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Science Plan and Implementation Strategy

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Prepared by the IGAC Transition Team:

Tad Anderson, United States
Timothy Bates, United States
Mike Bergin, United States
Olivier Boucher, France
John Burrows, Germany
Cristina Facchini, Italy
Fred Fehsenfeld, United States
Sandro Fuzzi, Italy
Laura Gallardo-Klenner, Chile
Didier Hauglustaine, France
Daniel Jacob, United States
Prasad Kasibhatla, United States
Katharine Law, United Kingdom
Mark Lawrence, Germany
Caroline Leck, Sweden
Shaw Liu, China (Taipei)
Ulrike Lohmann, Switzerland
Danny McKenna, United States
John Munthe, Sweden
Kevin Noone, Sweden
Ulrich Platt, Germany
David Parrish, United States
Lynn Russell, United States
Mary Scholes, South Africa
Ann Thompson, United States
Chandra Venkataraman, India

Preface

The IGAC Science Plan and Implementation Strategy lays out the scientific objectives and key research issues of the atmospheric chemistry project of the International Geosphere Biosphere Programme (IGBP) as both IGAC and IGBP enter their second phase. It also lays out a framework for addressing these objectives and issues, recognizing the need for collaboration with partner programmes and projects.

The scientific focus of this document emerged from the first decade of IGAC research, much of which was conducted in the context of focused, intensive measurement campaigns. The scope of IGAC in its next phase includes both regional characterisation and the extension into issues that cross more expansive boundaries in space, time and discipline. While local and regional-scale atmospheric chemical composition will be a primary focus, it is now clear that issues such as intercontinental transport and transformation of chemically active species and the interactions between atmospheric chemistry and climate must also be addressed in order to better understand atmospheric chemical composition and to provide guidance to the public and policy-making community.

It is also clear that atmospheric chemistry must be viewed within an Earth System context, and as such IGAC will collaborate with partner projects, including two new projects of the International Geosphere Biosphere Programme, the Integrated Land-Ecosystem Atmosphere Process Study (iLEAPS) and the Surface Ocean Lower Atmosphere Study (SOLAS), and two of the World Climate Research Programme's (WCRP) projects, Stratospheric Processes and their Role in Climate (SPARC) and the Global Energy and Water Cycle Experiment (GEWEX).

You can download a PDF-version of this Plan from the IGAC website or request hard copies via e-mail (igac.seattle@ noaa.gov) and we will distribute it free of charge. Alternatively, contact the IGAC International Project Office, NOAA-PMEL, 7600 Sand Point Way NE, Seattle, WA 98115, USA.

Acknowledgements

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Executive Summary

The International Global Atmospheric Chemistry (IGAC) project was created in the late 1980s to address growing international concerns over rapid changes observed in the Earth's atmosphere. The project is cosponsored by the International Geosphere-Biosphere Programme and the Commission on Atmospheric Chemistry and Global Pollution of the International Association of Meteorology and Atmospheric Sciences.

The past decade of international atmospheric chemistry research – much of which was initiated and coordinated within IGAC – has greatly increased the 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., 2003). 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 the research effort of the first decade of IGAC was directed towards assessing the effects of anthropogenic emissions on the "background atmosphere" – the present-day atmosphere in areas remote from emission source regions. While questions remain concerning the point at which observed global-regional mean trends in component concentrations unambiguously rise above background natural variability, it is now well recognised that human activities have perturbed the chemical composition of the atmosphere at local, regional and global scales. These perturbations arise from emissions from fossil fuel and bio-fuel combustion and other industrial processes, anthropogenic enhancements of biomass burning, and increases in the lofting and transport of mineral dust through human-induced land use change.

Air pollution (defined herein as elevated pollutant concentrations and deposition fluxes resulting from anthropogenic emissions) is a serious and growing problem in many regions of the world. Furthermore, it is becoming increasingly clear that pollutants from one region can be rapidly and efficiently transported to other regions (e.g. from Asia to North America and from North America to Europe), making local emissions an international concern. In the industrialised 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 major agricultural regions of the world are co-located with industrialised 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, such as tropical and extra-tropical Asia and Africa, anthropogenic emissions (which are already quite high) are projected to increase substantially in coming decades due to the increasing energy and food demands of a growing population. In addition, the development and growth of mega-cities (densely populated cities of more than ten million people) and urban agglomerations (large core cities with surrounding satellite cities) must be considered 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, giving rise to the objective and research agenda for IGAC for the coming decade.

The objective of IGAC is:

to understand the role of atmospheric chemistry in the Earth System and to determine the effects of changing regional emissions and depositions, long-range transport and chemical transformations on air quality.

The research agenda of IGAC for the coming decade consists of seven research foci organised into two research themes.

Theme 1
The Role of Atmospheric Chemistry in Amplifying/Damping Climate Change

Focus 1.1 Ozone and Its Effect on Climate Change

Focus 1.2 Aerosol Distributions, Properties and Direct Radiative Effects on Climate

Focus 1.3 Effects of Aerosols on Clouds, Precipitation and Regional Hydrological Cycles

Focus 1.4 Effects of Gas and Aerosol Emissions/Depositions on Spatial Patterns of Climate Forcing

Theme 2
Effects of Emissions/Depositions, Transport and Chemical Transformations on Air Quality and Tropospheric Composition

Focus 2.1 Export, Transport and Transformation of Oxidants, Aerosols and Aerosol Precursors

Focus 2.2 Impacts of Intercontinental Transport on Surface Air Quality

Focus 2.3 Anthropogenic Impacts on the Cleansing Capacity of the Atmosphere

Addressing these research foci will require coordination and collaboration across the international atmospheric chemistry research community and its interdisciplinary partners in order to: (i) accurately determine global distributions of both short- and long-lived chemical components in the atmosphere and document their changing concentrations over time; (ii) provide a fundamental understanding of the processes that control the distributions of chemicals in the atmosphere and their impact on global change and air quality; and (iii) improve the ability to predict the chemical composition of the atmosphere over the coming decades by integrating understanding of atmospheric processes with the responses and feedbacks of the Earth System.

The results of this research should provide decision makers with the tools necessary to develop judicious policies to manage the atmosphere and its role in global change. Enhanced outreach to the public will be vital to ensure that new knowledge leads to changes in public attitude, policy and legislation.

Introduction

Over the past century humanity has greatly altered the chemical composition of the atmosphere. Worldwide emissions from growing industrial and transport activity and increasingly intensive agricultural practices have resulted in higher atmospheric concentrations of photochemical oxidants, acidic gases, aerosols and some toxic chemical species. Many of these air pollutants are known to be detrimental to human health and ecosystems.

Additionally, fossil fuel consumption together with agriculturally driven increases in biomass burning, fertiliser use, crop by-product decomposition, and animal-based food and fibre production have led to increasing emissions of key greenhouse gases, such as carbon dioxide, methane and nitrous oxide. While a significant fraction of these emissions is absorbed by the oceans and by terrestrial ecosystems, the remainder accumulates in the atmosphere, changing its composition. The atmospheric accumulation of radiatively active trace gases and the increased particulate loading appear to be responsible for much of the observed climate change of the 20th century, particularly the warming of the last few decades (Houghton et al., 2001). Predicted impacts of climate change include disruption of agricultural productivity, fresh water supplies, and terrestrial and marine ecosystems, as well as changes in the patterns of human diseases. Significant increases in sea level and changes in the frequency of severe weather events are also forecast. The effects of all of these stresses on biogeochemical cycles could exacerbate atmospheric composition change, leading to further climate change. If current trends continue, the predicted significant warming could trigger other changes in the climate system. Anthropogenic emissions are of course likely to change as climate patterns shift, making accurate predictions difficult.

Twenty years ago, scientific programmes addressing global tropospheric chemistry and related climate change were in their infancy. There were almost no large-scale observations of tropospheric composition, many chemical transformation mechanisms were unknown, and global atmospheric chemistry models were rather crude. The past decade has seen global atmospheric chemistry

research blossom. Much has been learnt about the global cycles (sources, transformations and sinks) of most of the important atmospheric chemical species, and satellite observations have provided a wealth of data regarding the chemical composition of the stratosphere. Multi-platform process studies of atmospheric chemical processes have recently been conducted on an unprecedented scale. Global chemical transport models can now simulate with some success the distribution of key tropospheric chemical species, and can provide simulations of future global atmospheric composition under different scenarios. Furthermore, short-lived, radiatively active substances such as ozone and aerosols are now incorporated as active constituents in most global climate models. As scientific understanding of the elements of atmospheric chemistry has developed, the necessity of understanding the linkage between atmospheric composition and other components of the Earth System has been more explicitly realised. Ten years ago, an "Earth System" perspective was a rather abstract idea. Feedbacks between, for example, changing climate and changing terrestrial emissions, or changing climate and changing atmospheric chemical composition, were not included in models. Now, a more quantitative understanding of the role of atmospheric chemistry in Earth System processes has been attained, and strategies are beginning to be developed to use this knowledge in a predictive capacity.

Global tropospheric chemistry and its links to the global climate system have received growing attention not only from the scientific community but also from decision makers in government and industry. Over recent decades, society has increasingly recognised the importance and value of the environment, and hence atmospheric chemistry research has had an increasing influence on environmental policy. In some cases international treaties to reduce emissions have been enacted and actions to protect the global environment have been taken. In spite of substantial advances in the understanding of fundamental atmospheric chemical processes, the predictive capability required for informed decision making remains limited, and the uncertainties in air quality forecasts and climate change predictions remain

high. Additionally, new and challenging problems at the chemistry-weather, chemistry-climate and chemistry-ecology interfaces are emerging that require considerable attention.

The International Global Atmospheric Chemistry (IGAC) project of the International Geosphere-Biosphere Programme (IGBP) began in the late 1980s, with cosponsorship from the Commission on Atmospheric Chemistry and Global Pollution of the International Association of Meteorology and Atmospheric Sciences. The first decade of IGAC contributed greatly to the advances in atmospheric chemistry, and culminated in a major integration and synthesis activity (Basseur et al., 2003).

Beginning in 1999, and as part of the IGAC integration and synthesis activity, a series of discussions and workshops was held to define the atmospheric chemistry research challenges for the coming decade – called IGAC phase II. These discussions defined the objective for IGAC and led to the definition of seven research foci organised into two research themes.

The objective of IGAC is:

to understand the role of atmospheric chemistry in the Earth System and to determine the effects of changing regional emissions and depositions, long-range transport and chemical transformations on air quality.

The two research themes of IGAC are: (i) the role of atmospheric chemistry in amplifying/damping climate change; and (ii) effects of emissions/depositions, transport and chemical transformations on air quality and tropospheric composition. These research themes and the seven subsidiary research foci are described in the following sections. Additionally, under the description of each of the seven foci, the measurements and modelling that will be required are outlined. There is of course significant overlap in the measurement and modelling needs of the different foci, and IGAC activities will often address several foci.

Although the primary foci of IGAC continue to be the composition (gases and aerosols) and chemistry of the atmosphere, the research themes and foci recognise that the atmosphere, hydrosphere, biosphere and pedosphere, form an interacting system whose components collectively determine the future evolution of the Earth. The research efforts of IGAC will therefore be coordinated with other activities of IGBP and the other partners of the Earth

System Science Partnership (ESSP). In particular, many of the research questions articulated herein require an understanding of the chemical exchanges between the atmosphere and the ocean, the atmosphere and the land, and between the middle atmosphere (stratosphere) and the troposphere – the troposphere being where IGAC has historically focused its efforts. On these aspects IGAC will work closely with, respectively, the Surface Ocean–Lower Atmosphere Study (SOLAS), the Integrated Land Ecosystem–Atmosphere Processes Study (iLEAPS) and the Stratospheric Processes and Their Role in Climate (SPARC) projects. IGAC activities will both feed into, and benefit from, the work of the Global Emissions Inventory Analysis (GEIA) – an activity of the Analysis, Integration and Modelling of the Earth System (AIMES) project.

The Role of Atmospheric Chemistry in Amplifying/Damping Climate Change

Theme 1

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 substances is still extremely low (e.g. Figure 1) (Houghton et al., 2001). A major theme of IGAC is 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.

