

Science Plan and Implementation Strategy

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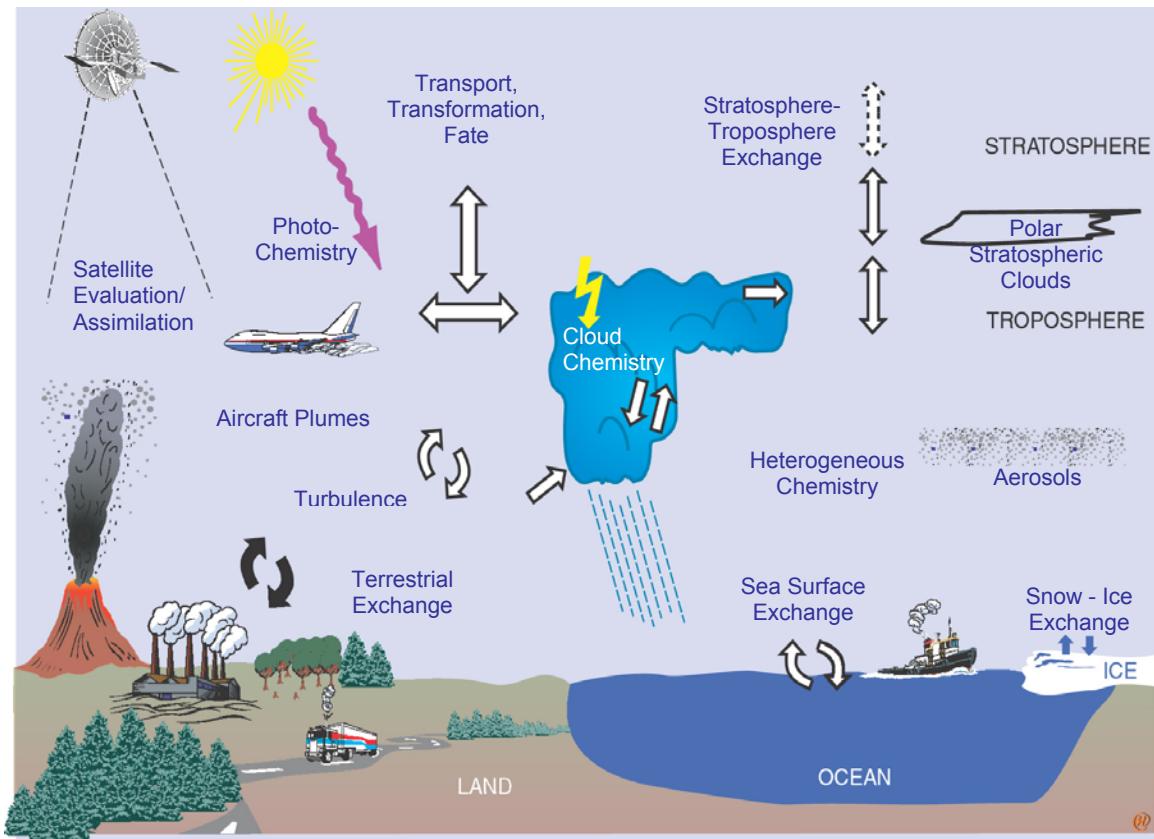


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SUMMARY

The International Global Atmospheric Chemistry (IGAC) Project, under joint sponsorship of the Commission on Atmospheric Chemistry and Global Pollution (CACGP) of the International Association of Meteorology and Atmospheric Sciences (IAMAS) and the International Geosphere- Biosphere Programme (IGBP), was created in the late 1980s to address growing international concerns over rapid changes observed in Earth's atmosphere. The past decade of international research, much of which was initiated and coordinated within IGAC, has greatly increased our understanding of the chemical composition of the troposphere, the fluxes of chemical components into and out of the troposphere, and the processes controlling the transport and transformation of chemical components within the troposphere (Brasseur et al., 2002). Through a large number of projects and activities, IGAC has created a worldwide community of scientists, enhancing international cooperation towards understanding global atmospheric chemistry.

Much of IGAC's research efforts during its first decade were directed towards assessing the effects of anthropogenic emissions on the 'background' atmosphere ('background' atmosphere defined here as the present-day atmosphere in areas some distance away from the emission source regions). While questions remain concerning the point at which observed global/regional mean trends in component concentrations (signal) unambiguously rise above background natural variability (noise), it is now well recognized that human activities have perturbed the chemical composition of the atmosphere on local, regional, and global scales. These perturbations arise from 1) emissions from fossil fuel/bio fuel combustion and other industrial processes, 2) anthropogenic enhancements of biomass burning, and 3) increases in the lofting and transport of mineral dust through human-induced land-use changes. On regional scales, air pollution (defined here as elevated pollutant concentrations and deposition fluxes resulting from anthropogenic emissions) is a serious and growing problem in many parts of the world. In the industrialized mid-latitudes of the Northern Hemisphere, elevated concentrations of ground-level ozone and particulate matter are of concern from a human health perspective. Moreover, the world's major agricultural regions are co-located with industrialized population regions in the northern mid-latitudes. As a consequence, the impacts of regional air pollution on world food production can be significant. In other regions of the world, such as tropical and extra-tropical Asia and Africa, anthropogenic emissions (which are already quite high) are projected to increase substantially in the coming decades as a result of the increasing energy and food demands of a growing population. In addition, the development and growth of megacities (cities with populations >10 million and high population densities) and urban agglomerations (a large core city with surrounding satellite cities which together act a large point source in a region) will necessitate their consideration in studies of regional and global atmospheric chemistry. The past decade of international research has clearly revealed a large number of atmospheric chemistry issues facing society as well as the challenges of studying and managing an integrated Earth System.

These issues and challenges were discussed within several IGAC symposia and workshops between 1999 and 2002. From this series of discussions several key issues of climate and air quality over large regional and global scales emerged:

1. What is the role of atmospheric chemistry in amplifying or damping climate change?
 - 1.1. What are the relative roles of stratosphere-troposphere exchange, anthropogenic and natural precursor emissions, and *in situ* photochemical processes in controlling O₃ and its effect on climate change?
 - 1.2. What are the sources, sinks, distributions and properties of aerosol particles and their direct radiative effects on climate?
 - 1.3. What are the effects of aerosol particles on clouds, precipitation, and regional hydrological cycles?
 - 1.4. How will changing emissions and depositions of gases and aerosol particles affect spatial patterns of climate forcings?
2. Within the Earth System, what effects do changing regional emissions and depositions, long-range transport, and chemical transformations have on air quality and the chemical composition of the planetary boundary layer?
 - 2.1. What are the export fluxes of oxidants, aerosol particles, and their precursors from continents (e.g., mega-cities, biomass burning, desert dust) to the global atmosphere?
 - 2.2. What are the impacts of intercontinental transport on surface air quality?
 - 2.3. How will human activities transform the cleansing capacity of the future atmosphere?

Addressing these global research questions will require international coordination and collaboration since the atmosphere knows no political boundary. The international atmospheric chemistry community and their interdisciplinary partners will need to:

- *accurately determine global distributions of both short and long lived chemical components in the atmosphere and document their changing concentrations over time.*
- *provide a fundamental understanding of the processes that control the distributions of chemical components in the atmosphere and their impact on global change and air quality.*
- *improve our ability to predict the chemical composition of the atmosphere over the coming decades by integrating our understanding of atmospheric processes with the response and feedbacks of the Earth System.*

With this knowledge we should have the tools necessary to develop judicious policies to manage the health of our atmosphere and its role in global change. Enhanced outreach to the public will be vital to ensure that the new knowledge results in changes in public attitude, policy and legislation.

SCIENTIFIC QUESTIONS

Theme 1 - What is the role of atmospheric chemistry in amplifying or damping climate change?

During the past decade the atmospheric chemistry research community has identified and quantified the distributions of a number of radiatively-active substances. However, in most cases, the level of scientific understanding in the calculated radiative forcing of these various substances is still extremely low. A major theme of IGAC will be to reduce the uncertainties in the calculated radiative forcing of the climate system and to understand the role of atmospheric chemistry in amplifying or damping climate change.

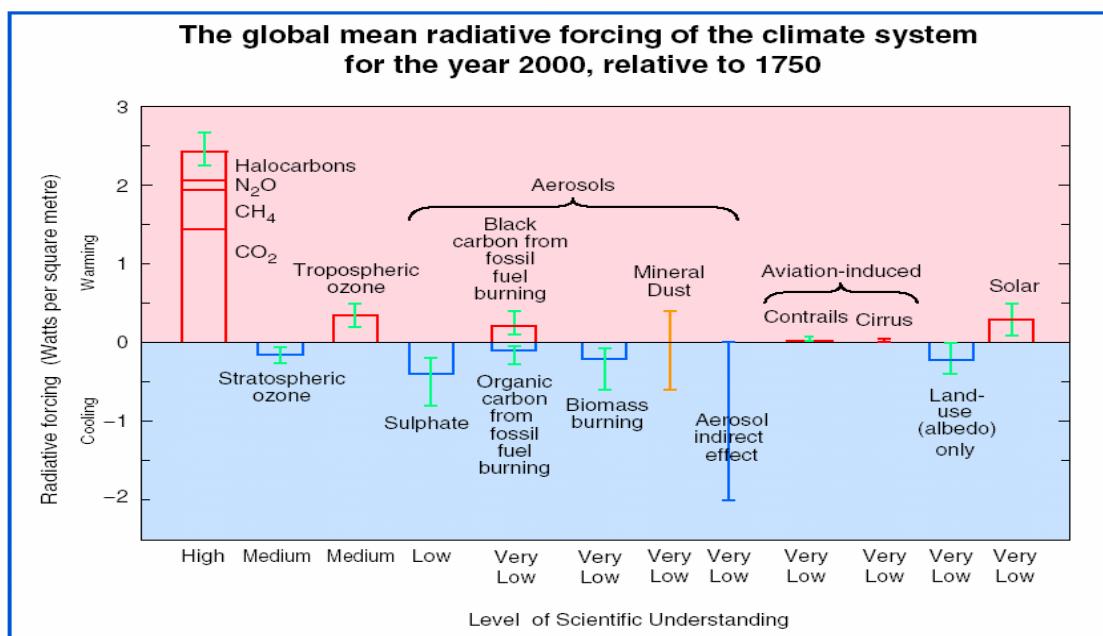


Figure 1. Anthropogenic and natural factors causing a change in radiative forcing from year 1750 to year 2000. (IPCC, 2001).

Part of the uncertainty associated with the calculated radiative forcing by various components is in defining the background atmosphere and the natural variability of this background. The "background" atmosphere, despite decades of research within IGAC and other programs, is still not clearly defined. Normally it has been taken to describe the chemistry of remote regions of the atmosphere, free from human influences. One of the lessons of past atmospheric chemistry research is that such regions apparently no longer exist - even the remotest regions, such as the central Pacific and the polar stratosphere show substantial influences from anthropogenic emissions at least for part of the year. Many attempts have been made to simulate pre-industrial atmospheric chemistry, but these modelling studies are highly uncertain and vary widely in their results; pre-industrial trace gas measurements are too sparse and (mostly too uncertain) to

allow a reliable assessment to be made. It is also unclear whether the changes in climate need to be accounted for in defining the background state of atmospheric chemistry versus anthropogenic changes. Changes in circulation patterns (source-receptor regions) and rainfall, for instance, can have substantial effects on various aspects of atmospheric chemistry. Furthermore, the natural variability in circulation and emissions patterns, which can be quite complex, needs to be taken into account in defining the range of states which can qualify as "background". Nevertheless, a "reference state" is needed in order to define the current and future changes brought about by human activities, and to be able to put this in a clear socio-political context. Furthering our understanding of the background state of the atmosphere and its natural variability will be (whether formally or informally) an underpinning activity within all of the future IGAC projects.

