in the coming years due to the processes noted above will be relatively small. As a consequence, our representation in atmospheric models of these processes will need to be comprehensive and precise in order to account for observed changes and to predict future changes.

Predictions of future changes in the atmospheric chemical composition will necessarily make use of meteorological forcing fields (*e.g.*, temperature, winds, water vapour, and convective fluxes) from General Circulation Model (GCM) simulations. It is therefore of vital importance to quantify uncertainties in those GCMs regarding future climate changes. One necessary preliminary step should be the systematic validation of the models for the current atmosphere (including its mean state, variability and trends over the last decades), which is now possible with the recently available reanalysed datasets (NCEP, ECMWF, NASA-DAO).

Calculated circulation parameters and LS temperatures in many models are biased with respect to observations. For example, temperature biases in the LS can reach several degrees Kelvin, which is enough to significantly affect simulations of high-latitude winter ozone depletion, especially in the Arctic. Mean meridional transport can also be quite different from model to model, either because of physical reasons (*e.g.*, different model parameterizations related to gravity waves or convection, which lead to different Brewer-Dobson circulations) or simply because of numerical reasons (*e.g.*, the location of the upper boundary of the model, numerical algorithms, etc). Major problems also appear in models' water vapour fields, especially in the UT/LS region. Thus, the continued availability of atmospheric meteorological observations will be essential to achieve and maintain the skill of GCM simulations for chemistry and climate processes in the UTLS.

Outstanding Issues

Understanding the photochemistry and dynamics that affect LS ozone and how these processes interrelate to climate change processes requires both observational and model studies.

For an accurate description of LS ozone abundances, improved understanding of the following chemical processes are needed:

- Gas-phase chemistry rate constants at low temperature (*e.g.*, the $HO_2 + O_3$ reaction).
- The nature of the particle surfaces present in the lower stratosphere (*e.g.*, formation and reactivity).
- Removal of water vapor and reactive nitrogen from the lower stratosphere in polar winters (*i.e.*, dehydration and denitrification)
- The transport and reactive conversion of very short-lived gases to the upper troposphere and lower stratosphere.

Similarly, an improved understanding of the following dynamical and transport processes is needed:

- Forcing and propagation of planetary waves, and stratospheric dynamical coupling to the troposphere
- Stratospheric response to resolved and parameterized wave drag
- Prediction of the Quasi-biennial and Semi-annual oscillations (QBO and SAO)

- Transport across subtropical and polar mixing barriers (*e.g.*, transport of ozone-depleted air from the winter polar vortices to lower latitudes)
- Details of extratropical stratosphere-troposphere exchange and mixing.

Process studies in the lower stratosphere are essential. The studies include multi-instrument campaigns located on the ground or on board research aircraft and include satellite instrument suites. Past studies have identified and confirmed key processes related to photochemistry and dynamics and provide key datasets for use in model validation. Similar studies should be continued in all regions of the lower stratosphere (*e.g.*, tropical, mid-latitude, and polar regions).

Model comparisons with other models and with observational and process-study datasets are important for assessing and guiding model development. A key step is the validation of stratospheric GCMs for the last 40 years (*i.e.*, an extended "Atmospheric Model Intercomparison Project (AMIP)-like" experiment) and the validation of chemistry modules in chemical transport models (CTMs). Other steps in model development should include GCM and CTM runs that have:

- several scenarios of atmospheric composition and meteorological parameters to assess the robustness of the models' response;
- ensemble simulations to assess the internal variability of the models' response; and
- comparison of models under common scenarios to assess how the response is model-dependent.

3. Tropospheric ozone and chemically active greenhouse gases

Key Points

The global burdens of methane and tropospheric ozone have increased by about a factor 2.5 and 1.3, respectively, since the pre-industrial era. These compounds are greenhouse gases. Tropospheric ozone changes contribute roughly 0.35 W/m^2 (about 8-15 %) to the total radiative forcing associated with greenhouse gas increase since the pre-industrial times. Tropospheric ozone (O_3) and other chemically active long-lived greenhouse gases (such as nitrous oxide (N_2O), methane (CH_4) and various halocarbons) contribute about 50% of the radiative forcing of climate since the pre-industrial, i.e., they contribute roughly as much as industrial carbon dioxide (CO_2).

In addition to the direct forcings, ozone and other chemically active greenhouse gases can also indirectly modify:

- the oxidation of several hydrocarbons of natural or anthropogenic origin that produce secondary organic aerosols, which add to the total aerosol load in polluted regions and affect the radiative budget of the atmosphere.

- the oxidizing capacity of the atmosphere. The lifetimes (thus the concentrations) of several long-lived greenhouse gases (such as CH_4 and HFCs) are controlled by the concentration of tropospheric hydroxyl radical (OH), which is derived mostly from ozone.

The uncertainties connected with estimates of these indirect effects are much larger than the uncertainties of estimates of tropospheric ozone's direct radiative effects, because the chemistry of CH_4, CO, NMHCs, NO_x, O_3, and some reactive halogen species is strongly intertwined.

Tropospheric chemistry can also affect the sources and sinks of CO_2 and hence the global carbon cycle. For example, there are indications that deposition of nutrients (air pollutants) and river runoff can affect the uptake of CO_2 by the biosphere.

Climate changes can potentially alter tropospheric chemistry (see Fig. 3 for some of the relevant reactions) via changes in:

- temperature and water vapor, which directly affect the rates of ozone and radical production and destruction processes.
- emissions of precursors and aerosols.
- scavenging processes of ozone and aerosol precursors, and the aerosols themselves.
- changes in the Brewer-Dobson circulation and the consequent alterations in the stratosphere-troposphere exchanges and global distribution of ozone in the troposphere,
- changes in convective activity or in weather patterns and the consequent alterations in the intercontinental transport of ozone and its precursors.