Part of the uncertainty in the calculated radiative forcing by various components stems from the difficulty in defining the "background atmosphere" and the natural variability of this background (e.g. Figure 2). The background atmosphere, despite decades of research within IGAC and

Figure 1. Mean global industrial era anthropogenic aerosol forcing (left axis) as estimated by forward (A–F) and inverse (G–L) calculations, and as used in applications (M–Q). Forward calculations are based on atmospheric aerosol physics and chemistry. Inverse calculations infer aerosol forcing from the total forcing required to match model simulations with observed temperature changes. Applications values are from model studies with anthropogenic aerosol forcing as a driver of climate change (and with input aerosol forcings consistent with inverse calculations). Circles with error bars are central values and 95% confidence limits. Bare error bars are stated range. Squares represent specific forcing calculations using alternative formulations within the same study. Right axis indicates total industrial era forcing assuming a non-aerosol forcing of 2.7 W m^{-2}. From Anderson et al. (2003).
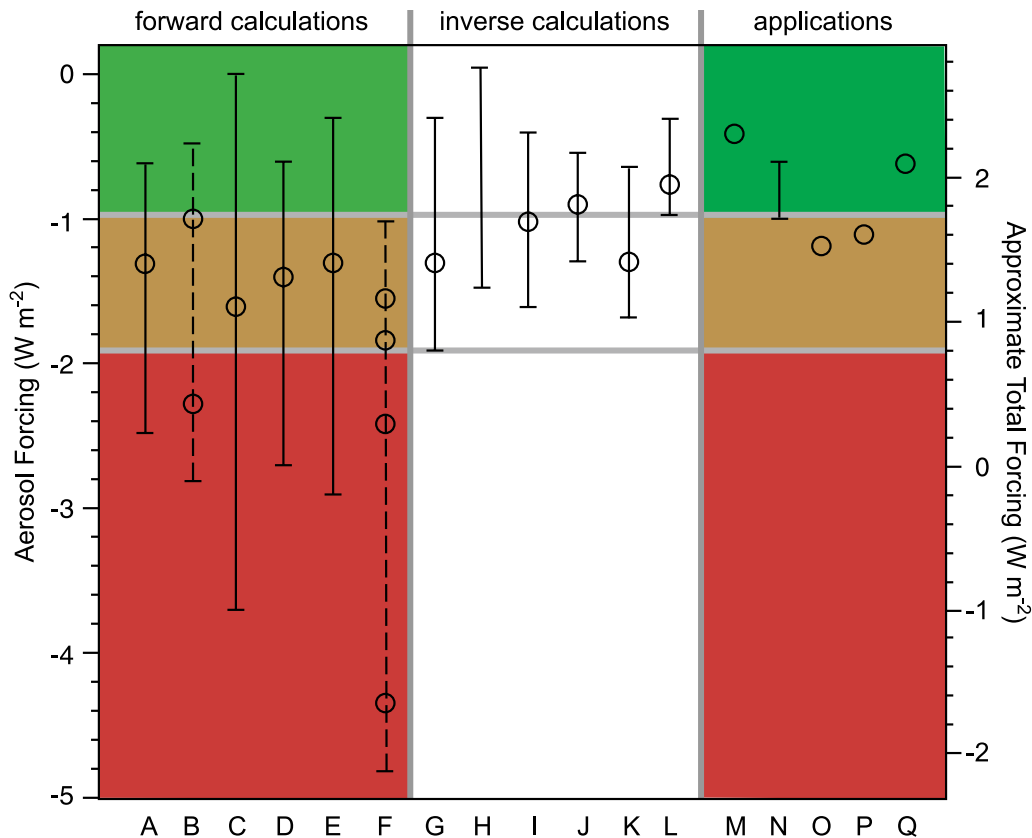
other programmes, is still not clearly defined. Normally it has been taken to describe the chemistry of remote regions of the atmosphere that are free from human influence. One of the lessons of past atmospheric chemistry research is that such regions no longer seem to exist; even the remotest regions, such as the central Pacific and the polar stratosphere, show substantial influences from anthropogenic emissions for at least part of the year (e.g. Chin and Jacob, 1996; Mari et al., 1998; Tegen et al., 2000; Hauglustaine et al., 2004; Quinn and Bates, 2005).

Many attempts have been made to simulate pre-industrial atmospheric chemistry, but the results of these studies vary widely (e.g. Shindell et al., 2005; Grenfell et al., 2001; Hauglustaine and Brasseur, 2001; Houweling et al., 2000; Wang and Jacob, 1998; van Dorland et al., 1997). Pre-industrial trace gas measurements are too sparse and generally too uncertain to enable reliable assessments. In defining the background atmosphere it is also unclear whether it is necessary to account for the indirect effects of climate change, for example, the changes in circulation

patterns (source/receptor regions) and rainfall that can affect atmospheric chemistry. Furthermore, the natural variability of circulation and emissions patterns, which can be quite complex, needs to be accounted for when defining the range of conditions that are considered "background". In spite of natural variations, a reference state is needed in order to determine current and future atmospheric change induced by human activities, and in order to assess these changes in a socio-political context. Improving the understanding of the background atmosphere and its natural variability will be an underpinning theme of all IGAC projects.

IGAC research to address these issues is organised into four foci: (i) ozone and its effect on climate change; (ii) aerosol distributions, properties and direct radiative effects on climate; (iii) effects of aerosols on clouds, precipitation and regional hydrological cycles; and (iv) effects of gas and aerosol emissions/depositions on spatial patterns of climate forcing. Each of these is considered in the context of a variable and changing climate system.

Figure 2. Box plots of regional sea salt concentrations for the sub- (a) and super-micron (b) size ranges and true wind speed (c) reported at a relative humidity of 55 ± 5%. Box values are the 25th, 50th and 75th percentiles, black squares denote the mean, gold bars denote the 5th and 95th percentiles, crosses denote the 1st and 99th percentiles, and the grey bars denote the maximum and minimum values. From Quinn and Bates (2005).

Experiment Substance	Regions
ACE 1 Marine "background" aerosols	Southern Ocean south of Australia
ACE 2 Cont Aerosols from Europe	Sub-tropical NE Atlantic
AER99 Dust African dust sources	Tropical Atlantic
AER99 BB Aerosols from biomass burning	Tropical Atlantic
IND Arabia Aerosols from Arabia	Northern Indian Ocean
IND India Aerosols from India	Northern Indian Ocean
AA Poll Pollution from Asia	E. coast of Japan, Sea of Japan, E. China Sea
AA Poll + Dust Pollution/dust from Asia	E. coast of Japan, Sea of Japan, E. China Sea
NEAQS Cont Pollution/dust from U.S.	U.S. Eastern Seaboard
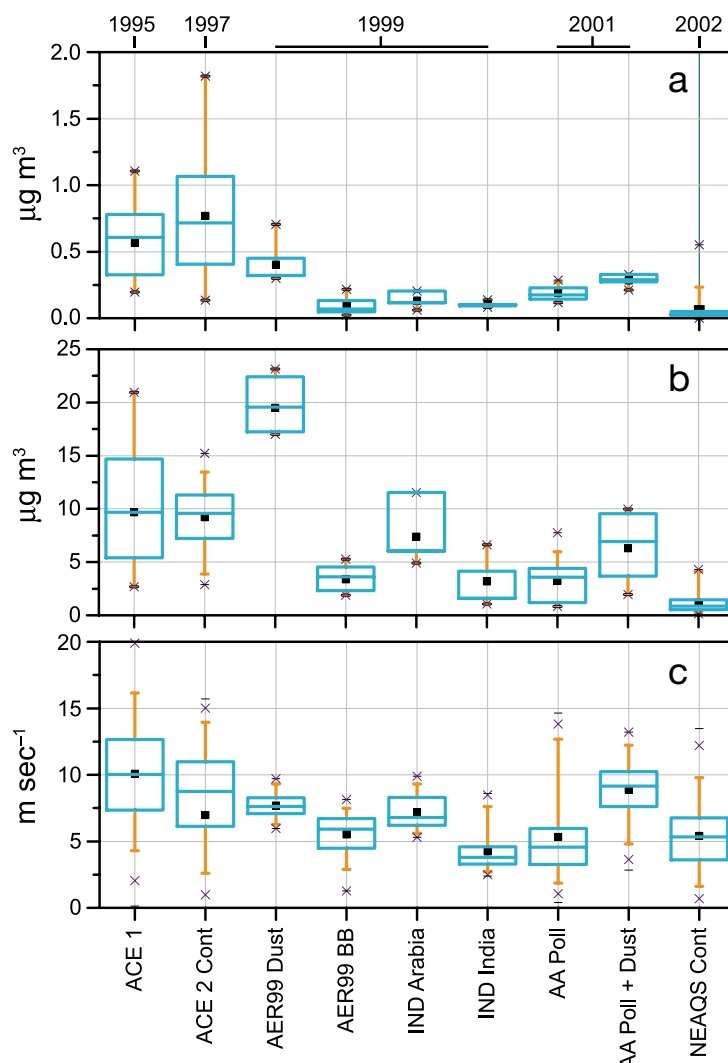
Focus 1.1: Ozone and Its Effect on Climate Change

This focus considers the relative roles of stratosphere-troposphere exchange, anthropogenic and natural precursor emissions, *in situ* photochemical processes and tropospheric transport in controlling ozone and its effect on climate change.

Ozone is a powerful oxidising agent and its presence in the troposphere affects human health (via lung tissue damage) and plant health (via leaf damage). While both particles and ozone affect human health, plant damage (including of native vegetation, production forests and agricultural crops) is largely caused by ozone. In developed countries most crops are grown in summer when ozone concentrations are highest, thus reducing crop yields. In many developing countries, population, ozone levels and agricultural demand are all increasing simultaneously, making this an important sustainable development issue.

In the first phase of IGAC the coverage of ozone observations (mapping and climatology) was greatly increased and the processes affecting ozone photochemistry and transport were elucidated. Models of various complexity and scale – from zero-dimensional to coupled global chemical transport models – have improved the ability to interpret observations, to model the past and to predict the future. *In situ* observations and modelling complement an enhanced global mapping capability provided by a new generation of satellite-borne instruments.

IGAC will tackle three aspects of tropospheric ozone chemistry. Firstly, IGAC will seek to improve the spatial, vertical or temporal resolution of observations of ozone and related constituents and parameters in order to quantify the processes controlling ozone and its effect on climate change. Secondly, IGAC will study transport processes as sources and sinks of ozone in the troposphere. Deep convective mixing and stratosphere-troposphere exchange closely tie this aspect to interests of SPARC. Boundary-layer exchange and long-range horizontal transport also control local concentrations of ozone, as considered under Focus 2.2. Thirdly, IGAC will study the basic

chemical processes related to the formation and destruction of ozone and other oxidants (see also Focus 2.3).

Ozone Measurement

There are several approaches needed for global ozone mapping: satellite observations, *in situ* measurements and modelling. Early in the first phase of IGAC, satellite observations were limited to climatologies of tropical and approximate (i.e. multi-month averages) mid-latitude concentrations. More recent refinements provide daily, near real-time tropospheric ozone column measurements in the tropics (since 1997; Ziemke et al., 1998 and 2000) and the extra-tropics (since 1995; Piters et al., 1998; Corlett and Monks, 2001). However, data coverage and accuracy are sometimes limited, and clouds and aerosols are known interferences (Koelemeijer and Stammes, 1999; Torres and Bhartia , 1999).

During the second phase of IGAC a new generation of ozone sensors – three on the NASA Aura satellite and the European Space Agency (ESA) Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) instrument – will be operational. These will complement the carbon monoxide and methane column measurements from the Measurements of Pollution in the Troposphere (MOPITT) instrument and other instruments (Gille et al., 2002; van der A, 2001; Deeter et al., 2003; Smith, 1997; Drummond and Mand, 1996). Satellite instruments will measure column ozone (and other trace gas) amounts; the challenge is to use the data from these instruments to their full advantage. Models that assimilate satellite data are being developed and these should provide some predictive capability.

Coordinated *in situ* measurements of ozone and other chemical constituents are required in order to better define the photochemical ozone budget and the interaction of chemical and dynamical processes within a given regime. This will require vertical profiles from soundings and from commercial aircraft at regular locations and intervals. The Measurements of Ozone and Water Vapour by In-service Airbus Aircraft (MOZAIC) and the Civil

Aircraft for Regular Investigation of the Atmosphere Based on an Instrument Container (CARIBIC) projects are providing routine *in situ* measurements of vertical profiles from commercial aircraft at major transportation hubs (Marenco et al., 1998; Zahn et al., 2002; Brenninkmeijer et al., 1999). A network of regular ozonesonde launches could record large-scale variability, stratosphere-troposphere exchange, pollution layers, convective transport and seasonal and inter-annual variations, together with vertical structure.