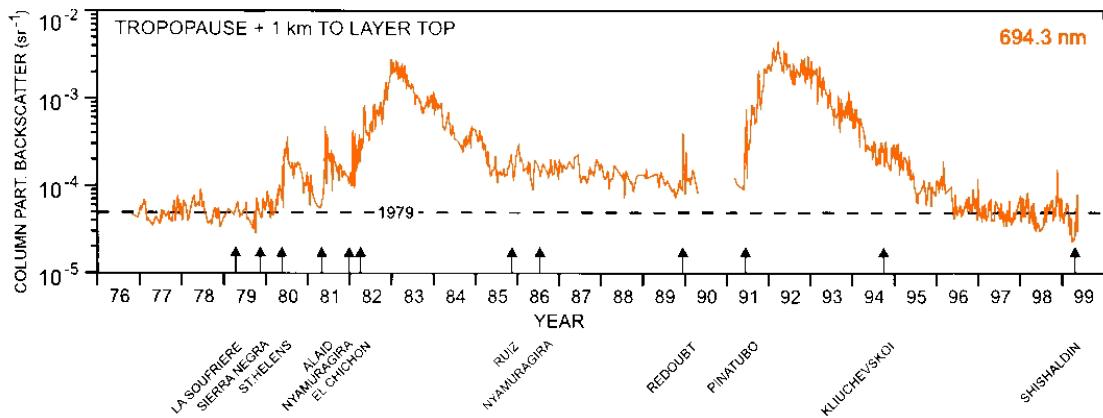


Figure 2: There is a large natural variability in the stratospheric aerosol optical depth due to the eruption of volcanoes. Natural variability is an important characteristic of essentially all variables describing the Earth system.

The research activities within IGAC addressing these issues have been divided into four questions covering emissions and deposition of gases; the processes controlling the distribution of ozone and its precursors; the sources, sinks, distributions and properties of aerosol particles and their direct radiative effects on climate; and the effects of aerosol particles on clouds, precipitation, and regional hydrological cycles.

Question 1.1 What are the relative roles of stratosphere-troposphere exchange, anthropogenic and natural precursor emissions, and *in situ* photochemical processes in controlling ozone and its effect on climate change?

Background

During IGAC-I there has been great progress in increasing the coverage of ozone observations (mapping and climatology) and in elucidating processes affecting its photochemistry and transport. Much of the advance is due to campaigns sponsored within IGAC activities. In addition, models of various scales, from zero-D to coupled-global-chemistry-transport (CTMs) have improved our ability to interpret observations and to look backward and forward in time. The in-situ view and modeling complement an enhanced global mapping capability provided by a new generation of satellite instruments.

Three areas emerge in tropospheric ozone chemistry as defining issues for IGAC. The first, current global tropospheric ozone observations (and related constituents and parameters) do not provide adequate spatial, vertical or temporal resolution to quantify the processes controlling ozone and its effect on climate change. The second issue focuses on transport processes as sources and sinks of ozone in the troposphere. Deep convective mixing and stratosphere-troposphere exchange closely tie this issue to SPARC. Boundary layer exchange and long-range horizontal transport tie this issue to question 2.2. The third issue focuses on basic chemical processes related to ozone (and other oxidant) formation and destruction. This third issue is discussed in more detail in question 2.2.3 under the broader heading of the cleansing capacity of the atmosphere.

Approach

Measurements of Ozone and Ozone Precursors

Global Measurement of Ozone

Several initiatives during the past decade were aimed at improving the expanding, but still incomplete, global picture of tropospheric ozone. There are several approaches needed for global ozone mapping: satellite, in-situ, and modeling. Satellite climatologies of tropical and approximate mid-latitude (several month averages) were all that was available from space in the early IGAC period. Refinements now provide daily, near real-time tropospheric ozone column measurements in the tropics (TOMS, method introduced in 1997) and extra-tropics (GOME, launched in 1995). Coverage and accuracy are sometimes limited and clouds and aerosol particles are known interferences in the retrievals. A new generation of ozone sensors (three on NASA's AURA plus ESA's SCIAMACHY) will be operational during IGAC along with CO and methane column measurement from MOPITT and other instruments. The community will benefit from the data but also be challenged to use it to maximum advantage. Models that assimilate some of the space-based data are developing; some predictive capability is anticipated.

Generally speaking, satellites will provide column ozone (and other trace gas) amounts. In-situ measurements should be an IGAC activity in two respects. Ozone combined with other chemical measurements in campaigns will be required to better

define the photochemical budget of ozone and the interaction of chemical and dynamical processes within a given regime. Vertical profiles from soundings and commercial aircraft at regular locations and intervals are needed. For example, a network of regular ozonesonde launches records large-scale variability, stratosphere-troposphere exchange (STE), pollution layers, convective transport, seasonal and interannual variations, along with vertical structure. In-situ measurements on commercial aircraft (MOZAIC and CARIBIC) are also providing routine vertical profiles at major transportation hubs.

Regular soundings are also the only way to derive global ozone trends throughout the troposphere. At WOUDC (World Ozone and UV Data Centre, at Environment-Canada, Toronto), ozonesonde data are archived for open community use. During the latter stages of IGAC-I, a focused effort to coordinate ozonesonde launches in the southern tropics and sub-tropics (SHADOZ = Southern Hemisphere Additional Ozonesondes) was successful in adding > 1300 ozonesonde profiles to a severely undersampled region. Balloons take on greater value when supplemented with campaign and satellite observations. For example, during the IGAC- SAFARI-92/TRACE-A (Southern African Fire Atmospheric Research Initiative/Transport and Atmospheric Chemistry near the Equator-Atlantic) campaigns, data from more than a hundred ozonesondes were used to evaluate the relative contributions of south Atlantic ozone pollution from the two adjacent continents. The WMO/GAW activity is a mechanism for adding ozone sounding capabilities to developing nation observing programs for global change. IGAC can also work with IGOS-P (Integrated Global Observing System) to develop a more complete global network.

Measurements of Ozone Precursors

The budget of tropospheric ozone is controlled primarily by photochemical production and loss within the troposphere. In order to understand and predict the trends of ozone, it is essential to have a good knowledge of the distributions and budgets of ozone precursors. The distributions of many key ozone precursors are highly variable and inhomogeneous because of their relatively short lifetimes and widely scattered emission sources. For example, the mixing ratio of nitrogen oxides ($\text{NOx} = \text{NO} + \text{NO}_2$) can vary by several orders of magnitude, from a few hundred ppbv in highly polluted urban centers to a few pptv in the remote troposphere. Some hydrocarbons, particularly naturally emitted hydrocarbons, can vary by similar magnitude. The large variability and lack of knowledge in some important sources of NOx as well as hydrocarbons make it difficult to accurately simulate the distributions of these ozone precursors in models. Because the photochemical production of ozone is a non-linear function of NOx and hydrocarbons, ozone budgets calculated by models can have large uncertainty as a result of the difficulty in simulating the distributions of the ozone precursors. Thus it is essential to have a good knowledge of the distributions and budgets of ozone precursors. Measurements of ozone precursors will not only provide important information on the distributions of these components, but also serve as pivotal tests for evaluating model capability.

In situ measurements of ozone precursors, including NO , NO_2 , NOy and a large number of NMHC, at ground stations and on aircraft during IGAC have been highly successful. In particular, aircraft measurements over remote areas such as the Pacific Ocean (IGAC/APARE), Atlantic Ocean (IGAC/NARE, TRACE-A, SAFARI), and Indian

Ocean (INDOEX) have greatly increased our knowledge of the distributions and budgets of ozone and ozone precursors. These measurements need to be continued. Furthermore, concurrent satellite measurements (GOME, TES) of ozone and ozone precursors can potentially revolutionize our understanding of the budgets and distributions of ozone and ozone precursors by substantially increasing the spatial and temporal coverage. Finally, satellite measurements will also provide the distributions of key tracers that can be help address key scientific questions of stratosphere-troposphere exchange discussed in the next section.

Other measurements need to be emphasized are components that contribute significantly to the budgets and distributions of odd hydrogen components, i.e. OH and HO₂. They are also pivotal to the cleansing capacity of the atmosphere. Water vapor is an essential component in the production of OH and HO₂. Accurate measurements and good spatial and temporal coverage of water vapor are still lacking, especially in the upper troposphere and lower stratosphere. H₂O₂, CH₃OOH, CH₂O, acetone, and other similar organic compounds have been shown to be important sources of OH and HO₂ in certain regimes of the troposphere. Satellite techniques for observing these components are currently being developed.

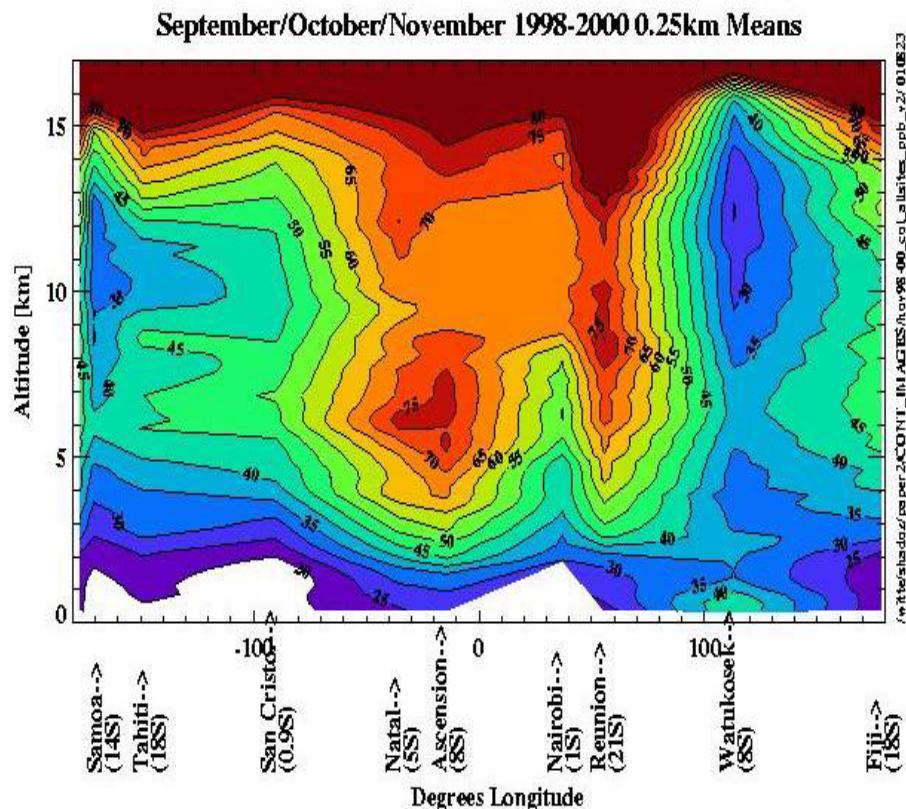


Figure 3. An accumulation of ozone over the south tropical Atlantic Ocean can be seen as a year-round feature, shown here for September-October-November. It feature appears in satellite data, aircraft sampling and in soundings (these from SHADOZ: <http://croc.gsfc.nasa.gov/shadoz>). The tropopause is more a "layer" than a thin barrier and dynamical as well as photochemical inputs of ozone appear to contribute to the enhanced ozone concentrations.

Investigations of Transport

IGAC is interested in four dynamical processes that affect global tropospheric ozone distributions: injection from the stratosphere (stratosphere-troposphere exchange) via large-scale circulation and small-scale processes (tropopause folds); deep convection; large-scale horizontal advection; and exchange between the Earth's surface and boundary layer.