These possible changes would also significantly alter regional air quality. Thus climate and regional air quality are strongly connected, for example through the intercontinental transport of pollutants that determines the background levels of ozone and its precursors.

Outstanding Issues

We must improve the fundamental building blocks of our understanding tropospheric chemical processes. These include improvements in characterizing the rates of gas phase, heterogeneous, and photolytic processes; process studies in the atmosphere to test our understanding of the chemical processes; and incorporation of these processes in a realistic way in global climate models. Improvements are needed specifically in the following areas:

- *Chemical processes*- Uncertainties still exist regarding the products of some basic, long-studied, reactions (*e.g.*, HOONO formation).
- *Tropospheric halogen chemistry*- Halogen tropospheric chemistry – in particular the iodine and bromine cycles – are still poorly understood.
- *Volatile Organic Compound (VOC) degradation*- there are still numerous uncertainties in the kinetics, the chemical pathways and the nature of secondary

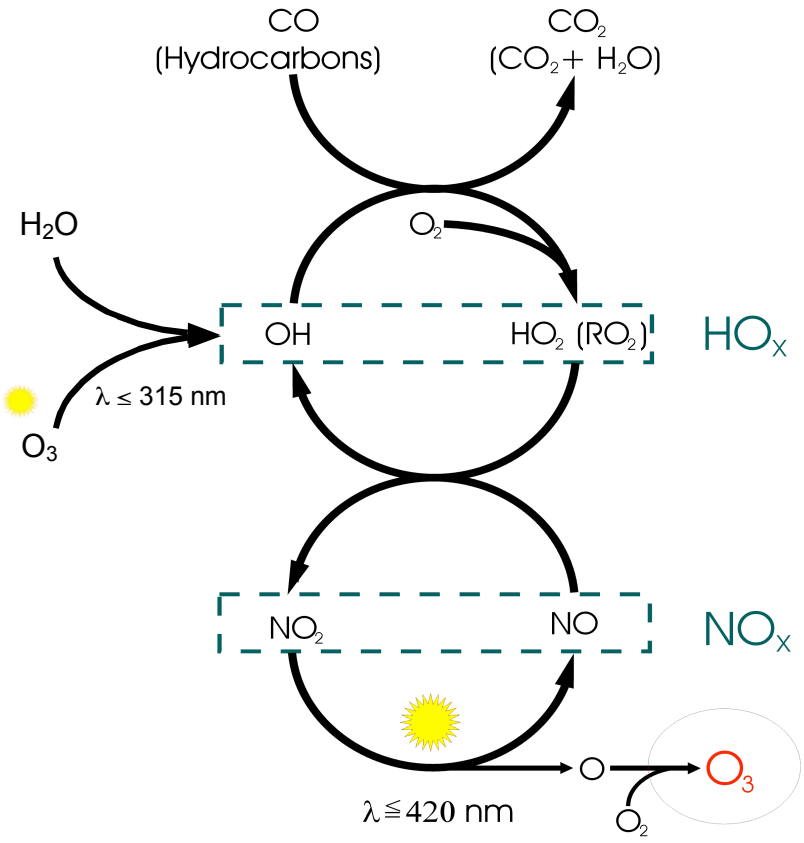

Figure 3. Schematic of the tropospheric photochemistry that oxidizes reduced carbon species. The oxidation is accomplished by a radical chain mechanism carried by HO_x radicals. The presence of NO_x catalyzes the formation of ozone in this oxidation process.

products in the degradation mechanisms and photochemistry of organic compounds. These uncertainties in VOC degradation affect not only the chemical ozone budget but also secondary aerosols production.

- *Heterogeneous processes*- The largest uncertainty in the chemistry of current global models is probably associated with the representation of heterogeneous processes. Heterogeneous reactions, which can either be scavenging or activation reactions, directly affect the budget of radicals and nitrogen species.
- *Photolytic processes in the presence of clouds and aerosols*- The role of aerosols and clouds in altering photolysis rates has to be considered carefully in global models.
- *Emissions*- Uncertainties in trace gas radiative forcing arise from our limited understanding of the global distribution, budget and evolution of greenhouse gases in the atmosphere. The budget of key species is governed by their emissions at the surface and by sinks in the atmosphere or at the surface. Natural emissions are responsible for a large part of the observed variability of long-lived greenhouse gases and ozone precursors such as NO_x and NMHCs. The magnitude of these emissions, their distribution and response to climate change is a major source of uncertainty.

Observations of atmospheric constituents are critical for testing models, understanding basic processes, and identifying new chemicals and processes. The major observational needs are:

- *In-situ gas-phase species measurements at high spatial and temporal resolution* - Measurements of atmospheric constituents using aircraft, ground stations, balloons, and LIDAR and other remote measurement techniques are essential for providing abundances that can be used for testing our understanding of processes and for bounding atmospheric concentrations. The mean concentrations of important greenhouse gases, their precursors and aerosol precursors must be quantified as a function of location and altitude, and the temporal variations on diurnal, seasonal and inter-annual timescales must be captured. Also informative is quantification of the variability of the concentrations and correlations in the variability of different species.
- *In-situ aerosol measurements at high spatial and temporal resolution*- Measurement requirements of aerosol species are similar to those for gas phase species but are much more complex. Specifically, detailed information is needed regarding their chemical composition and physical and optical properties.
- *Process studies designed specifically to reduce our uncertainty in key areas*- Ambient measurements can be designed to provide specific tests of emission inventories, transport mechanisms, and depositional processes. Two transport mechanisms are of particular importance. The first is transport from the boundary layer to the free troposphere. The large majority of emissions are released to the atmosphere within the continental boundary layer, which accounts for only a small fraction of the volume and mass of the troposphere. The boundary layer is also the site of the most rapid removal of many important species. It is very difficult for models to accurately quantify the fraction of emissions that are transported from the boundary layer to the free troposphere, since this transport occurs by many mechanisms (i.e. synoptic scale airstreams, convection, boundary layer growth and decay, mountain-valley circulations, land-sea circulations, small scale eddies, etc.) which have large spatial and temporal variability. It is these complex transport mechanisms that ultimately determine the influence of surface emissions. Second, stratosphere to troposphere exchange has a profound influence on the variability of ozone throughout most of the troposphere, particularly the upper troposphere.
- *Observations from space*- Space observations are extremely useful for evaluation of models because they provide wide, nearly global sampling of constituents that vary both spatially and temporally. In the troposphere, recent observations of O_3, NO_2, and CH_2O tropospheric columns from GOME/ERS-2, and of CO from MOPITT/EOS-Terra have provided evidence of the large-scale perturbation of the atmospheric composition by human activities. These datasets are now complemented by the observations of chemical species in the troposphere by SCIAMACHY/Envisat and soon by OMI and TES onboard EOS-Aura. In order to meet the stringent measurement requirements imposed by climate-chemistry studies, further development of instruments to probe down into the troposphere

would be useful. In particular, we need higher horizontal resolution to address pollution related issues; information on the vertical distribution of species (most specifically in the UTLS where sharp vertical gradients exist); information on the diurnal variation of chemical species; and measurement of new species of interest for tropospheric studies. This should be undertaken simultaneously with the monitoring from space of active fires and lightning flashes to impose further constraints on the models and on the emission of ozone precursors. Such satellite-based measurements have begun only recently. More work is critically needed to compare these measurements with in-situ data in order to quantify the accuracy and precision of the satellite retrievals. At present such assessments are largely lacking.

Enhancements in modelling capabilities are essential for further progress and for providing information that is sought from the community for policy decisions. Major improvements in modelling capabilities are needed in the following areas:

- *Coupled spatial regimes and processes*- The next generation models should treat dynamics, radiation and chemistry simultaneously, and they should treat both the troposphere and stratosphere simultaneously. Such coupling is essential for reproducing the observed trend in ozone in the troposphere and in particular in the UTLS region. Other changes, such as stratospheric water vapor increases or increased penetration of UV radiation into the troposphere and the subsequent impact on the oxidizing efficiency of the troposphere, will only be adequately represented if both the troposphere and the stratosphere are correctly represented in the models. Coupled models are currently under development; however, they have been used only with simplified tropospheric chemistry to investigate the impact of aircraft emissions on the composition of the atmosphere. Further improvements to include more detailed representations of tropospheric processes, non-methane hydrocarbon chemistry, and surface processes are needed.
- *Improved spatial resolution*- Current chemical-transport models of the atmosphere are run on a typical 2-degree by 2-degree resolution with 20-60 vertical levels. The next generation of models should have higher horizontal and vertical resolutions. High resolution is required in source regions to provide better representation of surface emissions, to account for non-linear effects in atmospheric chemistry and for better representation of sub-grid scale processes such as convection or boundary layer mixing. Similarly, high resolution is crucial for representing stratosphere-troposphere exchange, and the model top boundary is a critical parameter in the representation of the Brewer-Dobson circulation. Because of calculational limitations, it is important that these resolution requirements be quantitatively defined so models are optimized. For example, high resolution is needed but probably not everywhere and the degree of resolution will vary by region/altitude. Nesting of regional or plume models in global chemical transport models (CTMs), numerical zooming techniques and an ensemble of parameterisations will have to be developed in order to better represent the emissions, transport, and chemistry in the troposphere.

- *Deposition processes*- The washout and rainout of soluble species that constitute the ultimate loss of nitrogen and HO_X reservoir species are also a large source of uncertainty in global models. More physically-based parameterizations of these sub-grid scale processes need to be developed in global models. This is a research area where close collaboration with the regional modeling community would be beneficial by increasing the likelihood of adaptation and implementation of these parameterizations in models. A similar approach needs to be adopted in the case of surface dry deposition parameterizations, which also require representation of aerodynamical, surface, and biospheric sub-grid scale processes. Model representation of dry and wet nitrogen deposition (including precipitation) must be evaluated through comparison with surface measurements from networks. This type of evaluation provides a closure test and allows for further constraint on the global budget of nitrogen and other species, as well as providing a link between global tropospheric chemistry and regional air quality.
- *Data assimilation*- Data assimilation provides a means to interpolate observations in time and space; to integrate diverse elements of large measurement and modelling programs; and to combine, inter-compare, and characterize the observations from different types of instruments. Inverse modelling and chemical data assimilation techniques are advancing rapidly and are expected to reach a high level of sophistication in the near future. These techniques will, for example, allow us to infer the distributions of species that interact chemically with observed species but that are not observed directly by satellites. This will allow full advantage to be taken of satellite and network observations, improving the emission inventories of long-lived as well as more reactive source gases.
- *Improved coupled climate-chemistry models*- An important objective of the modeling community must be to develop and apply coupled chemistry-climate models. It is only recently that general circulation models have been fully coupled to chemical processes, and the first studies performed with these models provide exciting new insights on how the coupled climate system amplifies or damps chemical perturbations. The need for fully coupled models is emphasized by model simulations of the impact of climate change on the oxidizing efficiency of the atmosphere resulting from perturbations in humidity, temperature, cloud distributions, or convective activity (i.e. via changes in species transport, the scavenging of soluble species, and lightning NO_x emissions). It is also reinforced by recent findings showing a clear connection between stratospheric ozone distributions and the climate's variability, such as with oscillations in ENSO or the NAO. These models will have to be increasingly refined to include more detailed chemistry, coupling with the stratosphere, interactive emissions from the biosphere, and heterogeneous processes. Climate-chemistry feedbacks can arise from changes in biogenic emissions and dry deposition that were triggered by climate change; these, in turn, affect climate. The study of these feedbacks requires that tropospheric chemistry models be coupled to interactive continental and oceanic biosphere models that include emission and deposition modules for ozone and its precursors.