Globally distributed, regularly scheduled soundings are also critical for deriving global tropospheric ozone trends. At the World Ozone and Ultra-violet Data Centre of Environment Canada ozonesonde data are archived for open community use. Late in the first phase of IGAC the Southern Hemisphere Additional Ozonesondes (SHADOZ) activity coordinated ozonesonde launches in the southern tropics and sub-tropics to obtain more than 1,300 profiles for this under-sampled region (Thompson et al., 2003a,b; Figure 3). The value of balloon-borne measurements increases when supplemented with cam-

paign and satellite observations. For example, in the IGAC Southern African Fire Atmospheric Research Initiative (SAFARI) and the NASA Transport and Atmospheric Chemistry near the Equator – Atlantic (TRACE-A) campaigns, data from more than a hundred ozonesondes were used to evaluate the relative contributions of Africa and South America to ozone pollution in the south Atlantic. The Global Atmosphere Watch activity of the World Meteorological Organization (WMO) is adding ozone sounding capabilities to developing nation observing programmes. IGAC will also work with the Integrated Global Observing System Partnership (IGOS-P) to develop a more complete global measurement network, and to facilitate the synthesis of *in situ* and balloon-borne measurements with the growing body of satellite observations and modelling studies.

The tropospheric ozone budget is primarily controlled by photochemical production and loss within the troposphere. In order to understand and predict ozone trends it is essential to understand ozone precursors distributions and budgets. The distributions of many key ozone precur-

Figure 3. Contours of 0.25 km–averaged ozone values (ppbv) over the south tropical Southern Hemisphere based on SHADOZ soundings for September–November during 1998–2000. The tropopause (15–17 km alt.) is a layer rather than a thin barrier, and dynamic as well as photochemical inputs of ozone appear to enhance ozone concentrations. From Thompson et al. (2003b).
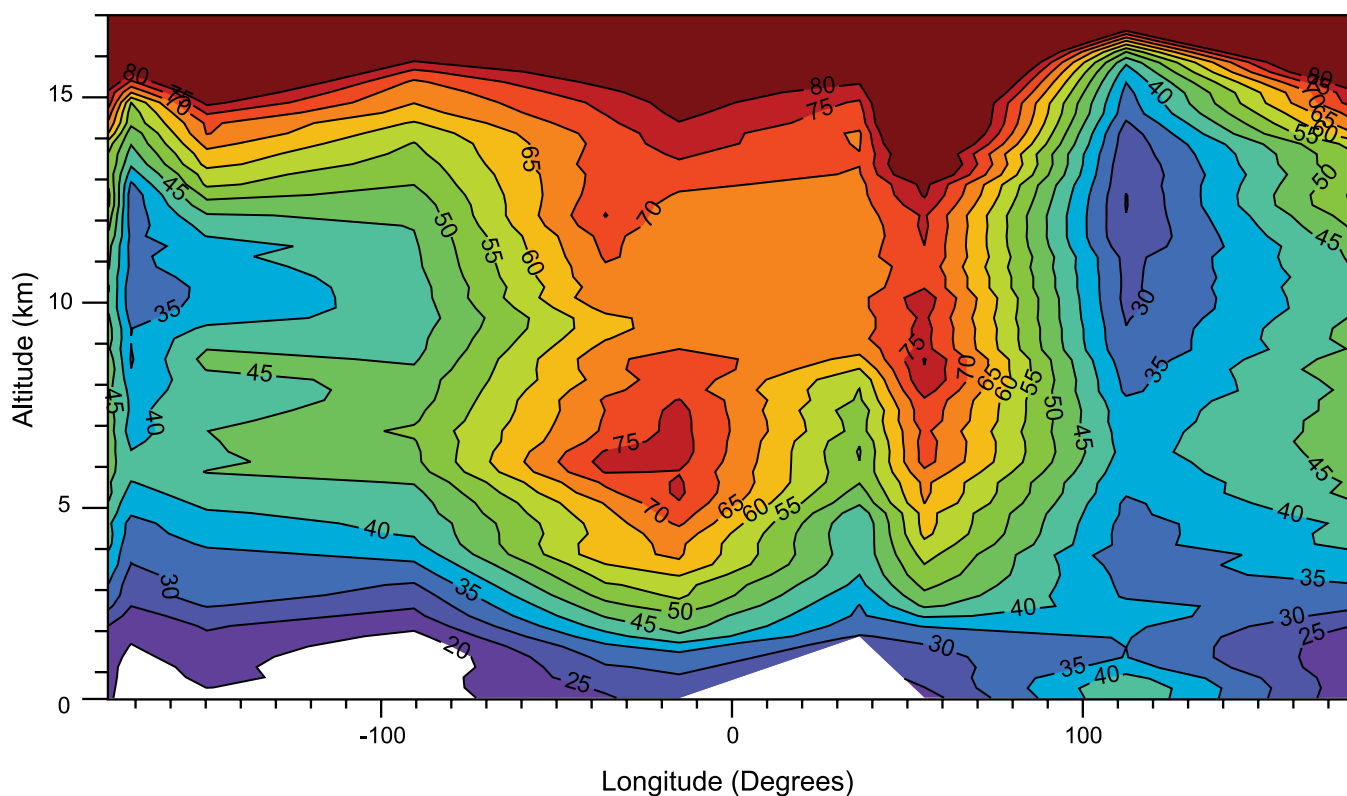
sors are highly variable and heterogeneous because of their relatively short lifetimes and widespread emission sources. For example, concentrations of nitrogen oxides can vary by several orders of magnitude – from a few hundred parts-per-billion by volume (ppbv) in highly polluted urban centres to a few parts-per-trillion in the remote troposphere (Brunner et al., 2001). Some hydrocarbons, particularly naturally emitted hydrocarbons, can vary by a similar degree. The large variability in, and lack of knowledge of, some important sources of nitrogen oxides and hydrocarbons makes it difficult to accurately simulate ozone-precursor distributions (Brunner et al., 2003; Lauer et al., 2002; Zhang et al., 2000; Holland et al., 1997). Photochemical ozone production is a non-linear function of the concentrations of nitrogen oxides and hydrocarbons, hence modelled ozone budgets can be uncertain due to the difficulty in simulating ozone-precursor distributions (Kleinman , 2005; Esler et al., 2004; Lawrence et al., 1999; Penner et al., 1998; Lin et al., 1988). Measurement efforts should therefore focus not just on ozone, but also consider the distribution of its precursors and on what controls these distributions. This process-level understanding will help in model evaluation.

In situ measurements of ozone precursors – including reactive odd nitrogen and a large number of non-methane hydrocarbons – have been highly successful both at ground stations and on aircraft in field campaigns. In particular, aircraft measurements over remote areas under the IGAC East Asia–North Pacific Regional Experiment (APARE) (Lui and Akimo, 1995; Hatakeyama et al., 2001), the IGAC North Atlantic Regional Experiment (NARE) (Peterson et al., 1998; Duderstadt et al., 1998; Penkett et al., 1998; Stohl et al., 2002), TRACE-A, SAFARI (Andreae et al., 1996), and the Indian Ocean Experiment (INDOEX) (Naja et al., 1999) have greatly increased knowledge of ozone and ozone-precursor distributions and budgets, and such measurements should continue. Concurrent satellite measurements of ozone and ozone precursors can potentially revolutionise the understanding of ozone and ozone-precursor budgets and distribution by substantially increasing the spatial and temporal data coverage, albeit at lower resolution than *in situ* measurements. Finally, satellite measurements will provide distributions of key tracers that can be used to answer questions regarding stratosphere-troposphere exchange.

Constituents that contribute significantly to the budgets and distributions of odd hydrogen components (i.e. the hydroxyl and hydroperoxyl radicals) should also be mea-

sured. Like ozone, these species help determine the cleansing capacity of the atmosphere. Water vapour is essential in the production of hydroxyl and hydroperoxyl radicals. Accurate measurements and good spatial and temporal coverage of water vapour are still lacking, especially in the upper troposphere and lower stratosphere. Hydrogen peroxide, methyl hydroperoxide, formaldehyde, acetone and similar organic compounds have been shown to be important sources of hydroxyl and hydroperoxyl radicals in certain tropospheric regimes. Satellite techniques for observing these components are currently being developed.

Transport Investigations

IGAC will investigate four dynamic processes that affect global tropospheric ozone distributions: (i) stratosphere-troposphere exchange via large-scale circulation and small-scale processes (i.e. tropopause folds); (ii) deep convection; (iii) large-scale horizontal advection; and (iv) exchange between the Earth surface, the boundary layer and the free troposphere.

Stratosphere-Troposphere Exchange

Originally it was believed that all tropospheric ozone was stratospheric in origin, owing to its relatively long photochemical lifetime and the clear connection through folding events in mid-latitudes and through large-scale circulation and mixing in the tropics. Once photochemical sources of ozone became evident in polluted regions, and models demonstrated that globally the photochemical source was comparable to transport, more attention was given to the role of chemistry (Lelieveld and Dentener, 2000). In its first phase IGAC focused on experimental and modelling initiatives to elucidate regional- to global-scale ozone generation processes. These processes include combustion in urban areas, biomass burning in rural areas and lightning (a major source of the ozone precursor nitric oxide).

Recent observations point to the critical role of dynamics and transport in determining tropospheric ozone concentrations, but show however, that the role of dynamics goes beyond simple stratosphere-troposphere transfer. For example, ozone and tracer data from aircraft and soundings (e.g. Figure 3) have overturned the view of the tropopause as a fixed barrier between the stratosphere and troposphere in the tropics (e.g. Dessler, 2002; Gettelman and Forster, 2002; Scott et al., 2001; Morgenstern and Carver, 2001). Stratospheric ozone in the subtropics penetrates as low as 2–3 km in connection with cut-off

low-pressure systems and deep troughs (Rondanelli et al., 2002; Baray et al., 1998). Additionally, throughout the tropical free troposphere there is a zonal "wave-one" feature – with 10–15 Dobson Units (1 Dobson Unit = 2.69 x 10^{16} molecules cm^{-2}) greater ozone column thickness over the Atlantic compared to the Pacific – which persists throughout the year. Although seasonal biomass burning contributes to a south Atlantic "ozone maximum" and to the wave, so do lightning and subsidence of upper troposphere–lower stratospheric ozone in all seasons (Thompson et al., 2003c; Fujiwara and Takahashi, 2001; Thompson et al., 2000; Randriambelo et al., 2000).

Complex dynamical processes in synoptic-scale systems impact ozone concentrations at local to global scales. Observational campaigns at mid-latitudes in the 1990s showed that tropopause fold events are very common. Sharp gradients at frontal system edges can be seen in satellite data, and at the polar front 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. Similarly, it has been shown that most mid-latitude ozone pollution events are associated with the penetration of moist, subtropical air beyond conventional "tropical" latitudes (e.g. Jaeglé et al., 2003; Shakina et al., 2001; Langford, 1999; Suhre et al., 1997; Gouget et al., 1996; Loring et al., 1996). Thus air masses have no latitude boundaries and hence definitions of tropical, subtropical, mid-latitude and polar air masses are not rigid.

Satellite data from the past twenty years show a connection between ozone and the processes associated with tropical climate variability (e.g. El Niño–La Niña and the Indian Ocean Dipole) (Stevenson et al., 2005; Ziemke et al., 2005; Ziemke and Chandra, 2003; Zachariasse et al., 2001; Thompson et al., 2001). Even during the extreme pollution event that followed the 1997 El Niño-triggered Indonesian fires, there was a large increase in upper tropospheric ozone brought about by perturbed dynamics which was comparable to the pyrogenic ozone over the larger Indian Ocean basin (Kita et al., 2000; Chandra et al., 1998).

Deep Convection

Building on mid-1980s observations, several campaigns by IGAC and other organisations established convection as a mechanism for introducing boundary-layer air into the free troposphere (Thompson et al., 1997). In areas with deep convection, both gases and aerosols emitted at the surface can be lofted out of the boundary layer, significantly increasing their lifetime. This has implications for the chemistry of both the upper troposphere and the lower stratosphere and affects the radiative impact of these species. 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). The TTL is several kilometres thick, extending from the secondary tropical tropopause (~12–14 km) – above which convective outflow begins decreasing rapidly – up to the tropical tropopause (~16–18 km) as defined by the temperature minima. The nature of these layers, including the climatology of water vapour and the evolution of thin cirrus (and its radiative forcing), is a subject of intensive field investigations. The transport of water vapour, methane and other chemically active species into the lower stratosphere is of particular interest to SPARC. In its second phase IGAC will collaborate more closely with SPARC in the study of stratosphere-troposphere exchange, its role in controlling various species and how it might change under a shifting and variable climate system.

Convection is not only an efficient transport mechanism, but is also important in scavenging chemical components from the atmosphere via convective precipitation. The role of ice in convective clouds is particularly crucial. Convection also produces lightning, most intensely over land, as seen by satellites and ground-based lightning detectors. The nitrogen oxides from lightning lead to ozone formation. The role of convection in global ozone formation can be assessed using models, although convective and lightning parameterisations still differ between models.