Stratosphere-Troposphere Exchange

Originally it was believed that all tropospheric ozone was stratospheric in origin, owing to its relatively long photochemical lifetime and clear connection through folding events in mid-latitudes and large-scale circulation and mixing in the tropics. As photochemical sources of ozone became evident in polluted regions, and models showed that globally the photochemical source was comparable to transport, more attention was given to the role of chemistry. IGAC focused on experimental or modeling initiatives to elucidate processes that make ozone on regional to global scale - combustion in urban areas, biomass burning in more rural locations, and lightning, a major source of the ozone precursor, nitric oxide. Recent observations point again to the critical role of dynamics and transport in determining tropospheric ozone observations. For example,

- (1) The view of the tropopause as a fixed barrier between stratosphere and troposphere in the tropics has been modified with ozone and tracer data from aircraft (MOZAIC) and soundings. Stratospheric ozone in the subtropics penetrates as low as 2-3 km (observed at La Reunion (21°S, 55°E) and at the GAW station Cerro Tololo (30°S, 70°W, 2200 m a.s.l) in connection with cut-off lows and a deep trough).
- (2) Throughout the tropical free troposphere there is a zonal wave-one feature, referring to 10-15 Dobsons Unit (1 DU=2.69x10¹⁶ molec/cm²) more ozone column thickness over the Atlantic than the Pacific. The wave-one persists throughout the year. Although seasonal biomass burning contributes to a south Atlantic "ozone maximum" and the wave, so do lightning and subsidence of upper troposphere/lower stratospheric ozone at all seasons.
- (3) Observational campaigns at mid-latitudes in the 1990's showed that tropopause fold events are very common. Sharp gradients at frontal edges can be discriminated in the satellite ozone record. At the polar front the intrusions of stratospheric air into the troposphere are visible. Along the trailing edge of these stratospheric episodes, "conveyor-belt transport" of pollution from continent to continent can be discerned. In a similar manner it has been shown that most mid-latitude ozone pollution events are associated with the penetration of moist, subtropical air beyond conventional "tropical" latitudes. In summary, we must bear in mind that air masses know no latitude boundaries and that definitions of tropics-sub-tropics-mid-latitudes-polar regions are fluid.
- (4) A connection between ozone and processes associated with tropical climate variability (eg El Niño/ La Niña, the Indian Ocean Dipole) has been established using satellite data to examine the past 20 years. Even during the pollution that occurred following 1997 Indonesian fires triggered by El Niño,

a large increase in upper tropospheric ozone brought about by perturbed dynamics was comparable to the pyrogenic ozone over the larger Indian Ocean basin.

Convection

Building on mid-1980's observations, a number of IGAC-related and other campaigns have established convection as a mechanism for introducing boundary-layer air into the free troposphere. The degree to which the tropopause is penetrated by deep convection, leading to mixing of stratosphere and tropospheric air, is still being determined. In the tropics, the rapid decrease in the penetration of deep convection into the higher levels of the upper troposphere has led to the definition of a tropical tropopause layer (TTL), that is several km thick, and extends from the secondary tropical tropopause (STT, ~12-14 km), above which convective outflow begins decreasing rapidly, up to the tropical tropopause (TT, ~16-18 km), defined by the temperature minimum. The nature of these layers, including the climatology of water vapor and the evolution of thin cirrus (and its radiative forcing role) is a subject of intense investigation in field programs. Convection is not only an efficient transport mechanism but also is important in scavenging chemical components from the atmospheric via convective precipitation. In particular, the role of ice in the convective clouds is a crucial. Convection also produces lightning, particularly intensely over land, as seen by satellites and ground-based lightning detectors. The nitrogen oxides from lightning lead to ozone formation. Assessing the role of convection in global ozone is left to the integrating perspective of models. Convective and lightning parameterizations are one cause of variations among models used by different research groups.

Advection/Frontal Transport

It could be said that the regional field initiatives sponsored under IGAC (eg APARE, NARE, STARE) have shown how truly global pollution transport is. This was observed during the ACE-Asia campaign in particular. During the 1990's campaigns in the Indian Ocean region and tropical Pacific found high pollution levels in areas expected to be relatively free of human influence. Clear evidence of North American transport reaching Europe and Asian transport reaching North America have been found. Accordingly, ITCT (Intercontinental Transport) is a recent IGAC initiative that is expected to be a major driver of IGAC activities.

Surface-Boundary Layer and Boundary Layer-Free Troposphere Exchange

Chemical processes in the boundary layer determine ozone concentrations near source regions. Inputs from biosphere-hydrosphere and human activities supply trace gases that lead to ozone formation. Complex processes involved in the chemical transformations within the boundary layer are an obvious IGAC focus, but the interaction between boundary layer and free troposphere determines the larger regional to global impact. Fluxes of ozone and its precursors from boundary layer to free troposphere are inadequately measured and not satisfactorily represented in simulations. Multi-scale models are needed, and for ozone, its precursors and associated gases (eg CO, HCHO, BrO) global satellite observations are useful, especially if vertical resolution can be improved.

Radiative Forcing by Tropospheric Aerosol

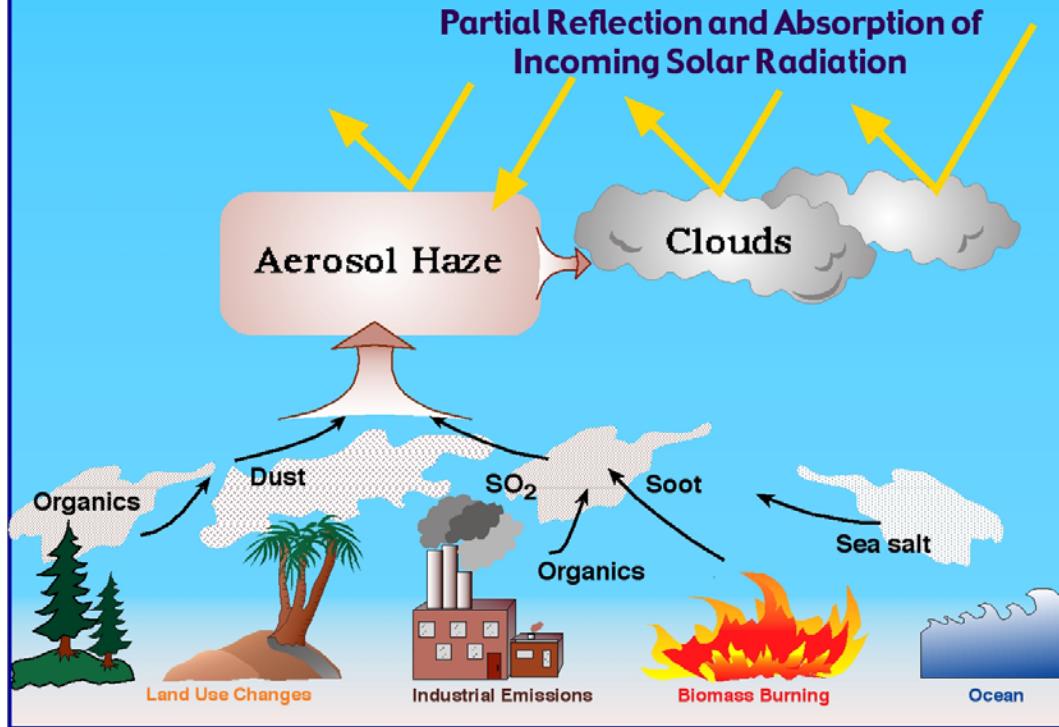


Figure 4. Atmospheric aerosol particles can affect the Earth's radiation budget directly by absorbing and scattering solar radiation. Additionally, aerosol particles can act as cloud condensation nuclei and thereby determine the initial cloud droplet number concentration, albedo, precipitation formation and lifetime of warm clouds.

Question 1.2 What are the sources, sinks, distributions and properties of aerosol particles and their direct radiative effects on climate?

Background

Atmospheric aerosol particles are the largest source of uncertainty in current IPCC estimates of climate forcing due to human activities. Several existing or imminent satellite missions offer an extraordinary opportunity for improving global knowledge of aerosol particle distributions. However, to understand the climate perturbations, aerosol radiative effects must not only be quantified but must be attributed to chemical components and, ultimately, sources. Thus, the critical challenge addressed by this activity will be a global aerosol particle assessment that links aerosol radiative properties to aerosol chemical components. Enormous uncertainties in the source strengths and lifetimes of major aerosol components, as represented in chemical transport models, require that this assessment be based on observations. Chemical attribution of observed, global aerosol optical depth (AOD) will provide a foundation for evaluating the present

direct climate forcing by anthropogenic aerosol particles as well as for estimating past and future aerosol particle impacts, including those associated with preindustrial aerosol particles (e.g. dust and biomass burning) and with aerosol particle source changes projected under various emission scenarios.

Approach

This project seeks the best estimate of existing AOD (including its geographical and temporal variations) and seeks to attribute existing AOD in terms of major aerosol components - sulfate, organic compounds, black carbon, nitrates, dust, and sea salt. Achieving this global characterization will require the integration of aerosol-sensing satellites, chemical transport models, and in-situ measurements. While satellites and chemical transport models operate naturally at the global scale, aerosol chemical measurements are necessarily local. Therefore, a major role of IGAC will be to foster the acquisition of high quality, consistent, and scientifically useful data across diverse regions. In addition, IGAC will provide a forum to identify the most critical regions and processes in order to encourage the scientific community to focus on the most important gaps in this global characterization.

Preliminary assessment and strategic planning

This will involve intercomparisons of chemical transport models (CTMs), intercomparisons of satellite-derived AOD climatologies, and integrated analysis of results from recent field campaigns. The goals will be (i) to form preliminary assessments of global AOD and the fraction of AOD due to major natural and anthropogenic components, (ii) to identify the major sources of uncertainty in these present estimates, and (iii) to develop a research strategy for reducing those uncertainties.

Coordination of ground-based measurements

A number of organizations currently support ground-based measurements of aerosol chemistry, although few sites have coordinated methodologies for sampling and reporting data, and even fewer have long-term support for collocated, simultaneous measurements of AOD. Use of such ground-based sites for chemical attribution of AOD relies on the assumptions that chemical fractions are homogeneous throughout the boundary layer and that boundary layer aerosol particles dominate the entire atmospheric column AOD. In coordination with column-closure experiments that test these assumptions ground-based chemical data can play a major role in the global assessment. However, considerable coordination will be required given that (i) measurement methods must be consistent and data quality must be known, (ii) chemical measurements must be collocated and simultaneous with AOD measurements, and (iii) local data must be pulled into a central archive for integrated analysis in the context of chemical transport models and satellite observations of AOD. Two high-leverage projects that can be achieved in under five years are (1) implementation of well-calibrated AOD measurements in polluted regions where chemical measurements are already being made and (2) implementation of aerosol chemical measurements at selected AERONET sites which are proving to be useful not only for satellite calibration (and evaluation) but also as a tool for studying aerosol particles.

Coordination of systematic aircraft measurements

Methodologies for “column closure” experiments with aircraft have advanced rapidly in recent years. These experiments produce tightly constrained, vertically-resolved information on aerosol particle properties that provide an excellent basis for attribution of AOD. What is needed now is to gather a statistically significant number of such data sets coincident with satellite overpasses in regions of high impact. Another mode of aircraft measurement that could provide large, semi-random data sets for chemical characterization beneath satellites would involve routine transit flights across regions of significant aerosol particle loading (e.g. Washington DC to Bermuda off the U.S. East Coast). These would serve the critical purpose of assessing horizontal variation, especially at the edges of globally significant aerosol particle plumes. Dedicated research aircraft would be required; conceivably, small or even unmanned aircraft could be deployed for this purpose. Flights would be mostly in the boundary layer (or main aerosol particle layer, if aloft) with occasional profiles to assess vertical variations.

Design of targeted experiments to address key uncertainties

To resolve critical questions that underlie the large discrepancies between models and observations, laboratory results must be combined with *in-situ* process studies to evaluate the accuracy and completeness of model parameterizations. Experiments need to be organized with sufficient measurement platforms and instruments to over-constrain a specific chemical or microphysical mechanism. Such targeted process studies rely on coordinated modeling to identify regions where specific processes are not well known and may have large impacts. An example is the processes that contribute sulfate, nitrate and organic components to mineral dust in Asian and Saharan outflow layers, which may mean that as much as 50% of “dust AOD” originates from man-made pollution sources. This question requires quantifying sources of these components as well as the processes that control their gas-phase reactions, gas-to-particle transfers, and surface reactions through laboratory studies, then measuring the evolution of these components *in-situ* by means of Lagrangian type experiments. The high cost and high level of coordination required for such an effort to measure a time history of chemical evolution in both the gas and particle phases necessitates multi-national coordination and funding.