4. Aerosols and their role in climate

Key Points

Direct forcing of climate by aerosols is perhaps the best-known quantity in aerosol-climate connections, though even this effect is not sufficiently well quantified. The magnitude of the forcing has been estimated; it appears to be substantial and rivals that due to stable greenhouse gases. For the most part it is opposite in sign to that of greenhouse gases, however the regional nature of aerosol forcing and its variable vertical distribution precludes simple conclusions of a global canceling of greenhouse gas warming.

In spite of advances in understanding over the past decades, the forcing by aerosols via absorption and scattering of radiation is still uncertain. The uncertainties arise because of poorly characterized variability in their vertical and horizontal distribution, composition, optical properties, hygroscopicity, and size distribution. Further, connecting aerosols' and gaseous precursor species' emissions to the spatial and temporal distributions, optical properties, hygroscopicity, and chemical composition of tropospheric aerosols is confounded by a lack of understanding of transport, transformation and aging processes, and of cloud processing. It is essential that such connections are represented accurately in global climate models, both for forecasting climate-state and climate-change, and for distinguishing natural from anthropogenic influences.

The indirect forcing by tropospheric aerosols (i.e. the impact of aerosols on cloud properties) is qualitatively understood but the magnitude of these effects is highly uncertain. Potential indirect effects that have been identified include changes in cloud albedo, the modification of ice and water clouds, changes in precipitation patterns and rates, and alteration to the composition of the atmosphere. In addition, the composition and size distribution of aerosols are affected by gas phase and cloud processes and the resultant effects on chemistry and radiation are highly uncertain. Possible impacts of these aerosol-cloud interactions have been explored to a limited extent through modeling sensitivity analyses. These studies indicate that the effects are substantial and need to be better quantified before an assessment with acceptable uncertainties is possible.

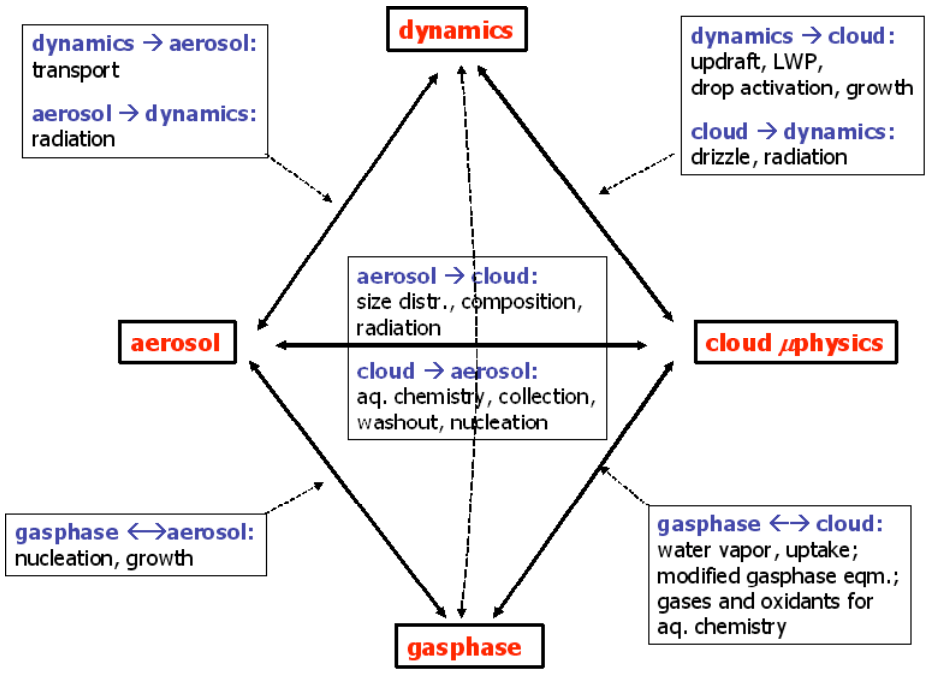

Figure 4. Schematic of the intricate coupling between the various processes that act together in the atmosphere and that should be considered when dealing with the role of aerosols in earth's composition and climate.

Stratospheric aerosols, in contrast to tropospheric aerosols, appear to be made up primarily of sulfuric acid, water, and, on occasion, nitric acid. Their impact on radiation and the stratospheric chemical composition is reasonably well understood even though many uncertainties exist. To first approximation, the climate impact of stratospheric aerosols appears to be amenable to representation in global climate models. Uncertainties in stratospheric aerosols include knowledge of which particles in the polar regions during cold periods grow to large enough sizes to sediment, and the impact of these processes on altering the water and nitric acid concentrations in the polar stratosphere. This has implications for the abundance of polar stratospheric clouds and therefore also springtime stratospheric ozone depletion.

The climate response to changes in aerosol composition and properties and the feedbacks that are involved is either unknown or, at best, only qualitatively understood. High priority questions include:

1) *What is the response of the hydrological cycle to changes in aerosols?* For example increases in aerosol concentrations are likely to suppress precipitation and increase cloud amount and spatial coverage. This results in an even stronger albedo modification than that due to the effect of the aerosol on drop size and reflectance alone. Conversely, increases in giant CCN concentrations may increase precipitation rates and reduce cloud amount, as well as aerosol and soluble gas concentrations.