Horizontal Advection

Many of the regional field initiatives of IGAC (e.g. APARE, NARE, the Southern Tropical Atlantic Region Experiment and especially the Aerosol Characterisation Experiment (ACE-Asia) have demonstrated the truly global nature of pollution transport (Huebert et al., 2003; Cahill, 2003; Price et al., 2003; Gong et al., 2003; de Gouw et al. 2004; Nowak et al., 2004). Campaigns during the 1990s in the Indian Ocean region and the tropical Pacific found high pollution levels in areas expected to be relatively free of human influence (Krishnamurti et al., 1998; Jayaraman et al., 1998; Satheesh and

Moorthy, 1997). Clear evidence of North American transport reaching Europe (Huntrieser and Schlager, 2004; Li et al., 2002; Derwent et al., 1994) and Asian transport reaching North America (Bertschi et al., 2004; Jaffe et al., 2004; Killin et al., 2004; Price et al., 2004; Weiss-Penzias et al., 2004) have also been found. This may have significant implications for pollutant regulations in receptor regions, and is certainly significant in understanding what controls the global distribution of chemically active species. There is a specific need to understand how densely populated areas (mega-cities and metro-agro complexes) impact atmospheric chemistry at regional to global scales, especially given the pace of urbanisation around the globe. Accordingly, Intercontinental Transport and Chemical Transformation (ITCT) and Mega-cities are important new IGAC activities.

Surface–Boundary Layer–Free Troposphere Exchange

Chemical processes in the boundary layer determine ozone concentrations near source regions. Inputs from the biosphere-hydrosphere and from human activities supply trace gases that lead to ozone formation. Complex processes involved in chemical transformations within the boundary layer are important, but it is interactions between the boundary layer and the free troposphere that determine the larger regional to global impact. Fluxes of ozone and its precursors from the boundary layer to the free troposphere are inadequately measured and poorly represented in simulations. Multi-scale models are needed for proper representation. For ozone, its precursors and associated gases (e.g. carbon monoxide, formaldehyde and bromine oxide), global satellite observations will be useful, especially when measured at improved vertical resolution.

Focus 1.2: Aerosol Distributions, Properties and Direct Radiative Effects on Climate

Atmospheric aerosols are the largest source of uncertainty in current estimates of anthropogenic radiative forcing (Houghton et al., 2001). They are both microphysically and chemically complex, and their spatial (both vertical and horizontal) distribution is highly variable. An improved understanding of the factors controlling aerosol composition and distribution is required to improve climate predictions and to guide sustainable development. Existing and imminent satellite missions offer extraordinary opportunities for improving knowledge of global aerosol distributions. However, to understand climate perturbations, aerosol radiative effects must not only be quantified, but must be attributed to chemical components, and ultimately, to sources.

The challenge for Focus 1.2 is to undertake a global assessment of aerosols, linking radiative properties to chemical components. The enormous current uncertainties in source strengths and lifetimes of different aerosols mean this assessment must be based on observations (Kinne et al., 2003). Chemical attribution of observed, global aerosol optical depth (AOD) will provide a foundation for evaluating present direct climate forcing by anthropogenic aerosols, as well as for estimating past and future aerosol impacts. These impacts include those associated with pre-industrial aerosols (e.g. dust and natural biomass burning) and those associated with aerosol source changes projected under various emission scenarios.

Focus 1.2 seeks the best estimate of current AOD (including its geographical and temporal variations) and its attribution in terms of major chemical components (i.e. sulphate, organic compounds, black carbon, nitrates, dust and sea salt; see Figure 4). While satellites and chemical transport models necessarily operate at the global scale, direct chemical measurement of aerosols is always at the local scale. Global characterisation will require integration of aerosol-sensing satellites, chemical transport models and *in situ* measurements. IGAC will foster the acquisition of high quality, consistent and scientifically useful

data across diverse regions, and will provide a forum for identifying critical regions and processes in order to encourage the scientific community to focus on the most important data gaps.

Figure 4. Radiative forcing by tropospheric aerosols: aerosols absorb and scatter solar radiation, and act as cloud condensation nuclei thereby determining the initial cloud droplet number concentration, albedo, precipitation formation and lifetime of warm clouds.
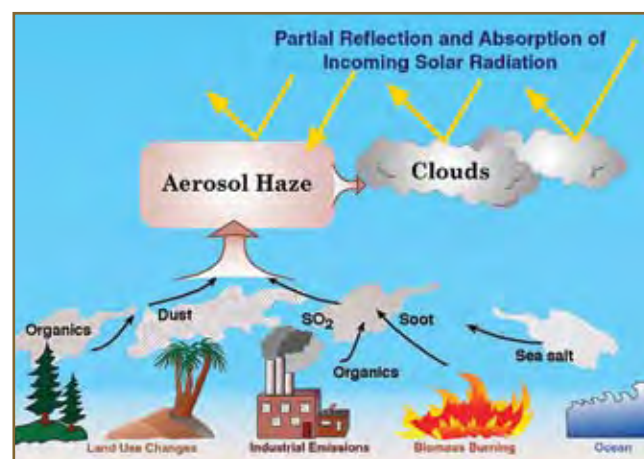
Initially, a preliminary assessment of global AOD and the fraction of AOD due to major natural and anthropogenic components will be undertaken by comparisons of chemical transport models, comparisons of satellite-derived AOD climatologies and integrated analyses of results from recent field campaigns. These comparisons will also identify the major sources of uncertainty in present AOD estimates, and will help with the development of a research strategy for reducing these uncertainties.

Ground-based Measurements

Several organisations currently support ground-based measurements of aerosol chemistry, although few sites have coordinated methods of sampling and reporting data, and even fewer have long-term support for co-located 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 aerosols dominate the entire column of AOD. In coordination with column-closure experiments that test these assumptions, ground-based chemical data can play a major role in a global assessment. However, considerable coordination will be required given that: (i) measurement methods must be consistent and data quality known; (ii) chemical measurements must be co-located and simultaneous with AOD measurements; and (iii) local data must be included in 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 less than five years are firstly, implementation of well-calibrated AOD measurements in polluted regions where chemical measurements are already being made, and secondly, implementation of aerosol chemical measurements at selected Aerosol Robotic Network sites which are proving to be useful not only for satellite calibration (and evaluation), but also as a tool for studying aerosol particles.

Aircraft-based Measurements

Methodologies for column-closure experiments with aircraft have advanced rapidly in recent years. Column-closure experiments produce tightly constrained, vertically-resolved information on aerosol properties that provide an excellent basis for AOD attribution. A global AOD assessment requires a statistically significant number of these data sets coincident with satellite overpasses in regions of high impact.

Routine transit flights by dedicated research aircraft (probably small or even unmanned aircraft) through regions of significant aerosol loading could also provide large, semi-random data sets for chemical characterisation beneath satellites. These would enable assessment of horizontal variation, especially at the edges of globally significant aerosol plumes. Flights would be mostly in the boundary layer – or main aerosol layer, if aloft – with occasional profiles to assess vertical variations (e.g. Figure 5).

Targeted Experiments

To explain 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 parameterisations. Experiments are required

using sufficient measurement platforms and instruments to over-constrain a specific chemical or microphysical mechanism. Such targeted process studies rely on coordinated modelling to identify regions where specific processes are not well known and may have large impacts. An example is the processes that contribute sulphate, nitrate and organic components to mineral dust in Asian and Saharan outflow layers. Recent measurements suggest that as much as half of dust-derived AOD originates from anthropogenic pollution (Arimoto et al., 2004). Resolving such issues requires quantification of component sources and of the processes controlling their gas-phase reactions, gas-to-particle transfers and surface reactions through laboratory studies, followed by *in situ* measurement of the evolution of these components using Lagrangian-type experiments. The high cost and high level of coordination required to measure a time-history of chemical evolution in both the gas and particle phases necessitates multinational coordination and funding.

Satellite-based Measurements

The global aerosol characterisation envisioned by IGAC will largely be based on satellite observations. It is therefore 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 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) through the meso-scale (updraft versus downdraft regions within cloud complexes) down to the scale of individual clouds (mapping concentration as a function of distance from cloud edge).

Aerosol Variability

Besides providing fundamental insights into the nature of aerosols, variability information is needed at sub-grid scales 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 necessary to establish the characteristic times and distances of the processes that change aerosol composition and loading. Variability assessment will require acquisition of statistically significant amounts of data at a resolution greater than the scales to be investigated, and over domains sufficiently large to encompass the scales to be investigated.

Aircraft-based measurements during IGAC's ACE-Asia campaign in March–April, 2001 revealed the high complexity of the aerosol layers in the east Asian region. In this profile, taken over the eastern Yellow Sea near Pyongyang from the NCAR C-130 aircraft, a highly layered structure is observed hundreds of kilometres downwind of the dust and pollution source regions. Below 2 km altitude the aerosol is dominated by pollution; above this, the fine mode (pollution) aerosol and SO_2 decrease and instead there are layers composed primarily of coarse mode aerosol (desert dust). This complex vertical structure shows the challenges of modelling the atmospheric chemistry in this region, and it points out the challenges of satellite-based instruments where retrievals are of column-integral values. Credit: Sarah Doherty, Tad Anderson and Byron Bloomquist.

Legend:
- SO$_2$ concentration
- <1 μm aerosol light scattering at 500 nm
- total aerosol light scattering at 500 nm

Y-axis: Altitude (m)
X-axis: Light Scattering (Mm^{-1}) & SO$_2$/100 (pptv)
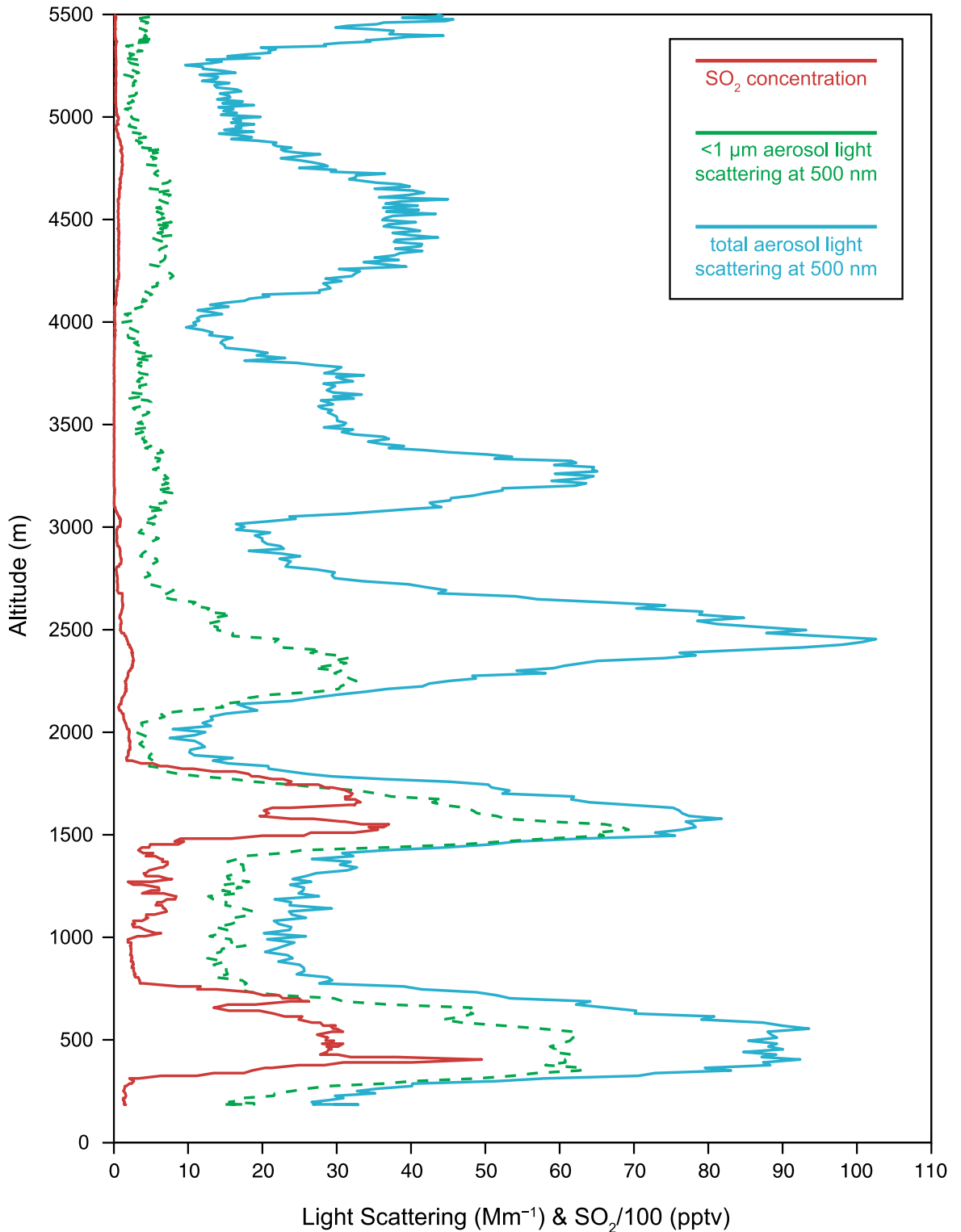
Focus 1.3: Effects of Aerosols on Clouds, Precipitation and Regional Hydrological Cycles

The indirect radiative effects of aerosols are certainly the most uncertain and potentially the largest of known aerosol forcings. They also represent an important non-linearity in the climate system. The relationship between the forcings (e.g. changes in aerosol concentration and properties), their effects (e.g. changes in cloud albedo, lifetime, extent or precipitation development) and climate responses (e.g. changes in surface temperature and precipitation patterns and intensity), and the concomitant changes in aerosol and aerosol-precursor emissions, are currently very poorly understood and are likely to be highly non-linear. A major challenge is bridging the gap between the scales of the driving process (micrometres and seconds) and the scales of the resultant effects (regional to global and days). A further challenge is bridging the gap between case-study observations of the indirect effects of aerosols (e.g. Figure 6) and the regional- to global-scale impacts.