Satellite-derived aerosol properties (cloud-clearing bias)

The global aerosol particle characterization envisioned herein will largely be rooted in satellite observations. For this reason, it is essential to understand the extent to which satellite observations of AOD are biased by the need to avoid cloud contamination. Targeted studies could assess whether aerosol particle concentrations (both dry mass and ambient extinction) are systematically higher in the vicinity of clouds and whether chemical composition varies systematically as a function of cloud proximity. Such investigations may need to consider a variety of scales from the synoptic (high-pressure ridges versus low pressure troughs) to the mesoscale (updraft versus downdraft regions within cloud complexes) to the scale of individual clouds (mapping concentration as a function of distance from cloud edge).

Aerosol particle variability

Besides providing fundamental insights into the nature of the aerosol particle phenomenon, variability information is needed at sub-grid scales for practical reasons - e.g. to assess the effects of time/space averaging by models and the effects of time/space offsets for measurement intercomparisons. In addition, studying variability is critical to establishing the characteristic times and distances for the processes that change aerosol particle composition and loading. Measuring variability requires acquisition of statistically significant amounts of data at a resolution that is higher than the scales to be investigated and over domains large enough to encompass the scales to be investigated.

Question 1.3 What are the effects of aerosol particles on clouds, their optical properties, precipitation, and regional hydrological cycles?

Background

The indirect radiative effects of aerosol particles are currently the most uncertain of the known climate forcings (IPCC, 2001). They also represent an important nonlinearity in the climate system. The relationship between the forcings (e.g., changes in aerosol particle concentration and properties) effects (such as changes in cloud albedo, lifetime, extent, or precipitation development) and climate responses (e.g., changes in surface temperature or precipitation patterns and intensity) are presently very poorly understood and are expected to be highly nonlinear. A major challenge in this respect is bridging the gap between the scales on which the processes causing the effect happen (spatial scales of micrometers and time scales of seconds) and the scales on which the effect is expected to be important (regional/global spatial scales and time scales of days).

Atmospheric chemistry is central to understanding the indirect radiative effects of aerosol particles. At a fundamental level, we need to understand and be able to predict the ways in which aerosol particles and gases are incorporated into cloud droplets, and the way that they influence the development of the droplet size distribution, optical properties and phase of clouds. The chemical composition of aerosol particles (both the organic and inorganic components) is central to this issue. We need to develop a deep understanding of a multiphase, multicomponent dynamical system with nonlinear feedbacks. A multitude of information is necessary to deconvolute this intricate system. Because of this complexity, any strategy to develop a predictive capability regarding the indirect effects of aerosol particles must be centered around a focused program intimately linking observations (both *in situ* and remotely sensed), laboratory studies and model development.

The central question to be addressed is:

“What is the role of aerosol chemical composition (both inorganic and organic) in determining the indirect radiative forcing of aerosol particles?”

In order to answer the question, we need to determine the processes by which different aerosol particle and gaseous components contribute to determining the microphysical and radiative properties at cloud top, as well as how they affect precipitation development. This means that we must determine the effects that *all* the

various chemical components have on cloud development, especially the organic components for which present knowledge is extremely limited. In addition, we need to be able to detect and quantify changes in cloud property influenced by aerosol particles. Detection of the effects is central in testing the veracity of model predictions.

Concrete results must be obtainable in a finite amount of time (a few years). There must be logical check points along the way to keep the program on track and insure that progress towards fulfilling the objectives of the program is being made.

Approach

This program must be very focused, since it is impossible in a practical sense to take on all the various aspects of the problem at the same time. It must have a combination *in situ* measurements of cloud and aerosol microphysics and chemistry, both detailed process and large-scale models, and airborne and satellite remote sensing measurements of cloud radiative characteristics to arrive at a better understanding of the processes relating aerosol physical and chemical properties, cloud albedo and precipitation development.

Laboratory studies

Laboratory studies can be performed to explore processes seen or expected in nature under more controlled conditions. Fundamental properties of organic compounds or mixtures of organic and inorganic components such as water solubility, vapor pressure over aqueous solutions, surface activity, etc. have not been determined for most organic compounds in the atmospheric aerosol, and laboratory studies should be performed to produce this information. These quantities are necessary to develop thermodynamic equilibrium models for aerosol particles that include organic components, and also for further developing detailed process models of aerosol-cloud interactions.

Observations

Observational campaigns provide the most direct information about what is happening in the atmosphere, but are limited in time, space, and to the conditions that Nature offers. Measurements will necessarily involve airborne observations of the microphysical and chemical properties of below-cloud aerosol particles, of cloud droplets and interstitial particles, and of the upwelling radiation above cloud top. The measurements will help us understand how inorganic and organic aerosol particles influence the microphysical development of the cloud drop size spectrum throughout the depth of the cloud (including precipitation development) to cloud top, where the radiative properties of the cloud are largely determined. These *in situ* measurements will be coupled with satellite retrievals of cloud albedo, effective radius, and precipitation.

The initial application of this approach would be best performed in an area where we expect substantial gradients in organic aerosol particles collocated with low-level clouds. A specific example would be an urban plume advecting into a relatively non-polluted marine boundary layer. Another example could be in a situation where the impact of carbonaceous aerosol particles are known to be of importance on a regional scale, such as biomass burning plumes in the boundary layers of central Africa or Amazonia. The rationale for this approach would be to contrast areas having the same dynamical characteristics, but that are characterized by very different chemical systems (aerosol

particles and gases, organic and inorganic aerosol particles). This would help to isolate the effects of organic aerosol particles, but still retain the advantages of *in situ* observations in the real atmosphere.

Modelling

Models are needed to explore situations not encountered or observable in field observations, and to be able to have a predictive capability. Because *in situ* and satellite observations alone cannot provide a complete picture of the atmosphere, we need models to test plausible mechanisms/hypotheses that arise from the analysis of the data. The models will be used to learn more about the processes involving organic aerosol particles that influence cloud properties. From the detailed model investigations, we will be able to develop parameterizations for large-scale models that are based on a sound knowledge of the underlying physics and chemistry at work in the clouds. We will obtain a better understanding of which processes that can safely be ignored or treated coarsely in large-scale models, and which processes must be treated in more detail.

Question 1.4 How will changing emissions and deposition of gases and aerosol particles affect spatial patterns of climate forcing?

Background

Observed changes in atmospheric composition provide the most robust and most dramatic evidence for human influences on the Earth System. Studies of future scenarios indicate that, while various anthropogenic and natural factors will contribute to climate change, continuing modification of atmospheric composition will become an increasingly dominant determinant of climate over the next century. This is likely to be the case even if greenhouse gas emissions are managed so as to stabilize changes in the climate system in accordance with the United Nations Framework Convention on Climate Change (UNFCCC).

The ramifications of atmospheric change involves all research areas covered by the Earth Systems Science Partnership (ESSP) of IGBP, the World Climate Research Program (WCRP) and the International Human Dimensions Program (IHDP). One of the roles of IGAC within this partnership will be to ensure that information on atmospheric chemistry change is available in forms that can be used by other disciplines. In particular, IGAC will include a focus on more accurate understanding of the linkages between atmospheric composition and climate.

Changes in atmospheric composition affect climate through changes in the abundance and distribution of radiatively active components thus altering the energy balance throughout the atmosphere. In the last decade, considerable progress has been made in determining the roles of different radiatively active components but new levels of complexity and new questions have emerged.

Climate change can no longer be thought of as driven by increases in CO₂ alone. Even though CO₂ may be the single most important greenhouse gas, the combined effect of other greenhouse gases and different types of aerosol particles, which have both cooling and warming effects, is at least as important. Validation of climate models using past observations, projections of future climate, and the policy tools and framework for

managing human influences on climate, all need to take into account the actual and potential evolution of a wide range of radiatively active components.

Our increased understanding of the linkages between atmospheric composition and radiative energy balance now challenges some of the simple approximations that have been used in the past to link atmospheric change to climate models and policy frameworks. The concept of radiative forcing has been used as a nexus between global mean greenhouse gas concentrations and the radiative perturbation used in climate models. However, radiative forcing is an approximation developed for long lived components that are well mixed throughout the atmosphere. Significant questions arise immediately when this concept is applied to short lived components which have large spatial variations. Similarly the effects of components that interact with radiation in very different ways, such as aerosol particles and greenhouse gases, may not be additive as often assumed in simple climate models. With the increasing spatial and temporal realism of climate models, and a growing emphasis on regional climate, it is necessary that the nexus between changes in atmospheric composition and perturbation of radiative energy balance be re-examined and broadened to capture new information on the role of different radiatively active components.

Atmospheric composition does not only affect climate through radiation balance and temperature. Precipitation can be affected directly through the influence of aerosol particles on cloud microphysical properties. Similarly the role of the biosphere in the climate system can be affected by nutrient transport through the atmosphere, physiological impacts due to pollutants, and the influence of cloud distribution and persistence on photosynthetically active radiation.

Changes in climate and other aspects of the Earth System also produce feedback effects on atmospheric chemistry. Some scenarios studied by the IPCC indicate that rising temperatures and water vapour concentrations in the atmosphere could alter CH₄ and O₃ concentration increases by up to 50% during this century. Natural emissions and deposition rates of a wide range of atmospheric components depend on surface temperature, wind speed, and precipitation. Lightning frequency is very sensitive to cloud top temperatures and is a significant source of NO_x, suggesting another potential direct linkage between changes in climate and atmospheric chemistry. An important challenge for the next decade will be to identify the key processes linking atmospheric change to climate change in a more comprehensive way than is done currently and to quantify these processes accurately in ways that improve the use of atmospheric science outside the atmospheric chemistry community.

Approach

We envisage that IGAC activities in this area will be characterised by one or more of the following three features:

- (1) They will involve a constructive synthesis of results from other IGAC activities addressing specific atmospheric constituents such as ozone or classes of chemical constituents such as aerosol particles.
- (2) They will involve linkages between the atmospheric chemistry community and other science communities including those involved in atmospheric radiation physics, satellite measurements, climate modelling, and the development of policy tools such as global warming potentials (GWPs).

(3) They will address aspects of the evolution of atmospheric composition over time.

Not all activities carried out within this sub-theme are expected to have all the above characteristics, but their net effect should be to produce a more comprehensive assessment of how atmospheric composition changes have affected and will affect climate at both regional and global levels in a form that is useful outside the atmospheric chemistry community.

The issues being raised here are all under consideration to a greater or lesser degree within existing science programmes and it is envisaged that the additional benefits from IGAC involvement will arise from fostering coordination and collaboration. In some cases coordination of activities dealing with issues addressed here may be conducted by other groups within the ESS-P. However, this sub-theme focuses on areas where underpinning atmospheric chemistry research is an essential component and direct involvement of the atmospheric chemistry community needs to be maintained. The role of IGAC will therefore include identification of appropriate linkages within ESS-P, identifying potential contributors from the atmospheric chemistry community, and assisting with the scientific leadership and coordination of collaborative activities. Critical indicators of the success of work in this sub-theme will include: more robust validation of climate models; more accurate climate projections for the future; and more comprehensive policy tools for managing emissions.

Opportunities and issues

IGAC activities should include a range of work conducted within the broad scope outlined in the background described above. Some activities may be specific to the atmospheric chemistry community, others may be established in collaboration with other disciplines, and some may be designed as gateway activities developing new linkages.

The following list of topic areas outlines possible opportunities and issues that would fit within this sub-theme.