2) *In what way is the climate response to the changes in aerosols (with their highly localized and seasonally varying forcings) different to that of greenhouse gases?* Quantification of the feedbacks shown in Figure 4 is a crucial for assessing the impact of aerosols on climate.

Outstanding Issues

Due to the large spatial and temporal variability in aerosol (and cloud) properties, aerosol radiative forcing is a regional scale issue. Within a given area, aerosol radiative forcings at the surface can be very large (-20 to 60 W m^{-2}). To better understand the climate impact of aerosols, an approach is required that produces regional scale assessments of top-of-atmosphere (TOA) and surface forcing along with the global mean forcing at TOA (for comparison to greenhouse forcing). The regional scale approach is also required for assessing the impact of aerosols on air quality. It is the regional scale emissions and processes that determine global scale contributions of natural and anthropogenic aerosols; such differentiations are crucial for policy decisions.

Uncertainty Estimates

In addressing strategies for improving the assessment of aerosol effects we must first evaluate how uncertainty in the aerosol abundance and properties translates into uncertainty in estimates of the radiative forcing and how the magnitude of this uncertainty compares to that associated with greenhouse gases. In this evaluation, particular attention should be paid to the following issues:

- Identification of quantities that may contribute to a non-linear dependence of aerosol forcing on burden. (Such relationships are central to climate change attribution studies used by the IPCC);
- Consideration of the radiative effects of the entire aerosol system, not just individual components;
- Distinguishing forcing from "noise" in the climate system;
- Assessing the effects of aerosol on the radiative properties of both water and ice clouds. This should encompass the entire lifecycle of clouds so that the effects on the hydrological cycle can be evaluated;
- Subjecting models to a range of input conditions and a degree of testing that has not yet occurred.

Direct Effects

Aerosol size and composition is complex and this complexity needs to be appreciated and accounted for.

- Aerosols are a mixture of soluble and insoluble species that respond differently to changes in relative humidity. Aerosol growth in response to relative humidity is a crucial aspect of assessment of direct forcing. Organic material on the surface of

the particles and reactions between chemical species within particles may be of importance.

- Particle surface tension (an important term for CCN activation) varies with solute concentration and type. Particle composition varies as a function of size and source region.

These factors, when included in cloud parcel models representing growth and activation of aerosol indicate that chemical effects on cloud properties can be as large as dynamic and aerosol size distribution effects when compared to a baseline case of soluble inorganic species.

Indirect Effects

Aerosol-cloud interactions and associated feedbacks to the climate system are highlighted as an example of a complex and coupled system between aerosols, clouds, dynamics, and gas phase species (Fig. 4). Because of these connections, there are myriad possible pathways for feedbacks in this system. Moreover, many of these feedbacks occur at small scales so that evaluating their importance is challenging.

The extent of the effect of aerosols on clouds and associated feedbacks depends, at least in part, on:

- how cloud macroscale properties such as cloud coverage, liquid water path, cloud depth, and precipitation change in response to changes in aerosol and CCN;
- whether anthropogenic increases in aerosol concentration are accompanied by changes in aerosol hygroscopicity;
- whether increases in aerosol/CCN concentrations are accompanied by increases in giant CCN concentrations which are effective at initiating precipitation;
- the extent to which clouds process aerosol and change the CCN size and composition, thus potentially modifying subsequent clouds;
- whether aerosols are strong absorbers, thus endowing them with the potential to modify atmospheric stability.

Clearly any feedback process that has the potential to modify cloud macroscale properties, and therefore cloud reflectance, is a primary concern. Improvements in the understanding of these processes and their representation in models are needed.

Data Requirements

Data needed for evaluations and forecasting are multidimensional and inhomogeneous and need to be carefully dealt with to take this complexity into account. In particular the following issues should considered:

- Evaluation of models against observations should take very careful note of the inherent limitations in observations, which are quite often retrieved, rather than directly measured quantities.

- Consistency should be required of the measurements used to evaluate models and satellite retrieval algorithms. To determine which effects are due to chemistry versus meteorology, the models have to be translated into observation space. For example, large-scale models could be sampled in the same way as observations.
- We need an overlap of satellite measurements and modelling. Multiple satellite measurements should be used synergistically. For example, absorbing aerosol information from TOMS and true color information from SEAWIFS can be used together. The initial approach should be to test the method with case studies before scaling up to the global level.
- There is a need for further systematic laboratory observations of the optical properties and hygroscopicity of aerosols and aerosol mixtures.
- We need to expand compilations of measured aerosol microphysical and chemical properties for better estimates of regional means and variability.

Other strategic challenges

1) Synthesizing system complexity

It is crucial that we evaluate the extent to which details of the climate system are important for the overall understanding of the climate system. For example: Is detailed knowledge of the aerosol size distribution and composition always required or are there adequate proxy measurements that are easier to perform? How many of the complex interactions represented in Fig. 3 are important, and at which spatial/temporal scales? Can we synthesize our understanding of the complexity into physically based parameterizations that capture the essence of the process under consideration? Will such parameterizations represent adequately the underlying physics when included in global scale models?

2) 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 aerosol-microphysics-chemistry processes. 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 aerosol-cloud interactions, and 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 microphysical-chemical models in climate models based on the principles of synthesis and parameterization outlined above, or of increasing the spatial and temporal resolution of the climate models. Predicting the impact of the aerosol indirect effect is an enormous challenge because it requires that models correctly predict both the co-location and timing of aerosol and cloud events, as well as the interactions between aerosol and clouds.