Atmospheric chemistry is central to understanding the indirect radiative effects of aerosols. A fundamental understanding and predictive capability is required of how aerosols and gases are incorporated into cloud droplets, and how they influence droplet size distribution, optical properties and phase of clouds. The aerosol-cloud-climate system is a multi-phase, multi-component dynamical system with non-linear feedbacks. Because of this complexity, development of predictive capability must focus on intimately linking observations (*in situ* and remotely sensed), laboratory studies and model development.

The central question concerns the role of aerosol chemical composition (inorganic and organic) in determining the indirect radiative forcing of aerosols. To answer this question the processes by which different aerosols and gases determine the microphysical and radiative properties at cloud-top and affect precipitation development need to be determined. This requires determining the effects that *all* aerosol chemical components have on

cloud development – especially organic components for which understanding is extremely limited (Kanakidou et al., 2005; Fuzzi et al., 2006). Additionally, it will be important to be able to detect and quantify changes in cloud properties due to the aerosol influences. Detection of the effects is required for testing model predictions.

Focus 1.3 must indeed be very focused, as it is impossible to tackle all aspects of the problem simultaneously. Focus 1.3 will combine *in situ* measurements of cloud and aerosol microphysics and chemistry, detailed process models and large-scale models, and airborne and satellite remote sensing measurements of cloud radiative characteristics, in order to improve the understanding of the processes relating aerosol properties, cloud albedo and precipitation development.

Laboratory Studies

Laboratory studies to determine the fundamental properties of organic compounds (or mixtures of organic and inorganic components) in aerosols such as water solubility, vapour pressure over aqueous solutions and surface activity should be expanded. These properties must be known in order to develop thermodynamic equilibrium models for aerosols with organic components, and to further develop detailed process models of aerosol-cloud interactions.

Observations

Observational campaigns provide the most direct information on atmospheric processes, but are limited in time and space and hence only sample a limited range of conditions. Important measurements include airborne observations of microphysical and chemical properties of below-cloud aerosols, of cloud droplets and interstitial particles, and of the upwelling radiation above cloud-top. These measurements will explain how inorganic and organic aerosols influence the microphysical development of the cloud-drop size spectrum throughout

Figure 6: Emissions from ships off the coast of France and Spain leave behind "ship tracks" in the otherwise clear areas between the marine stratus clouds. This image, from the Moderate Resolution Imaging Spectroradiometer (MODIS) on the Aqua satellite, is from January 27, 2003, a day with an unusually high number of ship tracks. While ship tracks present a very visually striking example of aerosol-cloud interactions, it is still unclear whether anthropogenic aerosols are resulting in climatically-significant change in cloud radiative forcing at a global scale. Image courtesy Jacques Descloitres, MODIS Land Rapid Response Team and Mark Gray, MODIS Atmosphere Science Team, both at NASA GSFC.

the entire depth of the cloud (including precipitation development). These measurements will also be coupled with satellite retrievals of cloud albedo, effective radius and precipitation.

Initial campaigns should target areas where substantial gradients in organic aerosols are likely to be co-located with low-level clouds. Examples include where urban plumes advect into a relatively non-polluted marine boundary layer, and where carbonaceous aerosols have regional-scale impacts – for example, biomass-burning plumes in central Africa or Amazonia. The rationale is to contrast areas with similar dynamic characteristics but very different chemical systems (e.g. aerosols and gases, organic and inorganic aerosols), in order to isolate the effects of organic aerosols, while retaining the advantages of *in situ* observations.

Modelling

Models are necessary for exploring conditions not encountered or observable in the field, and for prediction. Models can be used to study regional to global scale features (not measurable *in situ*) and features that are difficult or impossible to measure using satellite-based sensors (e.g. vertical structure, near-cloud effects and individual chemical reactions). Because *in situ* and satellite observations alone cannot provide a complete picture of the atmosphere, models are needed to test hypotheses that arise from data analysis. Detailed models will be used to study the organic aerosol processes that influence cloud properties, enabling parameterisation of large-scale models based on a sound knowledge of cloud physics and chemistry. This will help determine which processes can be omitted from, or treated coarsely in, large-scale models.

Focus 1.4: Effects of Gas and Aerosol Emissions/Depositions on Spatial Patterns of Climate Forcing

Observed changes in atmospheric composition provide robust and dramatic evidence for human influences on the Earth System. 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 stabilise changes in the climate system in accordance with the United Nations Framework Convention on Climate Change (UNFCCC).

Changes in atmospheric composition affect climate through changes in the abundance and distribution of radiatively active components, thus altering the energy balance of the atmosphere. In the late 1990s and early 2000s considerable progress was made in determining the roles of different radiatively active components, but this research also revealed new levels of complexity and posed new questions. Climate change can no longer be thought of as driven simply by increases in atmospheric carbon dioxide; even though carbon dioxide may be the single most important anthropogenic greenhouse gas, the combined effect of other greenhouse gases and different aerosols – with cooling and warming effects – is at least as important. Climate model validation, climate projections and the policy tools and framework for managing human influences on climate must all account for the actual and potential evolution of a wide range of radiatively active components.

Increased understanding of the linkages between atmospheric composition and the radiative energy balance challenges some of the simple approximations that have been used to link observed atmospheric change to climate models and policy frameworks. The top-of-atmosphere radiative forcing concept has been used as a nexus between global mean greenhouse gas concentrations and representations of radiative perturbation in climate models. However, radiative forcing is an approximation developed for long-lived components that are well mixed throughout the atmosphere. Questions arise when this concept is applied to short-lived components with large spatial variations arising from spatial variations in emissions (e.g. Figure 7). 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 is often assumed in simple climate models. With the increasing spatial and temporal resolution of climate models and a growing emphasis on regional climate, the nexus between atmospheric changes and radiative energy balance perturbation must be broadened to capture new information on the role of different radiatively active components.

Atmospheric composition does not only affect climate via the radiation balance and temperature. Precipitation can be affected directly by the influence of aerosols 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.

Climate and other Earth System changes also feed back to atmospheric chemistry. Some scenarios of the Intergovernmental Panel on Climate Change (IPCC) indicate that rising atmospheric temperatures and water vapour concentrations could alter methane and ozone concentration trends by up to 25–50% during this century (Johnson et al., 2001; Stevenson et al., 2000). Natural emission and deposition rates of many 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 nitrogen oxides, suggesting another potential direct linkage between climate change and atmospheric chemistry (Price and Rind, 1992; Thakur et al., 1999; Boersma et al., 2005; Ridley et al., 2005). An important challenge for the next decade is to more comprehensively identify and describe the processes linking atmospheric change to climate change.

Figure 7. Annual average NO$_2$ (10^{15} molecules per cm^2) for 2005 as retrieved by the satellite-based SCIAMACHY instrument. Regions of high NO$_2$ correspond to pollutant emissions (North America, Europe, India, Asia, southeast Africa) and to biomass burning (Brazil, central Africa, Indonesia and northern Australia). Courtesy of Andreas Richter, University of Bremen; data described in Richter et al. (2005).
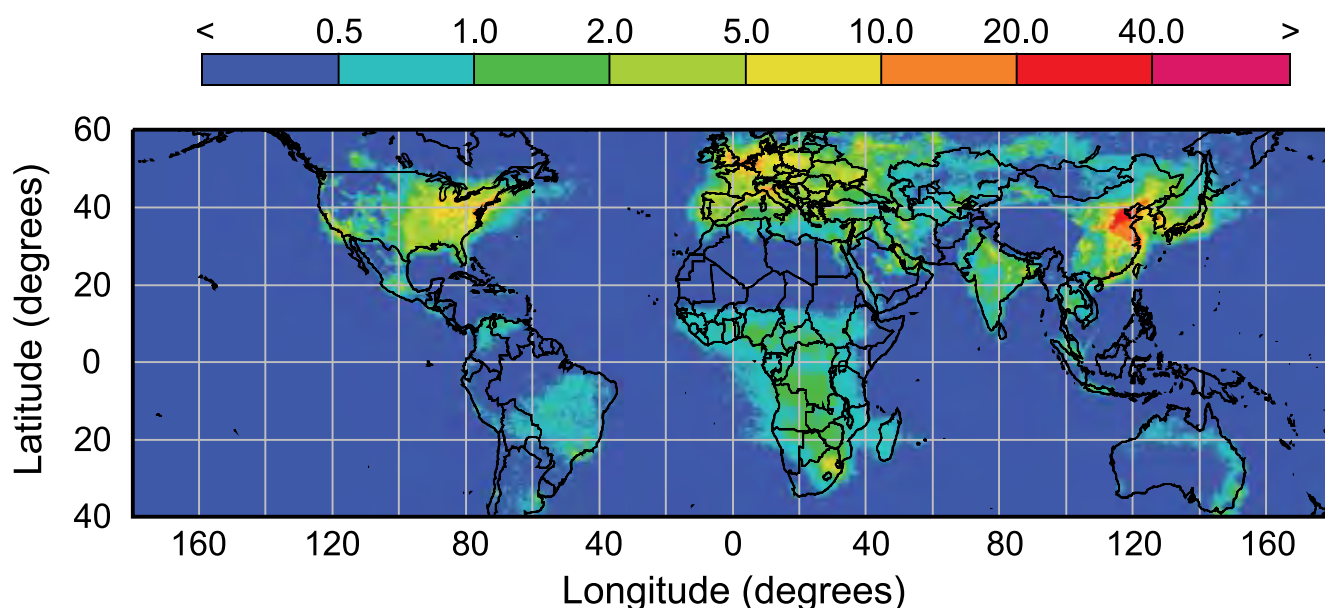
IGAC activities under Focus 1.4 will undertake one or more of the following: (i) synthesise results of other IGAC activities that address specific atmospheric constituents (such as ozone) or chemical constituent classes (such as aerosols); (ii) link the atmospheric chemistry community to other research communities, including those involved in atmospheric radiation physics, satellite measurements, climate modelling and policy tool development; and (iii) address aspects of atmospheric composition evolution over time.

While not all Focus 1.4 activities will have all these features, overall, Focus 1.4 should provide a more comprehensive and useable assessment of how atmospheric composition changes have affected, and will affect, climate at regional and global scales.

Focus 1.4 will include activities within the atmospheric chemistry research community and collaborative activities with other disciplines – some of which may develop new linkages. New methods should be developed and validated, including methods for remote sensing of atmospheric composition (recognising the forward-model versus inverse-model relationships between inferring radiation fluxes and inferring atmospheric composition), and methods for spatial and temporal extension of observations of short-lived radiatively active components (such as tropospheric ozone) for use in climate model validation.

Validations of models used for regional and global-scale atmospheric chemistry projections are required, since models disagree on the effect of future emissions; this will require targeted collaboration between field scientists and modellers. Some Focus 1.4 activities should combine measurements of the radiation balance and of concentrations of a comprehensive range of radiatively active components in order to investigate the additivity of individual radiative perturbations.