- Development of methods for remote sensing of atmospheric composition, and validation of such methods, recognising the forward-model vs inverse-model relationships between inferring radiation fluxes and inferring atmospheric composition.
- Use of combined measurements of radiation balance and concentrations of a comprehensive range of radiatively active components to investigate the additivity of radiative perturbations caused by individual components.
- Development of methods for spatial and temporal extension of observations of short lived radiatively active components, such as tropospheric O₃, to enable such components to be incorporated in the validation of climate models.
- Better understanding of trends in atmospheric oxidation rates to enable more robust analysis of scenarios for future emissions using atmospheric chemistry models. For example, recent CH₄ growth rates are near zero and CO appears to be decreasing raising the question as to whether this is consistent with model based projections that have both components increasing at present.
- Better understanding of the indirect effect of emissions of methane and other gases such as CO etc on radiative properties of the atmosphere through consequent changes in product components.

- Investigation of the applicability of concepts such as the global warming potential for assessing the effects of short lived components such as CO and NO_x through analyses and observations of regional emissions, concentrations and radiative balance. Note that the policy framework developing in response to the UNFCCC embodies concepts such as GWP which do not currently treat ozone precursors and aerosol particles.
- Critical validations of atmospheric chemistry models used for projections of regional and global scale atmospheric chemistry. Note that models do not agree on the effect of future emissions and resolution of this problem will require further and carefully targeted collaboration between those involved in observation and those involved in modelling.
- Investigation of the effect of urban and biomass burning plumes (which are sub-grid scale in most climate and chemical tracer models) through combined measurements of emissions, concentrations and radiative balance.
- Studies of the potential role of megacities on regional climate through the net influence of their emissions on radiative properties of the atmosphere. Similar studies might be carried out for the net effect of an economic sector such as pastoral agriculture. Note that the IPCC Special Report on Aviation and the Atmosphere showed that the net effect of aviation emissions was quite different to that which would have been estimated based on GWP weighting of direct greenhouse gases.
- Studies to improve understanding of feedback processes between climate change and atmospheric chemistry (e.g. changes in lightning frequency and distribution) and to validate model derived estimates of such feedbacks.

Theme 2 Within the Earth System, what effects do changing regional emissions and depositions, long-range transport, and chemical transformations have on air quality and the chemical composition of the planetary boundary layer?

Research during the past few years has shown that even short-lived substances such as aerosol particles and ozone can be transported intercontinentally. This transport is easily seen in satellite images of dust transport. On 7-8 April 2001 strong winds swept across the Taklimakan Desert in western China, the Gobi Desert in eastern Mongolia, and the industrial regions of Eastern Asia lifting dust and pollutants into the troposphere. The plume was tracked by satellite cross the Pacific Ocean and can be seen here as a haze over the mid-Atlantic United States and Atlantic Ocean on 22 April. A major theme of IGAC will be studying the intercontinental transport and chemical transformation of short lived components and their effects on regional climate and air-quality thousands of kilometers downwind of their sources. Research activities addressing these issues have been divided into three questions concerning the transport and transformation, effects on air quality, and effects on the oxidizing capacity of the atmosphere.

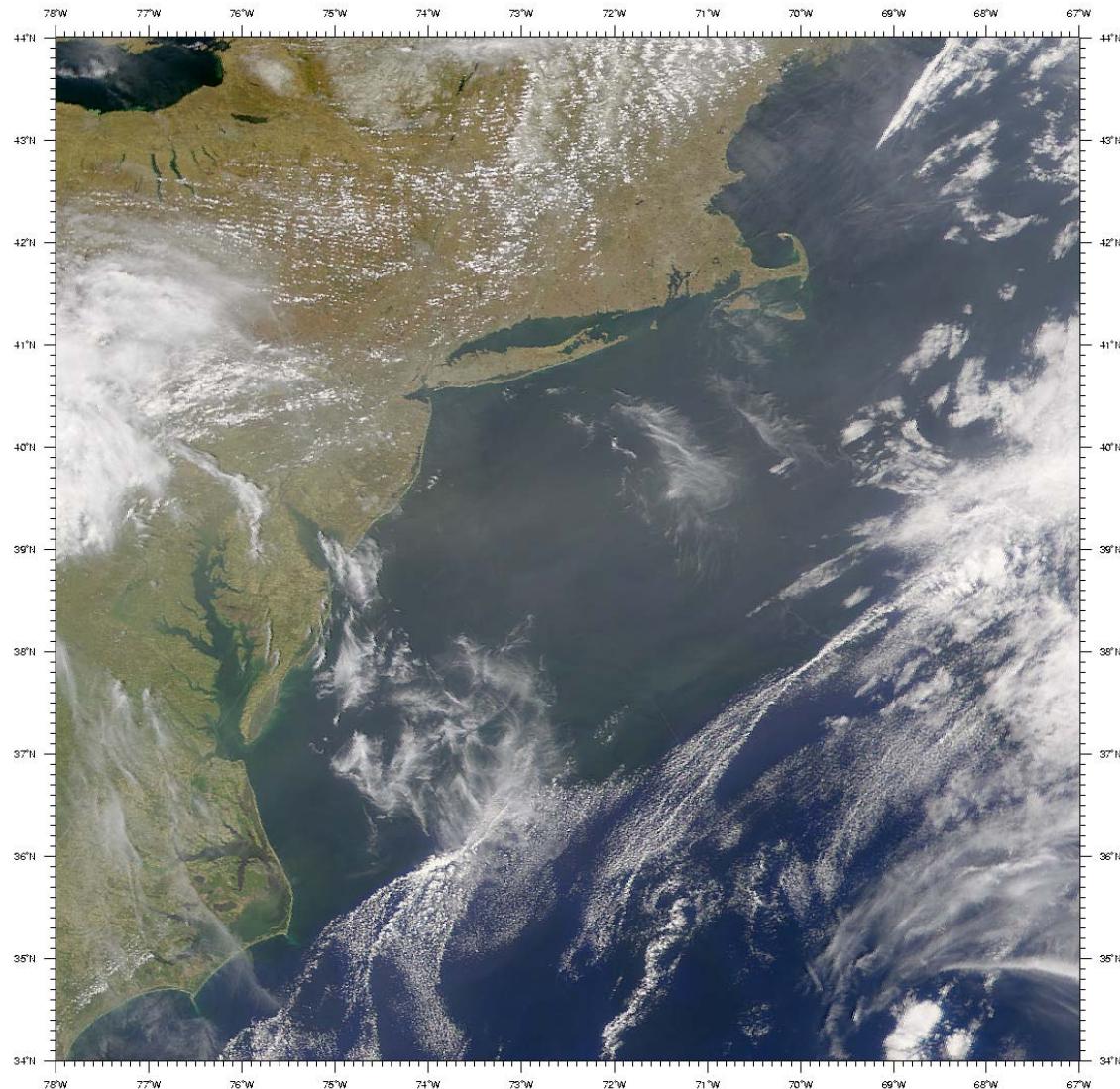


Figure 5. Transport of Asian dust two thirds of the way around the globe. This SeaWiFS image of the east coast of the United States shows the dust that left Asia two weeks earlier.

Question 2.1 What are the export fluxes of oxidants, aerosol particles, and their precursors from continents (e.g., mega-cities, biomass burning, desert dust) to the global atmosphere and how are these components transformed during transport?

Background

IGAC's research efforts during Phase 1 have contributed greatly to understanding and assessing the effects of anthropogenic emissions on the 'background' atmosphere and have elucidated some features of the transport processes that control the long-range transport of short-lived chemical compounds. ('Background' atmosphere is defined here as the present-day atmosphere in areas some distance away from the emission source

regions). Some of the recent scientific findings that form the basis for future investigations include:

- (1) *Ozone from North America dominates the ozone distribution over the North Atlantic during the summer.* - Surface measurements have shown that ozone pollution from North America is easily detectable 1500 km downwind from the North American source region in summer. This pollution has also been shown to enhance O₃ levels in the central North Atlantic, in the spring. These observations provide the basis for the quantitative assessment of the total flux of anthropogenic O₃ from North America to the North Atlantic in the summer. It totals on the order of 1.0 to 1.6 Gmol/day, which exceeds the natural O₃ flux from the stratosphere.
- (2) *The role that the oxides of nitrogen (NO_x) and volatile organic compounds (VOCs) play in the O₃ budget of the North Atlantic troposphere.* - In the remote, marine troposphere, the concentrations of carbon monoxide (CO) and methane are adequate to support significant photochemical ozone formation. Whether this photochemistry produces rather than destroys ozone is determined by the amount of NO_x available. Analysis of the correlation of NO_y with CO has demonstrated that only a small fraction of the NO_x emitted in the continental boundary layer is transported to the free troposphere or the marine boundary layer (MBL). Although transport is limited, model results suggest that eventual ozone production in the global troposphere from U.S. emissions is nevertheless about twice as large as the direct export of ozone from the U.S. boundary layer. The amount of NO_x delivered to the lower troposphere in the western North Atlantic during summer is sufficient to produce, on average, 1 to 4 ppbv/day of ozone throughout the region. In contrast, the central North Atlantic (e.g., the Azores) is a region of photochemical ozone destruction.
- (3) *Asian emissions influence the chemical composition over the northwest Pacific.* - Investigations were made of the long-range transport of atmospheric trace components over the northwest Pacific Ocean. The region of maximum outflow lay between about 20° N and 40° N. The delivery of NO_x from the Asian continent to the free troposphere and the subsequent redistribution by large-scale dynamics influences the ozone production over large areas of the Pacific basin.
- (4) *Oxidized sulfur over the western Pacific is attributed to Asian sources.* - Measurements were made of the chemical composition of the atmosphere over the western Pacific. These measurements indicated that in this region throughout the well-mixed troposphere the sulfur dioxide and sulfate in aerosol particles were largely associated with emissions of sulfur dioxide from sources located on the Asian continent.
- (5) *Mechanism for the transport of North America pollution to the North Atlantic.* - Meteorological measurements show that during the summer, inversions can effectively isolate the MBL from the lower free troposphere aloft, where the majority of the pollutants are transported in highly stratified layers. The processes that form these layers provide an effective mechanism for the transport of continental pollution into the mid- and upper-troposphere over the North Atlantic.
- (6) *The role that fronts play in transporting continental pollution.* - The primary direction for transport of North American pollution to the North Atlantic in the summertime is toward the northeast. Warm sector flow ahead of advancing cold

fronts has been identified as the most important process for the transport of pollution from the urbanized U.S. East Coast to the North Atlantic. This mechanism provides a means to rapidly and effectively transport large amounts of relatively short-lived pollution over long distances.

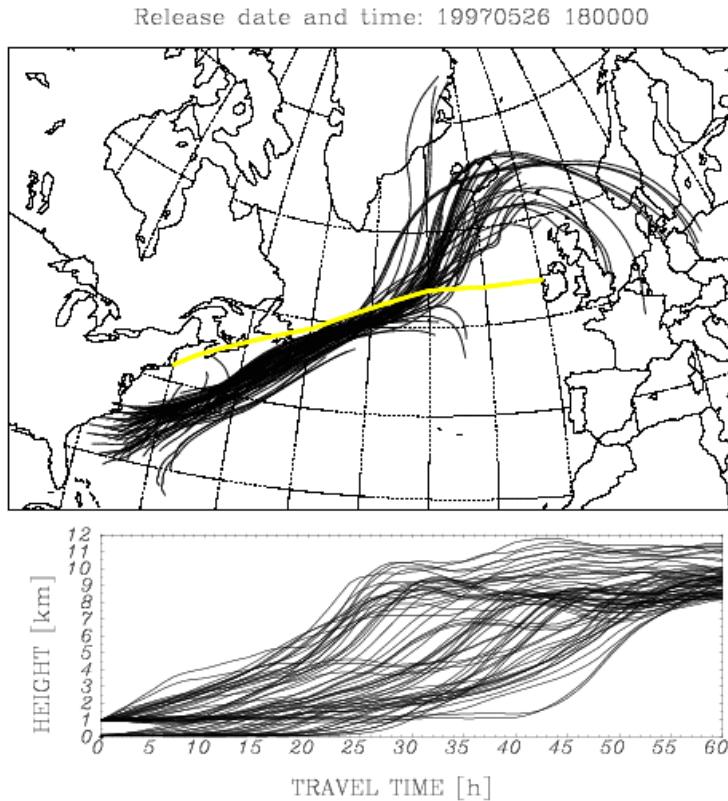


Figure 6. Three-dimensional trajectories with strong ascent that visualize the “warm conveyor belt” (WCB) flow. The upper part of the figure shows a horizontal projection, the lower part shows a time-height profile of the trajectories. The black line marks the leg of the MOZAIC aircraft that flew through the WCB and measured about 100 ppb O₃.