5. Water vapor and clouds

Key Points

Water vapor abundance and relative humidity

Water vapor feedback in climate models is known to be important. This is highlighted by the need to include water vapor feedback (long-wave component) in a GCM to adequately model the temperature response due to the Mt. Pinatubo eruption. Changes in tropospheric water vapor brought about by climate change may impact the stratosphere, if those changes affect the UTLS. As noted earlier, an increase in water vapor in the lower stratosphere is radiatively significant because of the large temperature difference between this region and the surface.

Similarly, changes to cloud properties, such as their reflectivity or lifetime or to the relative abundances of different cloud types, can have a very large effect on the climate system. While it is clear that the addition of long-lived greenhouse gases (GHGs) to the atmosphere will directly alter the earth's radiative balance, less well-understood is to what degree that change in radiative balance will lead to changes in cloud properties. However, it is possible that the radiative impact of such cloud feedbacks will be greater than that of the direct radiative GHG forcing. We are still in the process of identifying all of the potential cloud feedbacks under a changing climate, and the quantitative impact even of those that are well-established is still highly uncertain.

The importance of clouds in the climate system was demonstrated in one model study that showed that the air temperature at the top of cirrus clouds in the tropics does not change with increased sea surface temperatures. Such an effect would decouple the tropical outgoing long-wave radiation emission from the surface temperature. As a consequence, the tropical climate below the anvil clouds could be very sensitive to sea surface temperature changes, if only the long-wave emission is important. Additionally, anthropogenic activities can change cloud processes that in turn affect surface temperatures. In particular, work has shown that clouds formed through processes initiated by aircraft contrails actually reduce the diurnal temperature range.

Changes in cirrus clouds and highly convective clouds (whose tops reach into the UTLS region) will likely affect the chemical composition of the lower stratosphere. Changes in the frequency of these clouds or their composition may arise from changes in atmospheric dynamics or via the indirect effect of aerosols on cloud properties (such as with ice nucleation properties, as discussed in the previous section). Through stratosphere/troposphere exchange, these tropospheric cloud changes may result in alterations to the water vapor abundance, aerosol loading and the concentration of other chemical constituents in the lower stratosphere.

Understanding controls on water vapor and cloud processes in the tropics is key to understanding the climate system and future changes (see Fig. 3). The tropics is the source region for air entering the stratosphere, and thereby impacts stratospheric

composition. Changes in the radiative balance in the stratosphere have the potential to alter the radiation balance in the troposphere, and therefore near surface climate. Clouds, aerosols and humidity all play significant roles in the radiation budget of the upper tropical troposphere.

The processes that determine the relative humidity (RH) in the tropical troposphere layer are not understood well enough to predict the water vapor abundances. The mean tropical RH takes on a "C" shape, with large values in the boundary layer, low values in the mid troposphere, and increases again near the tropical tropopause. In particular, above the level of zero radiative heating, the frequency of super saturation increases significantly. Even though there are some plausible theories regarding controls on RH they are not tested. This makes it difficult to predict the abundance of water vapor in the upper troposphere, and knowledge of water vapor abundance is crucial for calculating water vapor enhancement and feedbacks. While the direct radiative impact of an increase in GHGs would produce an increase in surface temperature, the total response may be affected (or even dominated) by the as yet poorly constrained water vapor response.

What we do know is that there has been a multi-decadal increase in stratospheric water vapor abundance at northern mid-latitudes. Data is lacking at other latitudes for a sufficient period of time to determine whether mid-latitude Northern Hemisphere "trends" are global in nature. In the lowermost stratosphere, a statistically significant linear trend is difficult to determine, but indications are that since late 2000 water vapor abundance has dropped in response to Tropical Tropopause Layer (TTL) temperature decreases. It is uncertain whether these stratospheric trends are accompanied by trends in upper troposphere water vapor abundance.

Although long-term changes in UTLS water vapor are neither well understood nor well observed, there are annual and interannual variations that are well understood. There is a prominent annual cycle in lower stratospheric temperatures in the tropics (with a peak to peak range on the order of 10ºC). There is a corresponding large amplitude signal in tropical lower stratospheric water vapor (with a peak to peak range of at least 2 ppmv). There are associated QBO and ENSO related variations in tropical lower stratospheric water vapor that correspond to tropical temperature variations. These indicate a reasonable understanding of specific processes that have a large impact on TTL water vapor and subsequent transport to higher latitudes and altitudes. The smaller long-term variations are significant from a radiative and chemical standpoint, but are much more difficult to measure or model due to the fact they are a small perturbation compared to other variations in LS water vapor.

Aerosols' impact on clouds

There appears to be a NH/SH difference in the distributions of relative humidity outside of cirrus clouds, with the Southern Hemisphere showing higher frequencies of large super saturations. This difference has been attributed to the onset of freezing in polluted regions (NH) occurring at significantly lower RH than in the relative clean Southern

Hemisphere mid latitudes. The freezing processes are likely to involve competition between different types of freezing nuclei. Presumably, there are less ice nuclei in the cleaner Southern Hemisphere, and hence a higher frequency of homogeneous nucleation.

The introduction of anthropogenic aerosols may also impact cloud evolution and precipitation amounts. Observed NH/SH differences in aerosol (ice nuclei) loading are presumed to be a consequence of differences in anthropogenic emissions of aerosols, though whether this is leading to significant changes in cloud properties in the Northern Hemisphere is still uncertain. One modeling study presented during the workshop showed that increasing the aerosol loading and SO_2, presumably as a result of anthropogenic activities, altered the evolution of a mixed phase precipitating cloud.