Activities within Focus 1.4 should improve the understanding of trends in atmospheric oxidation rates, thus enabling more robust analysis of future emissions scenarios using atmospheric chemistry models. For example, recent methane growth rates are close to zero and carbon monoxide appears to be decreasing, raising the question as to whether this is consistent with model projections that suggest both are currently increasing. Similarly, studies are required to improve the understanding of the indirect effects of methane and other gaseous emissions on radiative properties of the atmosphere, due to consequent changes in product components.

Focus 1.4 will include investigations of the applicability of concepts such as the global warming potential (GWP) for assessing the effects of short-lived components such as carbon monoxide and nitrogen oxides, using analyses and observations of regional emissions, concentrations

and radiative balance. The policy framework developing in response to the UNFCCC embodies concepts such as GWP that currently do not consider ozone precursors and aerosols. Focus 1.4 will also investigate the effect of urban and biomass-burning plumes (e.g. Figure 8) – which are sub-grid scale in most climate and chemical tracer models – again by combining measurements of emissions, concentrations and radiative balance.

Investigations of the potential role of mega-cities on regional climate due to the net influence of their emissions on atmospheric radiative properties are required. Similar studies could be conducted to consider the net effect of an economic sector such as pastoral agriculture. IPCC showed that the net effect of aviation emissions was very different from that which would have been estimated based on GWP-weighting of direct greenhouse gases. Studies are also required to improve the understanding of feedbacks between climate change and atmospheric chemistry – such as changes in lightning frequency and distribution – and to validate model-derived estimates of these feedbacks.

Focus 1.4 issues are all being considered – to a greater or lesser degree – by other research programmes, however, IGAC will add value by fostering coordination and collaboration. This coordination may occur within ESSP, but in Focus 1.4 fundamental atmospheric chemistry is required, and hence direct involvement of the atmospheric chemistry research community is essential. IGAC will identify appropriate linkages within ESSP, identify potential contributors from the atmospheric chemistry research community, and provide scientific leadership and coordination. Indicators of success for Focus 1.4 include: (i) more robust validation of climate models; (ii) more accurate climate projections; and (iii) more comprehensive policy tools for managing emissions.

Figure 8. Smoke plume from lightning-ignited fires in southern Oregon (United States) July 2002, which increased albedo over land but decreased albedo over clouds. Ship-tracks are also visible in the otherwise clear areas between the marine stratus deck. Courtesy of J Allen and the NASA MODIS Rapid Response Team.
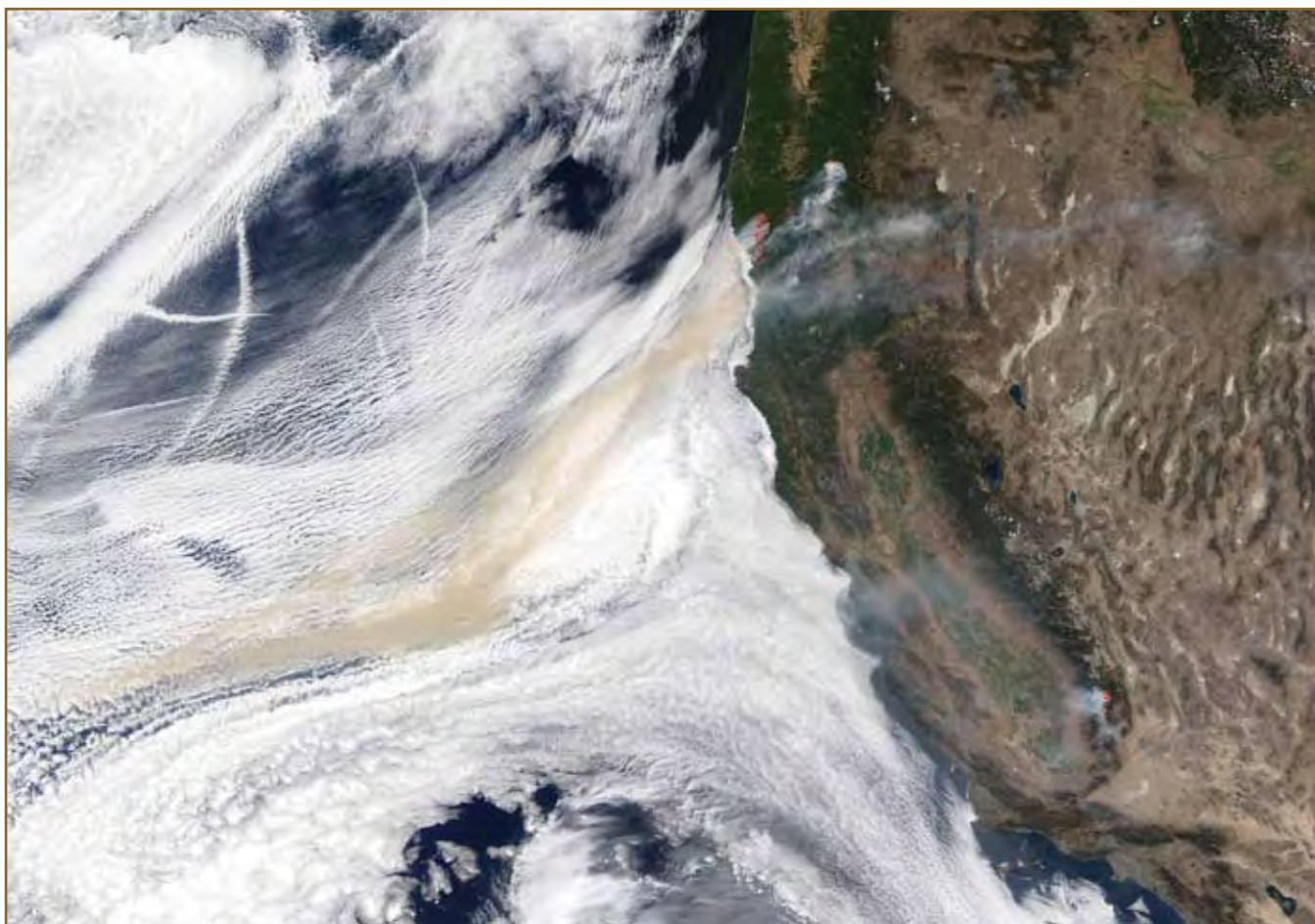
Theme 2 Effects of Emissions/Depositions, Transport and Chemical Transformations on Air Quality and Tropospheric Composition

Research in the early 2000s showed that even short-lived substances (such as aerosols and ozone) can be transported between continents (e.g. Bertschi et al., 2004; Gong et al., 2003; Huntrieser et al., 2005). For example, in early April 2001 strong winds swept across the Taklimakan Desert (western China), the Gobi Desert (eastern Mongolia) and industrial regions of eastern Asia, lifting dust and pollutants into the troposphere (e.g. Figure 9).

IGAC will study such intercontinental transport, the chemical transformation of short-lived components, and their effects on regional climate and air-quality thousands of kilometres downwind. Research to address these issues is being organised into three foci: (i) exports, transport and transformation; (ii) effects on air quality; and (iii) effects on the oxidising capacity of the atmosphere.
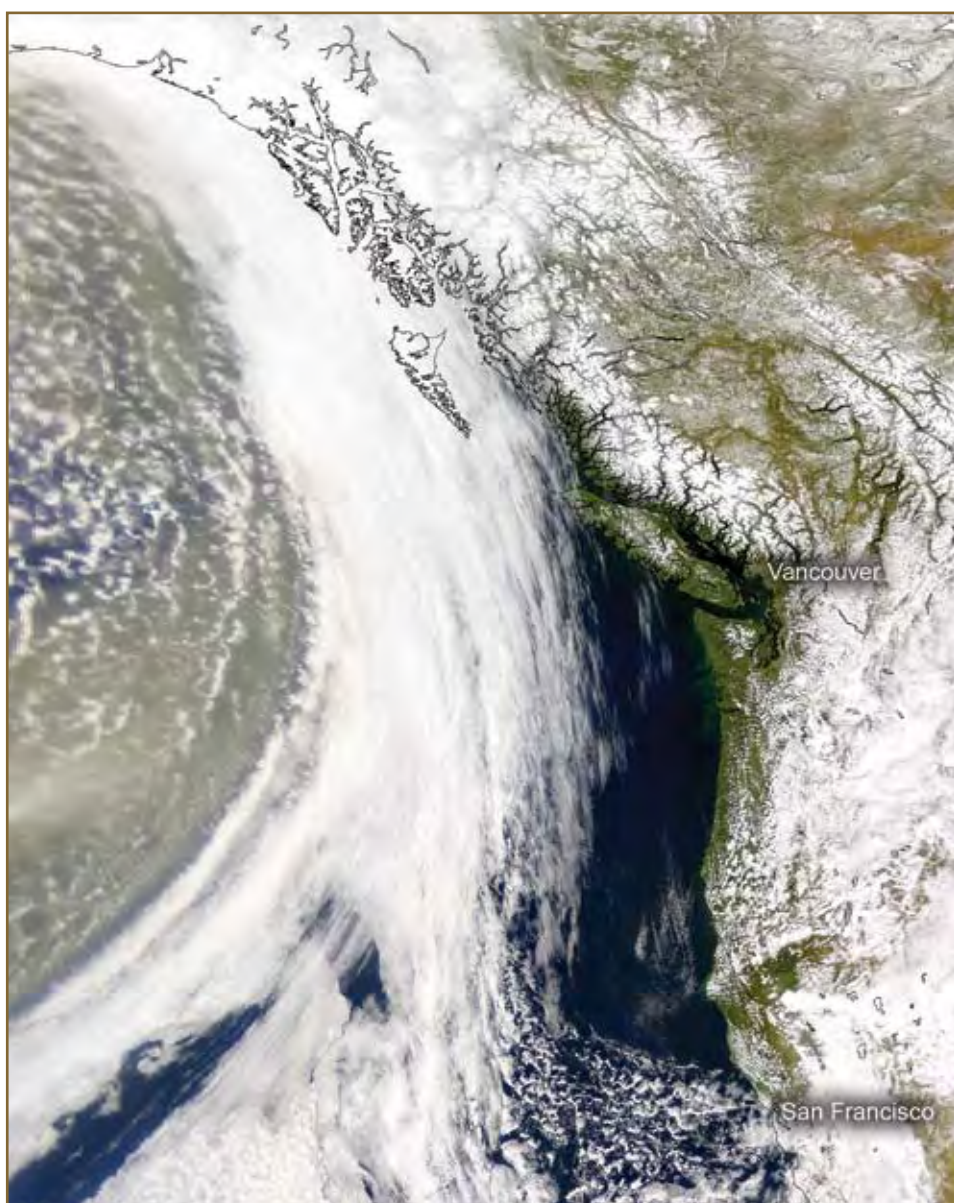
Figure 9. SeaWiFS image (11 April 2001) of dust embedded in a frontal system approaching the west coast of the United States. Originating in northern China as part of a particularly severe dust storm event, this plume was measured by the ACE-Asia array of platforms as well as by scientists across the United States as far east as New England. Image courtesy of SeaWiFS Project at NASA/GSFC and ORBIMAGE Inc.

Focus 2.1: Export, Transport and Transformation of Oxidants, Aerosols and Aerosol Precursors

Research during the first phase of IGAC contributed greatly to understanding and assessing the effects of anthropogenic emissions on the background atmosphere, and elucidated some transport processes that control the long-range transport of short-lived chemical compounds. The more important recent scientific findings – which provide the basis for future research – are summarised in the paragraphs below.

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 in summer 1,500 km downwind from the North American source region (Huntrieser et al., 2005; Li et al., 2002). This pollution has been shown to enhance spring ozone levels in the central North Atlantic (Chandra et al., 2004). These observations show that the summer flux of anthropogenic ozone from North America to the North Atlantic is 1.0–1.6 Gmol day^{-1}, which exceeds the natural ozone flux from the stratosphere (Li et al., 2002).

Nitrogen oxides and volatile organic compounds (VOC) play an important role in the ozone budget of the North Atlantic troposphere. In the remote marine troposphere, the concentrations of carbon monoxide and methane are sufficient to support significant photochemical ozone formation (Honrath et al., 2004). Whether this photochemistry produces or destroys ozone is determined by the availability of nitrogen oxides. Analysis of the correlation of odd reactive nitrogen with carbon monoxide has demonstrated that only a small fraction of the nitrogen oxides emitted in the continental boundary layer is transported to the free troposphere or to the marine boundary layer (Parrish et al., 2004a; Li et al., 2004). Although transport is limited, model results suggest that eventual ozone production in the global troposphere from North American emissions is about twice as large as the direct export of ozone from the North American boundary layer. The nitrogen oxides delivered to the lower troposphere in the western North Atlantic during summer are sufficient to produce, on average, 1–4 ppbv day^{-1} of ozone throughout the region (Liang et al., 1998). In contrast, the central North Atlantic (e.g. the Azores) is generally a region of photochemical ozone destruction (Parrish et al., 1998), although periodic intrusions of North American or European pollution can lead to ozone production (Honrath et al., 2004).