(7) *Role of aircraft emissions of NO_x relative to lightning and surface emissions in ozone formation.* - Recent studies have examined the sources of the oxides of nitrogen in the remote free troposphere over the North Atlantic. They indicate that emissions from aircraft and lightning in the free troposphere and transport from the surface all play a significant role in determining the NO_x distribution and the consequent formation of ozone in the free troposphere. These sources are found to vary with latitude: aircraft emission being relative more important at high latitudes, lightning at low latitudes.

(8) *Intercontinental transport of emissions from forest fires.* - The principal sources of CO in the Northern Hemisphere are fossil fuel combustion, biomass burning, and oxidation of methane and non-methane hydrocarbons. Recent research has elucidated the importance of boreal forest fires to northern hemispheric summertime CO background concentrations. Hemispheric background concentrations of CO declined from late 1980's through the middle of the 1990's. This decline was associated with emission control of CO in the industrial countries of the Northern Hemisphere. This trend has been perturbed by large variations in CO emissions associated with biomass combustion due to wild fires in the boreal regions of North America and Russia. The emission of the NO_x, combustion-produced VOCs, and carbon aerosol particles may also perturb the atmospheric chemistry in the Northern Hemisphere. The variability in these forest fire emissions parallels climate variability that has produced drought conditions in the boreal regions.

(9) *The seasonal variation of the anthropogenic influence on the tropospheric O₃ budget.* - All of the summertime studies in the western North Atlantic have found a positive correlation between CO and O₃, demonstrating that anthropogenic pollution produces ozone in the summer. However, in the wintertime a negative correlation between CO and O₃ is observed, both at surface sites and in the free troposphere. This negative correlation demonstrates that anthropogenic pollution provides a sink for ozone in the winter. This destruction is consistent with the expected reaction of O₃ with primary pollutant emissions of NO and unsaturated VOCs. This destruction is expected to occur in all seasons. However, photochemical formation of O₃ more than compensates for the destruction in summer. It has not been determined if the overall anthropogenic effect on O₃ in the winter is negative further from the continent. In winter, slow photochemical O₃ production likely occurs over longer transport times and distances in the troposphere. Long-range transport of anthropogenic precursors to low latitudes with more photochemical activity could possibly further enhance O₃ formation in the remote marine troposphere. A combination of these processes may compensate for the initial O₃ destruction.

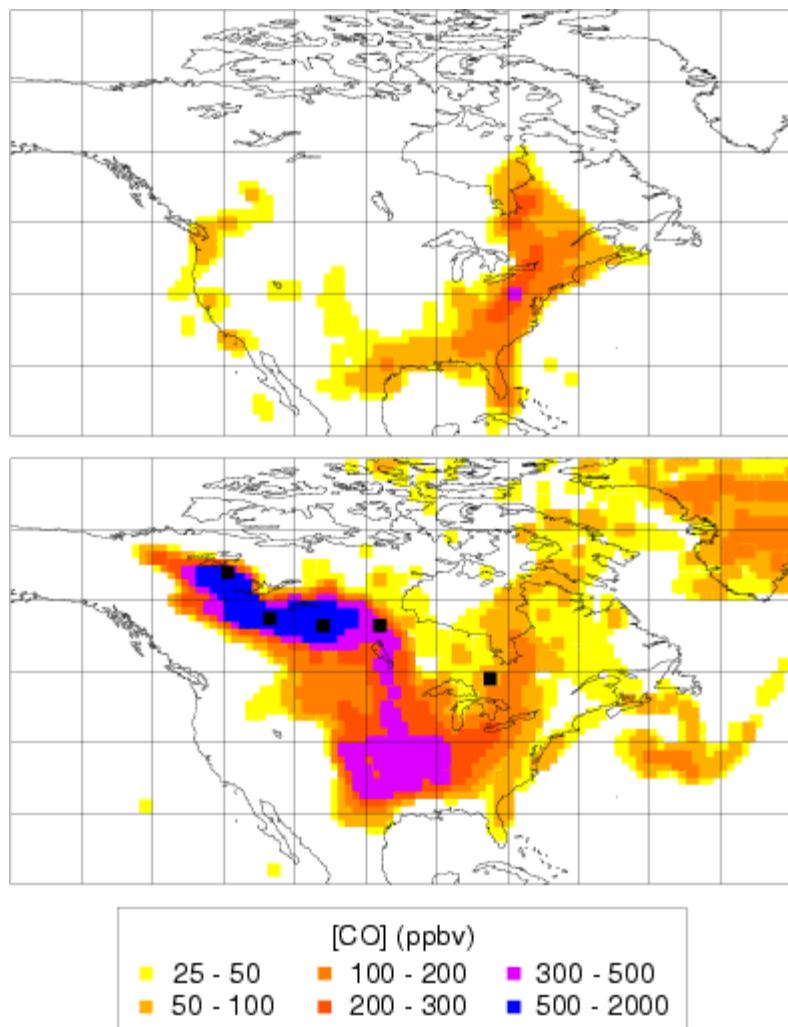


Figure 7. Calculated boundary layer CO concentrations from anthropogenic emissions (top) and from forest fires (bottom) for 1 July 1995. Forest fire locations are marked with black squares. The anthropogenic concentrations are high ahead of a cold front that parallels the east coast, while forest fire concentrations are high behind the cold front. (from Wotawa and Trainer, 2000).

The Bottom Line. - Our understanding of atmospheric chemistry and transport has evolved to a point where we now recognize that intercontinental transport of anthropogenic emissions (and of their chemical products) has significant effects not only on ‘background’ atmospheric chemical composition but on regional near-surface chemical composition and deposition fluxes as well. In the mid-latitude Northern Hemisphere, emissions from fossil-fuel combustion in eastern North America, western Europe, and eastern Asia, affect air quality in regions downwind of these major source regions. It is expected that the tropics will become increasingly important in terms of their impact on global atmospheric chemistry. Understanding the mechanisms of pollutant export from northern mid-latitude and tropical source regions, quantifying the

export fluxes from these regions, fully understanding their transformations, and assessing the impacts of these fluxes will be a central component of IGAC research.

Approach

Research tools

3-dimensional models will provide the quantitative answers to these research questions. They must resolve the coupling between transport, chemistry, and aerosol processes on synoptic scales and finer. Such models presently exist but their adequacy is highly uncertain. They need to be constrained and tested with atmospheric observations to a much more deliberate degree than has been achieved so far. One aspect of the experimental design for IGAC therefore involves a close coupling of atmospheric observations and models, in which the models are driven by assimilated meteorological observations for the period of interest, and the observations are targeted towards testing the key relevant features of the models. As discussed below, observations from ground-based, aircraft, and satellite platforms all have important roles to play in this design, both in testing model features, and in providing a wider base of exploratory measurements. Ship platforms are not specifically discussed but share some of the advantages and disadvantages of ground-based and aircraft platforms.

Ground-based platforms have two primary strengths. First, they provide the opportunity for field programs that simultaneously measure a broad spectrum of gaseous and aerosol constituents by research grade, prototype instrumentation. Such field studies expand the observational database that allows the identification and investigation of crucial atmospheric processes. During these studies many components can be measured by two or more techniques to allow for intercomparison of results. Such intercomparison efforts are critical for the development and improvement of measurement techniques. Second, ground-based platforms allow long-term observations at relatively low cost. Measurements can be made for a large number of components under well-calibrated conditions. A disadvantage of ground-based measurements is the lack of spatial information, which is critical for interpreting the long-range transport and chemical evolution of air masses. Lidars and sondes give vertical information but only for a few components (ozone, aerosol particles, water vapor) at present. The principal roles of ground-based measurements in this context are 1) to expand the observational data base of simultaneous measurements of a broad spectrum of atmospheric components, 2) to provide high-quality data on seasonal and interannual temporal trends, 3) to identify correlations between components that may provide important constraints for the models, and 4) to extend in time the information gained from intensive field studies.

Aircraft measurements have the advantage of providing large spatial coverage. High-quality measurements can be made from aircraft for a number of components. By designing the flight plans in the context of chemical model forecasts, the observations can be collected in a manner that provides an optimal test of the models. The disadvantages of aircraft observations are limitation in flight hours, so that the data are only snapshots in time, and the limits on instrumentation dictated by aircraft payload considerations. This time limitation can be overcome to some extent with regular observations from commercial aircraft or from chartered small aircraft, but the number of measured components is then even more limited and there is less flexibility in the observational strategy.

Satellite measurements can potentially provide global and continuous observations of tropospheric composition and are in that regard ideally suited for characterizing intercontinental transport. However, for the foreseeable future only a few components will be observable from space, the vertical resolution will be coarse (at most a few pieces of information in the troposphere), and the precision will remain limited. All current and planned satellites are in polar orbit and have a return time of 3 days or more for any given spot on the Earth; thus the data are not truly continuous. Measurements from geostationary orbit would provide truly continuous observations on the scales of relevance to ITCT but these measurements are still in a technology demonstration phase.

Research agenda

We envision a three-pronged research agenda involving (1) intensive field studies aimed at investigation of specific processes, (2) long-term observations to place these processes in a seasonal and interannual context, and (3) assessments directed at policy development.

Intensive field studies harness measurements from a number of platforms, together with supporting 3-D chemical transport models, into a concerted experimental design focused on answering a limited set of well-defined questions. To address the questions raised here, these field studies must involve one or more research aircraft. One useful tool to optimize the value of the measurements for testing the models will be to operate the models in forecast mode over the course of the intensive field study. These forecasts will then be available to guide flight planning on a day-to-day basis. Integration of satellite observations into the experimental design may also be helpful to place the limited aircraft observations into a broader spatial context. Developing this synergy between aircraft and satellite observations may call for inclusion of satellite validation flights in the aircraft mission plans.

Long-term observations are essential to address the ITCT questions by extending temporally the information from the intensive field studies into a seasonal and multi-year frameworks. Long-term measurement platforms may include ground-based sites, ships, commercial aircraft, and small, chartered aircraft. Satellites have been deployed to produce valuable long-term observations, but have the important limitations discussed above. The selection of platforms should be made with careful consideration of 3-D model results in order to provide the best test of the models towards addressing the science questions. Eventually, the long-term measurement program may evolve into a monitoring operation to document changes in anthropogenic emissions from continental source regions and their global implications for atmospheric chemistry.

Assessment. Periodic assessment must be made of the implication of the research findings in terms of the effects of projected changes in emissions and other forcing variables (e.g., changes in land use or climate) for intercontinental transport of pollutants. In this activity IGAC should foster a closer interaction between the policy communities of the large industrial countries. We expect that the policy community will provide guidance concerning the assessments of highest priority. These assessments will direct the scientific resources of the program towards the most critical uncertainties to be addressed.

Question 2.2 What are the impacts of intercontinental transport on surface air quality?

Background

The regional pollution theme of IGAC will focus on atmospheric chemistry studies that will have a direct bearing on human welfare and environmental quality. While a detailed assessment of the human health and environmental impacts of regional air pollution will be outside the purview of IGAC, the relationship between large-scale distribution and transport of pollutants and appropriate human health and environmental impact pollution metrics will be an important IGAC research question. Thus, the new regional air quality focus of IGAC will build on the understanding gained from the IGAC-I focus on the 'background' atmosphere, with the expectation that the results of these research activities will provide the scientific underpinnings for formulating effective regional and national air quality management policies.