However, aerosol/cloud interactions are complex and not easily predicted. Different pathways for freezing can result in different effects on clouds. Anthropogenic aerosols can change the balance between heterogeneous, homogeneous and immersion freezing, potentially impacting clouds, precipitation, and climate. A key point in regards to climate change is that changing the number concentration or composition of upper tropospheric aerosols can alter the number of ice crystals formed. This in turn can alter the radiative balance, and consequently affect surface temperatures and other climate parameters.

Clouds' impact on aerosols

The processing of aerosols by clouds is also highly uncertain. It is qualitatively known that changes in aerosol abundance, vertical distribution, size distribution, chemical composition, and optical properties can occur through cloud processing. However, quantitative evaluation of the changes is currently not available.

The role played by aerosols, and specifically their composition, in determining the atmospheric relative humidity is not clear; some studies imply very large impacts. Similarly, the impact of aerosols on cloud thermodynamics is unclear, but the development and the impact of the clouds could, in principle, be altered greatly by aerosols.

The chemistry that takes place in the liquid cloud droplets is probably the most well-understood of the issues related to water vapor and clouds. Still, not all the chemical processes that take place in clouds are understood or quantified.

Outstanding Issues

The major needs in this area are (1) accurate global data on water vapor abundance and trends, (2) understanding of processes that lead to transport, redistribution, and physical state of water, and (3) modeling detailed processes and including them in microphysical and global climate models.

1. The first and foremost requirement is the determination of the water vapor abundance, variability, seasonal and interannual cycles, and any long-term changes in UTLS water vapor. Particular attention needs to be paid to the following:

- How accurately can water vapor or humidity in the UTLS be measured?
- What is the climatological distribution of H_2O in the upper troposphere and, quantitatively, how are interannual variations in upper troposphere humidity related to geophysical phenomena such as the QBO and ENSO cycles?
- Are regions of super saturation in the upper troposphere well characterized?
- How long of a data record is required to assess whether there is a (global) long-term change of UT humidity, given the observed spatial and short-term variability?
- What are the implications of a change in UT humidity and are models taking into account all the processes necessary to answer this question?

2. Accurate global modeling of water vapor in the UTLS is in its infancy. Many parameterizations are applied to get model water vapor fields to resemble observations. What is needed to improve model representations is a good understanding of the processes that control UTLS water vapor. In particular, we need to understand:

- The exact mechanisms for the observed dehydration of air entering the stratosphere.
  - Is it via cooling through gradual ascent, interactions with deep convection , or a combination of processes?
- What are the relative roles of convective and non-convective transport on the UTLS water vapor budget?
  - Can this be modeled, and what do the roles of these pathways tell us about the transport of other tropospheric species into the upper troposphere and stratosphere?

3. Clouds are connected with the water vapor distribution, and also with radiative and chemical processes in the UTLS. Key questions related to cloud processes and cloud modeling are:

- What is the role of cirrus and subvisible cirrus in the definition of the TTL?
- What is the role of particles in affecting both cloud processes and the composition of the TTL?
- How can both our understanding and model representations of convective transport and precipitation be improved?
- What are the best ways to upscale microphysical and dynamical processes from small-scale to large-scale models?
- What are the levels of detrainment and entrainment in deep convective clouds and how do they affect the composition of the cloud outflow and UT properties?
- What are the dynamical and microphysical processes controlling the uptake and release of soluble species in warm and mixed clouds?
- What is the impact of ice on chemistry in convective clouds and the UT?

- What are the processes controlling the formation of ice in the TTL?
  - What is the relative role of homogeneous vs. heterogeneous processes?
  - How do the natural and anthropogenic aerosols affect the ice formation?

The overarching issue that encompasses all the questions mentioned above is that we need to understand how UTLS aerosols and clouds impact both stratospheric and tropospheric chemistry and radiation and additionally how aerosol and cloud processes will be affected by a changing climate. To help in addressing these outstanding issues, *in situ* and remote measurements of water vapor, clouds, aerosols (composition, size distribution, and other properties such as hygroscopicity) are required. A thorough understanding of the accuracy of the water vapor and aerosol measurements is also needed. The nature of water makes it difficult to measure *in situ*, and its high spatial variability makes it difficult to interpret satellite measurements, which have low spatial resolution. *In situ* and remote aerosol measurements are similarly challenging, but are critical both for understanding cloud processes and validating global satellite measurements. Addressing these needs should form the cornerstones of water vapor, cloud, and aerosol research in the UTLS.

Concluding remarks:

Various research needs in the field of climate/chemistry interactions have been highlighted here. In prioritizing these needs, the following unifying set of strategies are recommended:

1. *The upper troposphere and the lower stratosphere must be studied as an interactive system.* The impact of the lower stratosphere is most pronounced on the upper troposphere and vice-versa, though their influence propagates beyond UTLS. While there are distinct demarcations in processes and timescales, these two regions are inherently coupled and to properly understand climate and chemistry in either it is essential that they be investigated together.
2. *Investigations of Tropical Tropopause Layer (TTL) in particular should be emphasized.* This region is the key gateway for transport into the stratosphere, can affect upper tropospheric composition (especially with respect to water vapor and clouds), and is essential for understanding chemical changes for short-lived gases.

3. *Disparities in the spatial and temporal resolution and coverage of in-situ observations, remote observations and models must be reconciled.* In-situ measurements can have very high temporal resolution (i.e. fractions of a second for state parameters and some gas species but up to a day for chemical filter samples) and high spatial resolution, but they are confined in location and may not be regionally representative. In contrast, satellite-based remote measurements, which are generally for a specific instant in time, have poor spatial resolution – particularly in the vertical – but have regional to global coverage. Models have moderate spatial and temporal resolution (depending on the model)

but can have complete global coverage. Issues of up- and down-scaling when comparing or combining these data sets have confounded atmospheric community. It is important that the impact of regional forcings be accurately represented while at the same time care has to be taken to derive global scale forcing and impacts.