Investigations of the long-range transport of atmospheric trace components over the northwest Pacific Ocean have shown a region of maximum outflow between about 20–40° N. The delivery of nitrogen oxides from the Asian continent to the free troposphere, and their subsequent redistribution by large-scale dynamics, influences ozone production over large areas of the Pacific Basin (Parrish et al., 2004b; Price et al., 2004). Measurements of atmospheric chemical composition over the western Pacific indicate that throughout the well-mixed troposphere, sulphur dioxide and sulphate in aerosols were largely associated with sulphur dioxide emissions from Asian continental sources (Koike et al., 2003).

Meteorological measurements show that during summer, inversions can effectively isolate the marine boundary layer from the lower free troposphere, where the majority of the pollutants are transported in highly stratified layers (Prados et al., 1999). The processes that form these layers provide an effective mechanism for the transport of North American continental pollution into the mid- and upper-troposphere over the North Atlantic. The primary direction for summer transport of North American pollution to the North Atlantic is north-eastward. Warm sector flow ahead of advancing cold fronts is the most important process for the transport of pollution from the urbanised east coast of the United States to the North Atlantic (Cooper et al., 2001, 2002a, b; Hess, 2005); this rapidly and effectively transports large amounts of relatively short-lived pollution over long distances (e.g. Figure 10).

Figure 10. Ensemble of trajectories visualising "warm conveyor belt" (WCB) transport of ozone from the polluted North American boundary layer to Europe: (a) horizontal projections and (b) time-height profiles. Trajectories began from a dense horizontal grid on 26 May 1997 at 1800 UTC but only those ascending by at least 8000 m are shown. The red line indicates the flight path of a MOZAIC aircraft, which encountered the ozone pollution plume contained in the warm conveyor belt and measured ~100ppb ozone. From Stohl and Trickl (1999).
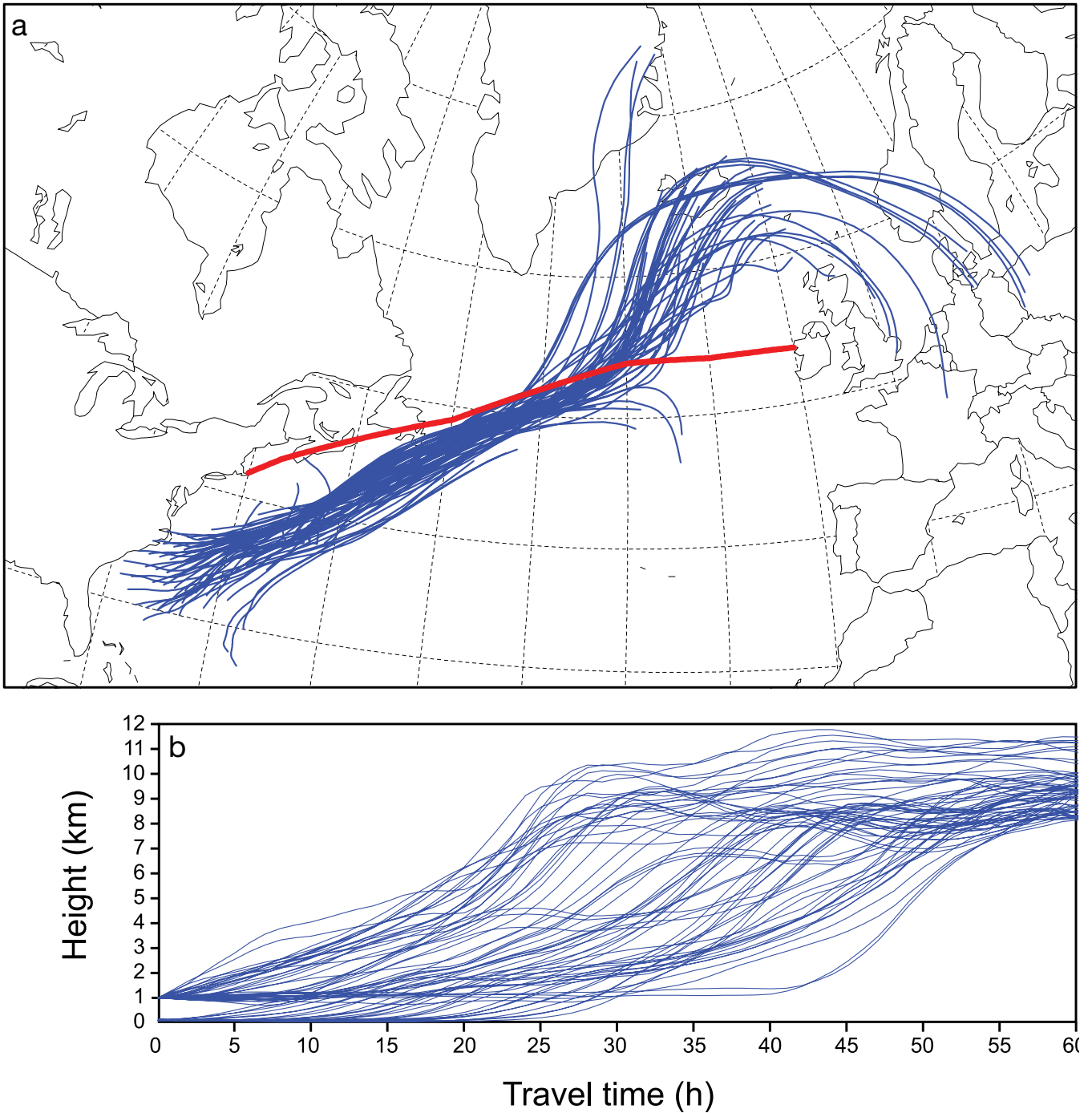
Recent studies of the sources of nitrogen oxides over the North Atlantic have shown that aircraft and lightning emissions in the free troposphere and transport from the surface, help determine the distribution of nitrogen oxides in the free troposphere and the consequent formation of ozone (Meijer et al., 2000). These sources are found to vary with latitude, with aircraft emissions relatively more important at high latitudes, and lightning relatively more important at low latitudes.

The principal sources of carbon monoxide in the Northern Hemisphere are fossil fuel combustion, biomass burning, and oxidation of methane and non-methane hydrocarbons (Bergamaschi et al., 2000). Recent research has elucidated the importance of boreal forest fires to background carbon monoxide concentrations in the Northern Hemisphere during the summer (Yurganov et al., 2004, 2005; Wotawa et al., 2001). Background concentrations in the Northern Hemisphere declined from the late 1980s through the turn of the century, due to carbon monoxide emission controls in the industrial countries of the Northern Hemisphere. This trend has been perturbed by large carbon monoxide emissions associated with wild fires in the boreal regions of North America and Russia (Novelli et al., 2003). The emission of nitrogen oxides, combustion-produced VOC and carbon aerosols from these fires, may also significantly perturb Northern Hemisphere atmospheric chemistry. The variability of emissions due to these forest fires parallels climate variability that has led to drought conditions in boreal regions (e.g. Figure 11).

Studies of summer conditions in the western North Atlantic show a positive correlation between carbon monoxide and ozone, indicating that anthropogenic pollution produces ozone during the summer. However, in the winter, carbon monoxide and ozone are negatively correlated – both at surface sites and in the free troposphere (Parrish et al., 2000). This negative correlation indicates that anthropogenic pollution provides a winter sink for ozone – consistent with the expected reaction of ozone with primary pollutant emissions of nitrogen oxide and unsaturated VOC. This destruction probably occurs in all seasons, but in summer the photochemical formation of ozone more than compensates. It has not yet been determined whether the overall anthropogenic effect on ozone during winter is negative further from the continent. In winter, slow photochemical ozone production probably occurs over longer transport times and distances in the troposphere. Long-range transport of anthropogenic ozone precursors to low latitudes with more photochemical activity, could possibly further enhance ozone formation in the remote marine troposphere. A combination of these processes may compensate for initial ozone destruction.

Given the understanding of atmospheric chemistry and transport summarised above, it is now recognised that intercontinental transport of anthropogenic emissions – and of their chemical products – has significant effects not only on background atmospheric chemical composition, but also on regional near-surface chemical composition and deposition fluxes. In the mid-latitude Northern Hemisphere, emissions from fossil-fuel combustion in eastern North America, western Europe and eastern Asia, affect air quality in downwind 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 central components of IGAC research.

Research Agenda

Progress in Focus 2.1 will require (i) intensive field studies to investigate specific processes; (ii) long-term observations to place these processes in seasonal and multi-year contexts; and (iii) assessments directed at policy development.

Intensive field studies will need to be designed to answer a limited set of well-defined questions. These studies will involve multiple platforms (in particular one or more research aircraft) and will both utilise and support the development of three-dimensional chemical transport models. To optimise the value of the measurements for testing models, it will be useful to operate the models in forecast mode during the field study, and use the forecasts to guide day-to-day flight planning. This approach was used successfully in INDOEX, ACE-Asia and the International Consortium for Atmospheric Research on Transport and Transformation campaigns. Integration of satellite observations into experimental designs may also help place limited aircraft observations in a broader spatial context. Developing this synergy between aircraft and satellite observations may require satellite validation flights in aircraft mission plans.

Long-term observations are essential to address ITCT questions by extending information from intensive field studies to seasonal and multi-year records. Long-term measurement platforms may include ground sites, ships, commercial aircraft and small chartered aircraft. Satellites have been deployed to produce valuable long-term observations, but they have important limitations. The selection of platforms should be made with careful consideration of three-dimensional model results in order to provide the best test of models' abilities to answer key questions. Eventually, the long-term measurement programme may evolve into a monitoring operation to document changes in anthropo-

genic emissions from continental source regions and their implications for global atmospheric chemistry.

Periodic assessments should be made of research implications in terms of the effects of projected changes in emissions and other forcing variables (e.g. land use or climate change) for intercontinental pollutant transport. Focus 2.1 will foster close interaction between the international science community and the policy communities of large industrial countries. We expect that these policy communities will help identify the highest priority assessments, so as to direct scientific effort toward the most critical uncertainties.

Figure 11. Calculated boundary-layer carbon monoxide concentrations from anthropogenic emissions (a) and forest fires (b) for 1 July 1995. Forest fire locations are marked with black squares. Anthropogenic concentrations are high ahead of a cold front which parallels the east coast; forest fire concentrations are high behind the cold front. From Wotawa and Trainer (2000).
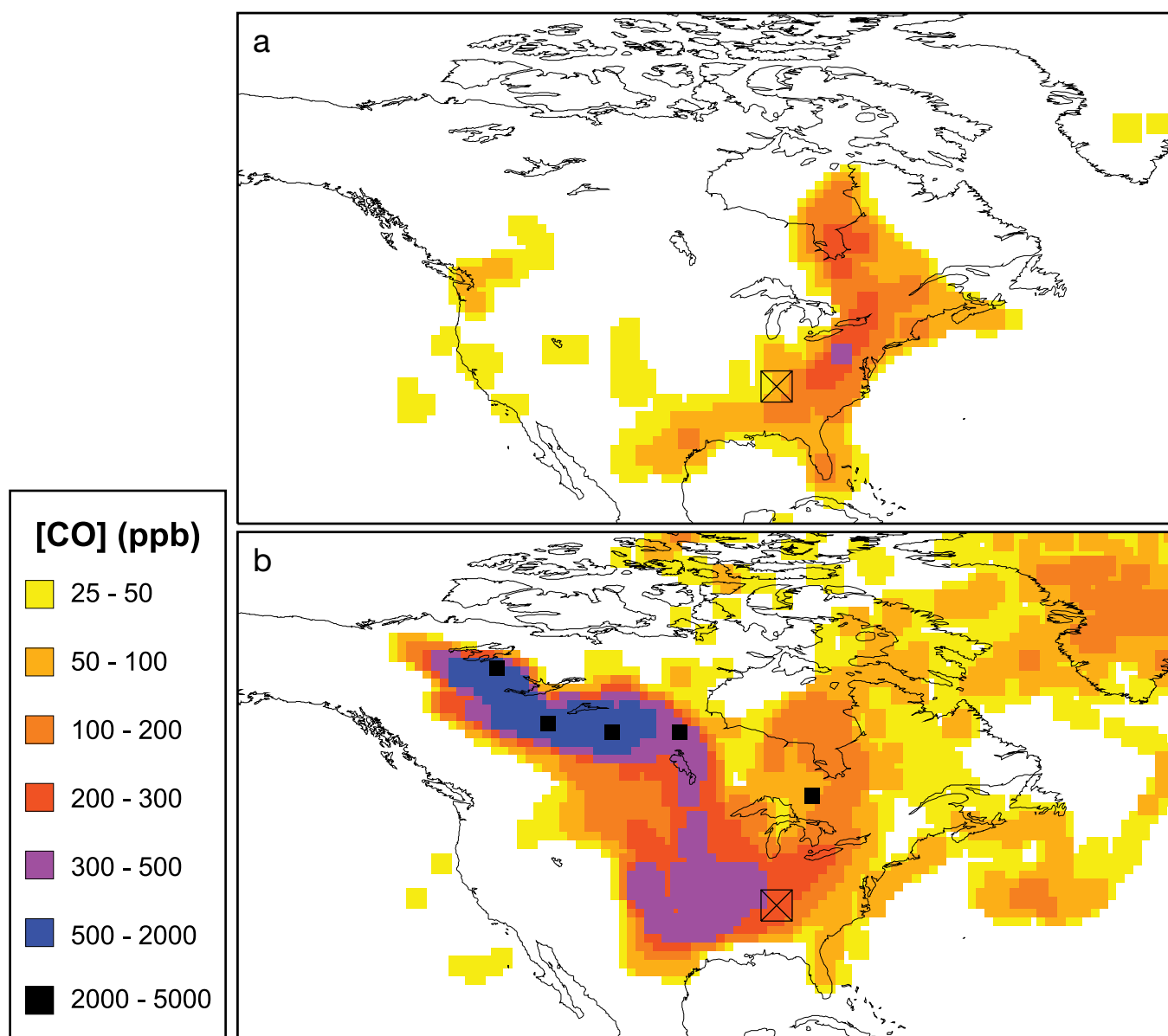
Research Tools