Already ample evidence exists establishing the importance of the impact of long-range transport of ozone, fine particles, and their precursors. The long-range transport of dust particles from Asia and Africa has been extensively documented. Estimations also indicate that there is an increasing trend in the mixing ratio of ozone over the Northern Hemisphere that is probably associated with anthropogenic influences on the chemical composition of the atmosphere.

More specifically, several reports have appeared in the published literature documenting evidence for intercontinental transport of ozone, ozone precursors, and other chemical compounds. Measurements made from island locations in the Atlantic and Pacific have recorded events of increased concentrations of ozone and CO. Measurements made during the spring on the West Coast of the United States have detected Asian influence on the levels of several atmospheric components including O₃, CO, peroxyacetyl nitrate (PAN), nitric acid (HNO₃) and light hydrocarbons during periods when trajectory analysis indicated rapid transport from Asia. Model simulations reproduced the observed enhancements in CO and PAN, and indicated that the ozone over the western United States was also enhanced. Other model calculations indicate that significant enhancements in the average ozone concentration over the Western United States will occur if Asian emissions of ozone precursors increase significantly.

Elevated concentrations of ozone that are attributed to long-range transport from the boundary layer over North America were observed over Europe during an event that occurred in the spring of 1997. Ozone mixing ratios as high as 100 ppbv were measured in the free troposphere. The high concentrations recorded during this episode were explained by the effective transport without significant dilution of boundary layer air that was carried by an ascending air stream at the leading edge of a trough.

Numerous examples of long-range transport have been investigated, but a systematic and quantitative understanding of the budgets of the important trace components on a hemispheric to global scale is missing. This is particularly true for the O₃ budget; there is as yet no agreement on the relative importance of natural and anthropogenic sources of ozone to the Northern Hemisphere troposphere. One of the major tasks facing IGAC is to provide this quantitative analysis.

While tropospheric ozone and aerosol particles will continue to be of central importance in IGAC, other environmentally important components will also be a focus of research. For example, mercury is a priority pollutant in many international conventions (e.g. UN ECE CLRTAP) and there is a growing awareness that a significant fraction of the deposition flux of mercury in a particular region can originate from outside the region. Formulating effective mercury (and other priority pollutants) deposition control policies will therefore require the development of source-receptor relationships for mercury on hemispherical and global scales. This in turn will require a better understanding of the speciated distribution of mercury compounds in the global atmosphere, and of the underlying processes that control these distributions.

Approach

A new activity that has emerged during IGAC is global "chemical weather" forecasting. The "chemical weather" is the short-term state of the atmosphere in terms of the time-dependent distributions of trace gases and aerosol particles. Using chemistry-transport models driven by meteorological forecasts from weather centers such as NCEP and ECMWF, predictions of the global chemical weather have been computed and used extensively in the day-to-day flight planning in several major field campaigns (e.g., INDOEX, TRACE-P, ACE-Asia, ITCT-2K2). Global chemical weather forecasting is expected to become one of the major foci of atmospheric chemistry activities in the next decade, since it integrates many of the efforts discussed here (improved emissions inventories, long-range transport, chemical transformations, deposition, and assimilation of observations, especially satellite-based). The applications of chemical weather forecasts will also expand beyond campaign planning into health and agricultural issues. The atmospheric chemistry community may profit from being able to work in a more "operational" mode like the meteorological community, working towards measurable improvements in community-wide products. A careful coupling of global and regional-scale forecasts will be needed. Although there is already considerable experience with ozone forecasts on an urban scale, the strong global interconnections that have been pointed out during IGAC also point towards the need for embedding regional models in the framework of global chemical weather forecasts.

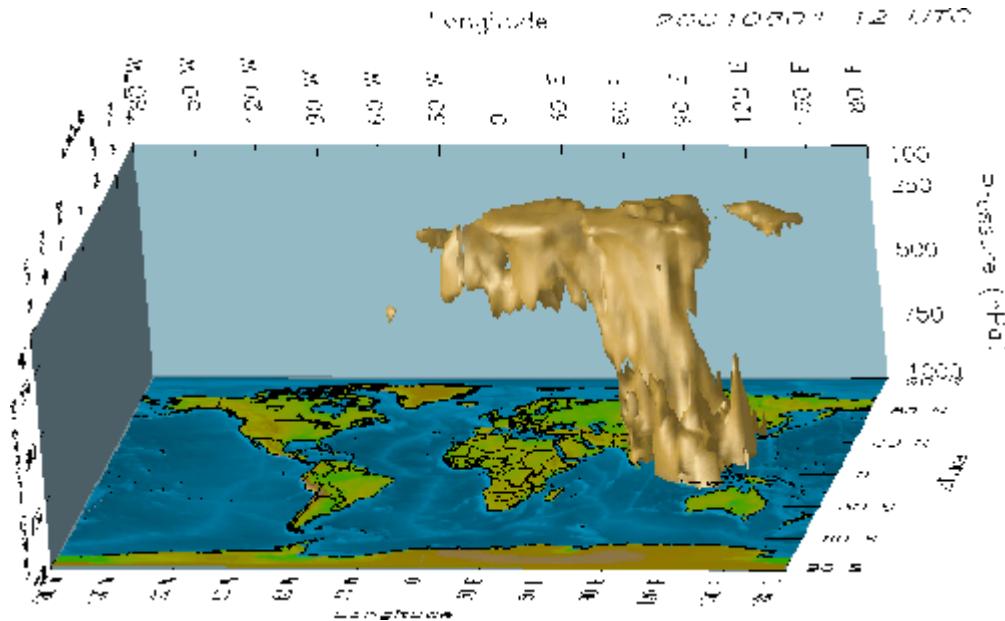


Figure 6. 3D image of the southern Asian monsoon plume computed with the global MATCH-MPIC chemical transport model. The plume shows one of the major pathways for export and intercontinental transport of the emissions from Asia. This major intercontinental transport feature was successfully predicted using global chemical weather forecasts for the MINOS field campaign planning.

Understanding Processes

The coupling of emissions generated in one region to ambient pollution levels in another is controlled by a number of key atmospheric processes (mixing, transport, chemical transformation, deposition, etc.). The prediction of ambient pollution levels relies on an understanding of how these processes influence the formation and distribution regardless of whether the approach to forecasting involves a parametric or deterministic approach. Therefore, reliable forecasts of chemical composition in a particular region require an improved understanding of the entire ensemble of salient, intervening atmospheric processes along with reliable inventories of pertinent emissions.

Model Evaluation

The models required for global "chemical weather" forecasting are closely related to those required to answer the previous science question. The evaluation process outlined in that section will be required here as well. This evaluation of model performance is required to quantify forecast reliability and to identify areas needing improvement. There is a need to evaluate model performance in all key areas, including: 1) emissions, 2) meteorology and transport, 3).chemical transformation, and 4) deposition.

Observations

Observations play a key role in model evaluation. A well-designed observing system is essential to the success of any air quality forecasting system. The need for information on the chemical constituents of the atmosphere is analogous to the meteorological observing system that supports the current weather forecasting system. Information on

atmospheric pollution is needed to both initialize and evaluate the forecast. Specific research needs include: 1) data to initialize models and perform operational evaluation; 2) data for diagnostic evaluation of complex grid models; 3) Information on the vertical distribution of pollution; and 4) access to data in a timely manner.

Question 2.3 How will human activities transform the cleansing capacity of the future atmosphere?

Background

The oxidizing or cleansing capacity of the atmosphere is generally defined by the level of the hydroxyl radical, OH. It is crucial because it determines the rate at which atmospheric pollutants, greenhouse gases and short-lived replacement HCFC compounds are removed from the atmosphere. To a large extent the oxidizing capacity is controlled by O₃, water vapor, UV radiation and levels of trace gases like CH₄, CO and VOCs. Also, due to its extremely high reactivity and rapid cycling with other HO_x components (HO₂, H), the OH concentrations are very low (~ 10⁶ molecules cm⁻³) and highly variable. Only in the last decade has it been possible to measure OH directly with some degree of accuracy. Even so, it still remains impossible to measure OH on a global scale in order to determine trends directly. Therefore, global chemistry transport models and calculations based on the use of network observations of gases like CH₃CCl₃ and HCFC-22 have been used to derive the change in OH levels over the past 20 years or so, or in the case of models, since pre-industrial times. In general, global models have estimated ~10% decrease in global OH over this period with results being sensitive to assumed changes in emissions of NO_x, CO, CH₄ and hydrocarbons. This is in contrast with recent estimates based on the analysis of CH₃CCl₃ data using inverse modeling techniques suggesting increases in global OH of similar magnitude. One difference was that stratospheric O₃ depletion was not taken into account in most modeling studies. This should result in an increase in tropospheric UV levels and hence in global mean OH. This mechanism has also been forward to explain, at least partially, observed changes in the CH₄ and CO growth rates over the last 20 years. Therefore, it still remains very uncertain how OH has changed in the past and how it will change in the future in response to changing anthropogenic emissions and climate change.

Approach

Three approaches are required to determine whether or not the earth's oxidizing capacity is changing: chemical models based on better emissions inventories, evaluated with global data; observations, including in-situ and satellite measurements of key components in today's atmosphere, along with data from ice cores and snow to interpret the past; coupled chemistry-climate models for interpretation of past and present and for making projections of chemical forcing and climate response and vice-versa.

Models, Inputs, Model Evaluations

It is clear that, without direct measurements of fully global distributions of OH or even of CH₃CCl₃, any methods for estimating changes in the oxidizing capacity have their limitations and that improved estimates will only come about by reducing

uncertainties, particularly in global models. This requires improvements in emission inventories of O₃ precursors etc. which have very large uncertainties (more than a factor of 2), particularly related to their temporal variability (via continuation of GEIA activity) and better understanding about chemistry-climate interactions (e.g. how do lightning NO_x emissions respond to changes in convection?). It also suggests the need for continued evaluation of model performance (chemistry, transport and deposition) through inter-comparison and by comparison with observations (aircraft, sondes, satellites). This could take place via continued activity within IGAC (GIM) and in conjunction with activities organized by SPARC (e.g. strat/trop. exchange), WCRP (transport and deposition processes) and IPCC (emissions, O₃ budget).

Observations

Observations also play a key role in improving our understanding about the oxidizing capacity. This includes, in particular, the need to continue long-term monitoring of trace gases such as halocarbons, CO, CH₄, O₃ combined with setting up new monitoring stations in regions which are very data sparse and/or which make measurements of key trace gases about which trend information is extremely uncertain (e.g. NO_x, VOCs). One pressing need, which will be addressed, to a certain extent by new satellite data, is the need for long-term measurements in the free troposphere. Commercial aircraft (e.g. MOZAIC O₃ 1994 to 2002) offer one possibility but the development of new instrumentation to fly regularly on sondes should be encouraged. It is also clear that dedicated research campaigns are also necessary. They need to be focused on areas where our current understanding is poor and which are sensitive to climate change (e.g. West Africa, Eurasia). In this case, measurements of a wide variety of chemical components, including HO_x, have already been shown to provide stringent tests of our current understanding of tropospheric photochemistry (e.g. PEM campaigns). Tracer measurements also allow representations of transport schemes in models to be evaluated and improved. In evaluating chemistry-transport and chemistry-climate models and possible oxidizing changes, historical data of chemical constituents provide useful constraints. Methane in ice-cores is one example of data that have proven useful in published studies. Further extraction and measurements of gases related to ozone, OH and other oxidants, through IGAC and/or IGAC-linked projects within IGBP are envisioned.

New Model Initiatives

Use of new modeling techniques (e.g. data assimilation, inverse modeling), combined with new datasets will open up the possibility of gaining a better insight into how the oxidizing capacity might be changing in the past, present and future. As well as better quantification of how anthropogenic emissions are changing in different regions (IPCC), it will also be necessary to use Earth system models that include links between the chemical and climate system. In this way, it will be possible to study links between climate change (human-induced) and changes in the natural system (emissions, dynamics, water vapor cycle). Therefore, over the period of IGAC it is envisaged that the development of chemistry-climate models and their use to understand the Earth system will be an important goal leading to better understanding about the oxidizing capacity and how it might be changing.