4. *Global and targeted measurements, as well as highly coupled models need to be employed*. There are some key issues that are common to many climate studies and, hence, of importance to climate-chemistry coupling also. They include the need for highly integrated and coupled models, which couple other climate related processes with chemistry and deal with spatial inhomogeneity in processes and forcing. Coupling between complex models (i.e. atmosphere, ocean, biosphere, carbon cycle, chemistry and aerosol) constitutes the way forward if we hope to identify and better quantify the indirect climate forcings associated with atmospheric chemical constituents and to understand and predict the impacts of climate change on atmospheric composition. However, we should be aware that uncertainty in estimates of these forcings would also increase as model complexity increases. In order to reduce these uncertainties we will have to carefully evaluate the model representations of the physical, dynamical, chemical, and biospheric processes that link atmospheric chemistry and climate. This will require not only large-scale (i.e. global) campaigns and satellite observations (for example, to investigate the inter-continental transport of pollutants) but also carefully targeted campaigns that focus on a given region or on specific processes that are poorly understood and/or not well represented in models . These coupled models will have to be carefully tested (by calculating the specific observed parameter under the same spatial and temporal resolution as the observations) against observations before a reasonable level of confidence can be achieved in their evaluation of radiative forcings and climate feedbacks.

5. *Transfer knowledge across measurement and modeling domains in an appropriate way*. This includes transforming information from laboratory studies to the atmosphere, from microphysical models to regional and global models, from process studies to regional and global models, and from regional models to global models.

6. *Study regional air quality and climate together whenever possible*. It will be beneficial to both communities to recognize the commonalities in the climate and regional air quality issues with regards to aerosols, short-lived chemically active greenhouse gases, and tropospheric ozone. These include common investigational approaches and a common interest in species formation, sources, and chemistry.

7. *Recognize the differences between the radiative forcings by well-mixed and spatially inhomogeneous species, and use them appropriately*. The radiative forcing concept, which was originally formulated for the global and annual mean climate system, has been applied to even smaller spatial domains and to shorter

time-averaging periods, such as when assessing the impact of short-lived species with a distinct geographical and/or seasonal character (*e.g.*, ozone or other indirect forcings). Several recent studies have shown that these in-homogeneous forcings result in climate sensitivities that are characteristically dissimilar to the forcing/sensitivity relationship of long-lived, globally distributed greenhouse gases. For these species, climate sensitivity is quite dependent on the spatial (altitude and geographical) distribution of the applied forcing agent. The metrics used to quantify the impact of greenhouse gas emissions on climate (*e.g.*, Global Warming Potentials, or GWPs) have to be calculated with care and modified or refined in order to include species involving chemical species/aerosols with heterogeneous forcings and indirect forcings on the climate system (for example, CH_4, CO, NO_x and NMHCs).

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Figure 5. Photograph of the participants in the Climate Chemistry Interactions Workshop.

Bibliography

Folkins, I., K. K. Kelly, and E. M. Weinstock, A simple explanation for the increase in relative humidity between 11 and 14 km in the tropics, *J. Geophys. Res.,* Vol. 107, No. D23, 4736, 10.1029/2002JD002185, 2002.

Haag, W., G. Kärcher, J. Ström, U. Lohmann, J. Ovarlez, and A. Stohl, Freezing thresholds and cirrus cloud formation mechanisms inferred from in situ measurements of relative humidity, *Atmos. Chem Phys.,* 3, 1791-1806, October 27, 2003.

Hartmann, D. L. and K. Larson, An important constraint on tropical cloud-climate feedback, *Geophys. Res. Lett.,* Vol 29, 1951, 10.1029/2002GL015835, 2002.

Jacob, D.J., Heterogeneous chemistry and tropospheric ozone, *Atm. Env.*, 34,2131-2159, 2000.

Kärcher, B, and U. Lohmann, A parameterization of cirrus cloud formation: Homogeneous freezing of supercooled aerosols, *J. Geophys. Res.,* 107, D2, 10.1029/2001JD000470, 2002.

Kärcher, B, and U. Lohmann, A parameterization of cirrus cloud formation: Homogeneous freezing including effects of aerosol size, *J. Geophys. Res.,* 107, D23, 4698, 10.1029/2001JD001429, 2002.

Kärcher, B. and U. Lohmann, A parameterization of cirrus cloud formation: Heterogeneous freezing. *J. Geophys. Res.,* Vol. 108, No. D14, 4402, 10.1029/2002JD003220, 2003.

Soden, B. J., R. T. Wetherald, G. L. Stenchikov, A. Robock, Global cooling after the eruption of Mount Pinatubo: A test of climate feedback by water vapor, *Science*, Vol. 296, 727-730, 2002.

Travis, D.J., A. M. Carleton, and R. G. Lauritsen, Contrails reduce daily temperature range, *Nature*, Vol. 418, 601, 2002.

Nenes, A, R. J. Charlson, M. C. Facchini, M. Kulmala, A. Laaksonen, and J. H. Seinfeld, 2002: Can chemical effects on cloud droplet number rival the first indirect effect? Geophys. Res. Lett., Vol 29, No. 17, 1848,10.1029/2002GL015295

Feingold, G., 2003: Modeling of the first indirect effect: Analysis of measurement requirements. Geophys. Res. Lett., 30, No. 19, 1997, doi:10.1029/2003GL017967.

"Ozone-climate interactions", Air pollution research report 81, European communities, 2003. ISBN 92-894-5619-1.