LINKAGES WITH OTHER PROGRAMS

Emissions inventory. Major uncertainties exist in present inventories. In addition, methods of updating inventories need to be greatly streamlined to keep them up to date. Global-scale assessment and the incorporation of inverse modeling techniques will both depend upon and contribute to advancing knowledge of emissions. The role of IGAC will be to identify important gaps in inventories and encourage evaluation by models and measurements. To expand and improve the IGAC Global Emission Inventory Activity (GEIA), the new IGAC will need to work closely with the IGBP Projects studying the ocean-atmosphere (SOLAS) and land-atmosphere interfaces (ILEAPS).

Megacity analysis: Megacity plumes can provide excellent calibration targets for satellite sensors. High concentrations increase spatial resolution of the remotely sensed quantities, thereby improving the ability to couple them to ground-based and airborne in-situ measurements. Chemical characterization in these important source regions will assist long-range transport and transformation studies. The role of IGAC will be to foster the acquisition of high quality, consistent, and scientifically useful data across diverse regions.

Air quality. Vast amounts of aerosol particle data are routinely gathered around the world for the purpose of monitoring air quality and designing control strategies to minimize effects on human health. IGAC will provide a forum for linking these programs into an effort to assess regional and global-scale effects. This is also an area where IGAC can join with the IHDP.

Paleoclimate interpretation. Paleoclimatic records indicate wide variations in natural aerosol particle loadings and trace gas concentrations. For example, both Greenland and Antarctic ice-cores indicate that dust loadings during the last glacial maximum were many times higher than at present. However, to estimate the climatic impacts of these changes, it is necessary to know the radiative effect of present levels of these same natural aerosol particles. Attribution of present AOD to natural and anthropogenic aerosol components, as envisioned herein, will lay the groundwork for such paleoclimatic analysis.

Troposphere-stratosphere exchange. To fully understand the processes controlling the distribution of chemical species in the atmosphere we must integrate the measurements and expertise of the two communities. IGAC and the WCRP SPARC program must develop joint research strategies for these common research questions.

Aerosols and the hydrological cycle. The concentration, size and composition of aerosol particles that can act as cloud condensation nuclei determine cloud properties, evolution and the development of precipitation. This is an area where IGAC and the WCRP GEWEX program have many overlapping interests. Both communities have unique expertise to address questions involving the hydrological cycle. This is an area where IGAC can also contribute to the ESSP water project.

IMPLEMENTATION STRATEGY

Underlying programme principles:

Human activities have profoundly perturbed the chemical composition of the atmosphere over the past two centuries. Although atmospheric chemistry and issues of air quality were initially regarded as local or regional problems, it is now clear that the atmosphere transforms and transports a wide range of chemical components intercontinentally and in many cases globally. Atmospheric chemistry is thus a global issue that must be addressed in an international context. IGAC provides a framework in which to develop and study scientific questions concerning the chemistry of the atmosphere that require a global strategy. The following principles will form the basis of a new atmospheric programme.

- IGAC's goal is to promote and facilitate international atmospheric chemistry research that will lead to a better understanding of the Earth System.
- The programme will be driven by scientific questions that require a global strategy.
- Within this global strategy, activities will be targeted in regions of high impact on the global system.
- The programme will position atmospheric chemistry within the Earth System framework to assess responses and feedbacks within the system.
- The programme will operate across traditional organizational boundaries at regional to global scales, integrating traditional troposphere (IGAC), stratosphere (SPARC), and measurement (GAW, IGOS) programmes.
- The programme will develop strategies in concert with ocean (SOLAS, JGOFS, GLOBEC) and terrestrial (BAHC, LUCC, GCTE) scientists to quantify the exchange of chemical components, between the atmosphere/ocean/land/biosphere and to build a common interactive emission database.
- Research activities within the programme will strive to be inclusive and promote international cooperation.

Mode of operation:

IGAC's goal is to promote and facilitate international atmospheric chemistry research that will lead to a better understanding of the Earth System. Since IGAC does not fund research per se, research efforts within IGAC must come from pro-active scientists. Thus, where appropriate, research will be organized on a task basis directed towards answering specific scientific questions defined by the scientists involved. Tasks will be defined with a finite lifetime, initially approved and periodically reviewed by the IGAC Science Steering Committee, and will have as a final goal a publication(s) in a peer-reviewed journal(s). Certain tasks, such as the development and maintenance of a global database for surface emissions of chemical compounds, will continue on an on-going basis with period reviews and published summaries.

IGAC will promote interaction among the atmospheric chemistry community through biannual scientific symposia. The symposia will be held throughout the world to facilitate a broad geographical representation.

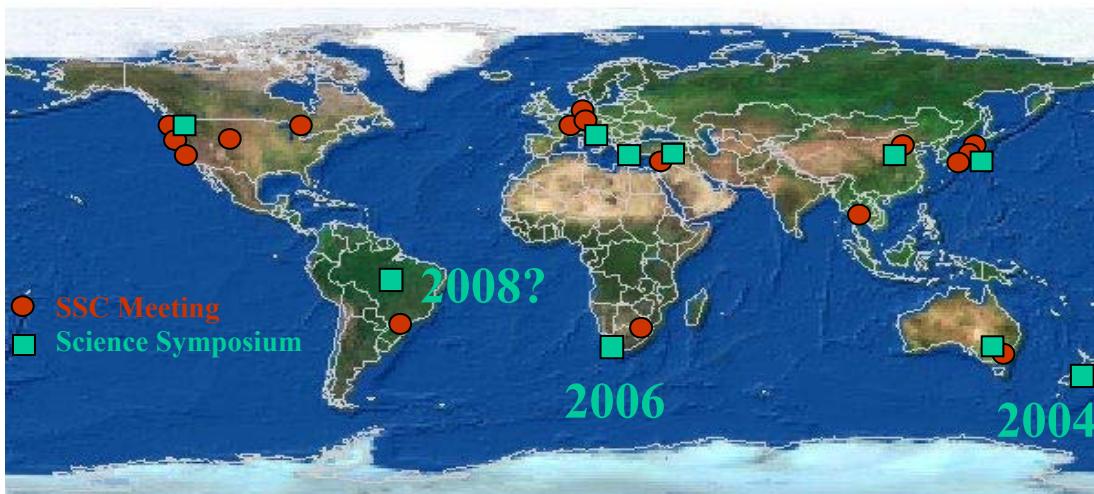


Figure 7. IGAC SSC meetings have been held on six continents. Upcoming Science Symposia are planned for New Zealand, South Africa (in coordination with the CACGP quadrennial conference), and South America.

Organizational Structure

IGAC research activities and Scientific Conferences will be approved and reviewed by the Scientific Steering Committee (SSC). Each SSC member will also serve as a liaison with one other Activity with closely related research interests (e.g. SOLAS, ILEAPS, SPARC, PAGES, GAIM, IGACO/IGOS, IPCC). Funding permitting, each SSC member will attend an annual IGAC SSC meeting and their liaison activity meeting.

IGAC operations will be coordinated through one or more International Project Offices. Initially the project offices will be housed in Taiwan, the US, and Europe. The distribution of activities between these offices will depend on funding resources in each country/continent. In general, the International Project Offices (IPO) will:

- promote and facilitate international atmospheric chemistry research that will lead to a better understanding of the Earth System;
- assist the IGAC SSC in planning and carrying out new scientific research;
- serve as a channel of communication between scientists working in different countries on various aspects of global change and between IGAC and the scientific community as a whole; and

- assist the SSC in collating information on national and regional programmes of global change research relating to IGAC.

Implementation of Tasks

Guidelines for proposal submission:

IGAC's research activities under phase II will be organized under Tasks. The Tasks, proposed, organized and run by proactive scientists will be initially approved and annually reviewed by the IGAC SSC. IGAC Tasks must include:

- a research plan which focuses on specific scientific question(s), as described in the IGAC Science Plan, that require an international research framework;
- where appropriate, a section dealing with the natural variability of the processes under investigation and any attempts to better define this variability;
- a timetable of research activities with a completion date. Research activities should strive to be inclusive to promote wide participation and international cooperation;
- a quality assurance and data plan that includes public access to the data;
- a plan for educational and capacity building efforts;
- plans for peer-reviewed manuscript(s), including possible titles, special issues and targeted journals, as the final products; and
- the name(s) and full contact address(es) of the task(s) coordinator(s). The IGAC SSC co-chairs will assign an IGAC SSC member as the main point of contact for the task.

Proposals for Tasks should be no longer than 8 pages and should be submitted to the US IGAC project officer. Proposals can be submitted at any time and will be circulated by email to the SSC for approval. Tasks will be reviewed annually at the SSC meetings. Annual task summaries (see below) should be submitted six weeks prior to the annual SSC meeting.

Although IGAC can not provide direct research funding for its Tasks, the IGAC SSC will:

- Serve as an international advisory board;
- Provide project advocacy and promotion, enlisting wide international participation in the project;
- Serve as a channel of communication between scientists working in different countries on similar projects; Encourage co-ordination with other components of the IGBP, and other relevant international research programmes; and Advocate for national and international funding of its Tasks.

Guidelines for the annual report

Each task will be asked to submit an annual report, of no more than 3 pages, addressing:

- the scientific highlights of the last year;
- the deliverables which have been met in line with the approved proposal; and
- any changes to the original proposal.

Copies of published manuscripts, training manuals or any products should accompany the annual report. These reports will be due six weeks before each SSC meeting.

REFERENCES

G. Wotawa and M. Trainer, *Science*, 288, 324-328, 2000.

Brasseur et al., 2002

IPCC 2001

DEFINITIONS OF ACRONYMS

- ACE: Aerosol Characterization Experiment
- AERONET:
- APARE: East Asia/North Pacific Regional Experiment
- BAHC: Biospheric Aspects of the Hydrological Cycle
- BIBLE: Biomass Burning and Lightening Experiment
- CACGP: Commission on Atmospheric Chemistry and Global Pollution
- CARIBIC:
- ESSP: Earth System Science Partnership
- EXPRESSO: EXPeriment for Regional Sources and Sinks of Oxidants
- GAIM: Global Analysis, Integration, and Modeling
- GAW: Global Atmospheric Watch
- GEIA: Global Emissions Inventory Activity
- GCTE: Global Change and Terrestrial Ecosystems
- GLOBEC: Global Ocean Ecosystem Dynamics
- IAMAS: International Association of Meteorology and Atmospheric Sciences
- IGAC: International Global Atmospheric Chemistry Project
- IGACO: Integrated Global Atmospheric Chemistry Observation
- IGBP: International Geosphere Biosphere Program
- IGOS: Integrated Global Observing Strategy
- IHDP: International Human Dimensions Program
- ILEAPS: Land-Atmosphere Project
- INDOEX: Indian Ocean Experiment
- IPCC: Intergovernmental Panel on Climate Change
- JGOFS: Joint Global Ocean Flux Study
- LUCC Land-Use and Land-Cover Change
- MINOS:
- MILOPEX: Mauna Loa Observatory Photochemical Experiment
- MOZAIC
- NARE: North Atlantic Regional Experiment
- PAGES: Past Global Changes
- PEM:
- SAFARI: Southern African Fire-Atmosphere Research Initiative
- SHADOZ:
- SOLAS: Surface Ocean – Lower Atmosphere Study
- SPARC: Stratospheric Processes and their Role in Climate
- STARE:

- TARFOX: Tropospheric Aerosol Radiative Forcing Observational Experiment
- TRACE-P: Transport and Chemical Evolution over the Pacific
- TRAGNET: United States Trace Gas Network
- UNFCCC: United Nations Framework Convention on Climate Change
- WCRP: World Climate Research Program
- WMO: World Meteorological Organization
- WOUDC:

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