Focus 2.1 will employ three-dimensional models and measurements from ground sites, aircraft and satellites. Three-dimensional models will be key to developing a quantitative understanding of regional- to global-scale impacts of local emissions. To do so, they must resolve the coupling between transport, chemistry and aerosol processes on synoptic scales and finer. Such models exist, but their results are highly uncertain and they need to be far more deliberately constrained and tested with atmospheric observations. IGAC will promote a close coupling of atmospheric observations and models, in which models are driven by assimilated meteorological observations for the period of interest, and observations are targeted towards testing relevant model features.

Measurements from ground sites, aircraft and satellites will all be important in both model testing and in providing a wider base of exploratory measurements. Ship platforms are not discussed in detail, but share some of the advantages and disadvantages of ground sites and aircraft platforms.

Ground sites have two primary strengths. Firstly, they enable simultaneous measurement of a broad spectrum of gaseous and aerosol constituents by prototype, research-grade instrumentation. They therefore expand the observational database that allows the identification and investigation of crucial atmospheric processes. Many components can be measured by two or more techniques, enabling instrument comparisons that are crucial for the development and improvement of measurement techniques. Secondly, long-term observations from ground sites are relatively cheap, thus allowing measurement of many components under well-calibrated conditions. A disadvantage is the lack of spatial information – both vertically and horizontally – which is crucial for interpreting the long-range transport and chemical evolution of air masses. Light Detection and Ranging (LiDAR) instruments and balloon sondes do of course provide vertical information, but currently only for ozone, aerosols and water vapour. The principal roles of ground-based measurements in this context are to (i) expand the observational database of simultaneous measurements for a broad spectrum of atmospheric components; (ii) provide high-quality data on seasonal and multi-year trends; (iii) identify correlations between components that may provide important model constraints; and (iv) extend in time the information gained from intensive field studies.

Aircraft have the distinct advantage of allowing a broad range of high quality measurements to be made over large areas. By using chemical model forecasts to select flight paths, observations can be collected in an optimal manner for model testing. The disadvantages of aircraft-based observations are the limited duration of flights and limited instrumentation due to aircraft payload restrictions. The time limitation can be partially overcome by regular observations from commercial aircraft or from small chartered aircraft. However, in these cases there are even more severe limits on instrumentation and hence observational flexibility.

Satellite-based measurements can provide global and continuous observations of tropospheric composition, and hence are ideally suited for characterising intercontinental transport. The trace gases which can be observed from space are limited, but include several of great significance – ozone, nitrogen dioxide, water vapour, carbon monoxide, formaldehyde, carbon dioxide and methane. Radiative transfer theory indicates that passive remote sounding of trace gases has limited vertical resolution in the troposphere, with the best resolution being achieved by combining simultaneous remote measurements of gases in different spectral regions. The data from several missions – for example, the ESA ENVISAT satellite and the NASA Aura satellite – will extend the passive remote sensing capability within the troposphere. Space-based LiDAR has already been demonstrated as being able to provide high vertical resolution for aerosol and cloud data products. Several imminent and planned satellite missions will further enhance this capability (Spinhirne et al., 2005; Winker et al., 2004).

Finally, it is well recognised that to be of optimal value, remote sensing of the troposphere requires high spatial and temporal resolution. Currently, tropospheric composition can be determined only from low orbits and from a limited number of platforms. Proposed measurements from a set of geostationary platforms would provide the most relevant data for investigating ITCT of atmospheric species.

Focus 2.2: Impacts of Intercontinental Transport on Surface Air Quality

Focus 2.2 will concentrate on atmospheric chemistry studies that have a direct bearing on human welfare and environmental quality. Some of the most highly polluted regions of the globe are also the most populated, with clear consequences for public health; mega-cities in particular are prone to severe pollution events (Figure 12). A detailed assessment of the environmental and human health impacts of regional air pollution is beyond the scope of IGAC; however, IGAC will relate large-scale pollution distribution and transport to appropriate human health and environmental impact metrics. Focus 2.2 will build on the description of the background atmosphere from the first phase of IGAC to provide the scientific underpinnings for formulating effective regional and national air quality management policies.
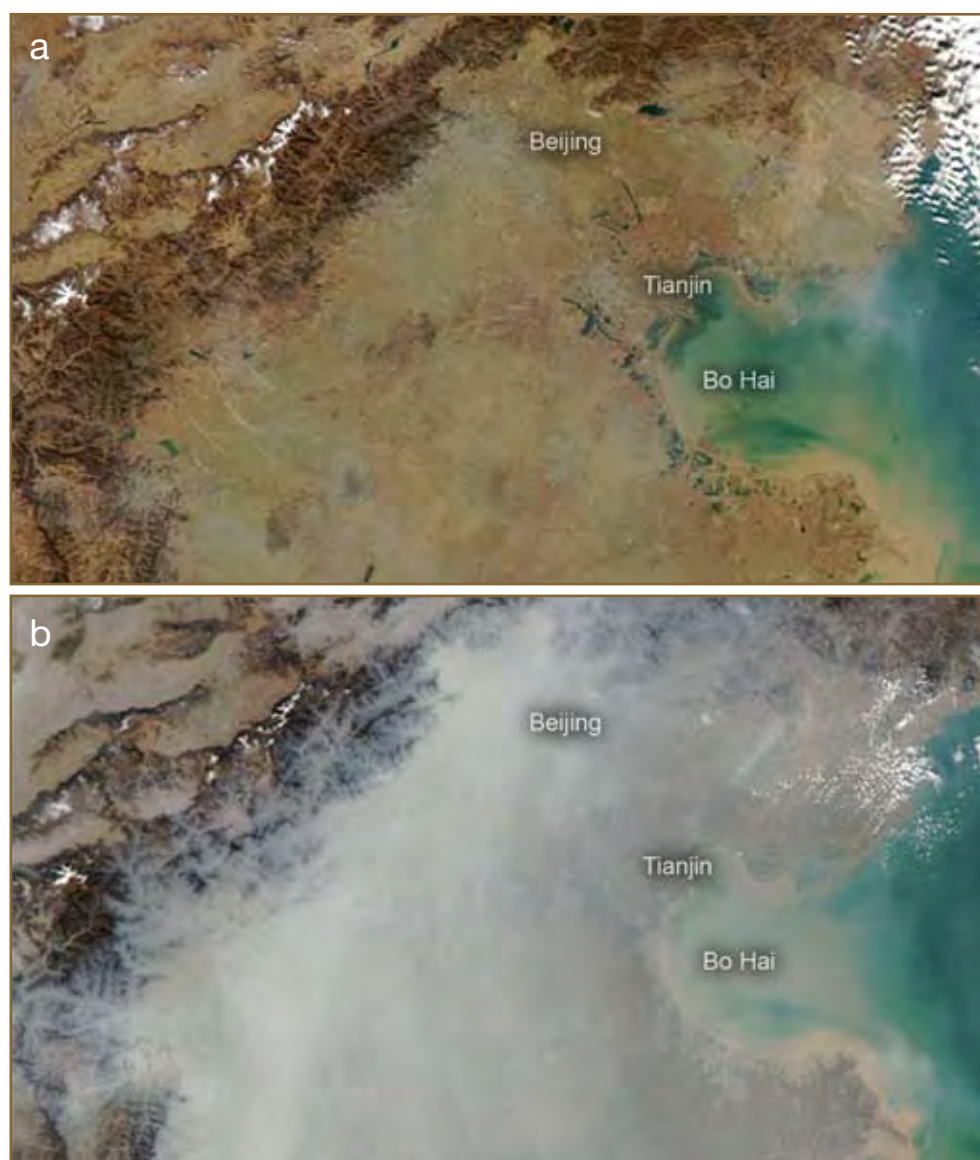
Figure 12. Beijing region on 2 November 2005 (a) and 3 November 2005 (b). Differences were induced by a change in meteorology that trapped pollution in the flatlands around the city. The pollution gathered overnight and lingered until 5 November; by 6 November a new haze cloud had accumulated. True-colour images from the Moderate Resolution Imaging Spectro-radiometer (MODIS) instrument aboard the NASA Aqua satellite. Courtesy of MODIS Land Rapid Response Team.

The impacts of long-range transport of ozone, aerosols and their precursors are well established, and the long-range transport of dust from Asia (e.g. Holzer et al., 2005; VanCuren et al., 2005; Darmenova et al., 2005; Jaffe et al., 2003a; VanCuren and Cahill, 2002) and Africa (e.g. Prospero and Lamb, 2003; Reid et al., 2003; Chiapello and Moulin, 2002; Prospero, 1999; Moulin et al., 1998; Perry et al., 1997) is well documented. The concentration of ozone over the Northern Hemisphere appears to be increasing, probably due to anthropogenic influences on the chemical composition of the atmosphere (Berntsen et al., 1999; Jacob et al., 1999; Wang et al., 2006). More specifically, there is good evidence for intercontinental transport of ozone, ozone precursors and other chemical compounds. Measurements from islands in the Atlantic and Pacific have recorded events of increased ozone and carbon monoxide concentrations. Measurements during spring on the west coast of the United States have detected an Asian influence on the levels of several atmospheric components (including carbon monoxide, peroxyacetyl nitrate, ozone, nitric acid and light hydrocarbons) during periods of rapid transport from Asia (Parrish et al., 2004c; Price et al., 2004; Bertschi et al., 2004; Jaffe et al., 1999, 2003b). Model simulations reproduced the observed enhancements in carbon monoxide, peroxyacetyl nitrate and

ozone (Liang et al., 2004; Yienger, 2000). Other model calculations indicated that the average ozone concentration over the western United States would increase significantly if Asian ozone-precursor emissions were to increase significantly (Jacob et al., 1999).

Elevated ozone concentrations (attributed to long-range transport from the boundary layer over North America) were observed over Europe during a spring 1997 event (Stohl and Trickl, 1999), with concentrations as high as 100 ppbv measured in the free troposphere. These high concentrations were due to effective transport (without significant dilution of boundary layer air) by an ascending air stream at the leading edge of a trough. Since then, measurements from aircraft and ground sites together with modelling studies have shown that North American pollution regularly impacts ground-level ozone concentrations in Europe during the summer (Huntrieser et al., 2005; Stohl et al., 2004; Derwent et al., 2004; Trickl et al., 2003; Li et al., 2002).

Numerous examples of long-range transport have been investigated, but a systematic and quantitative budgeting of important trace components at hemispheric to global scale has not been attempted. In particular, there is no agreement on the relative importance of natural

Figure 13. South Asian monsoon plume – a major pathway for export and intercontinental transport – as computed by the global Model of Atmospheric Transport and Chemistry (Max Planck Institute for Chemistry version), and successfully predicted by global chemical weather forecasts. Courtesy of Mark Lawrence, Max Planck Institute for Chemistry.
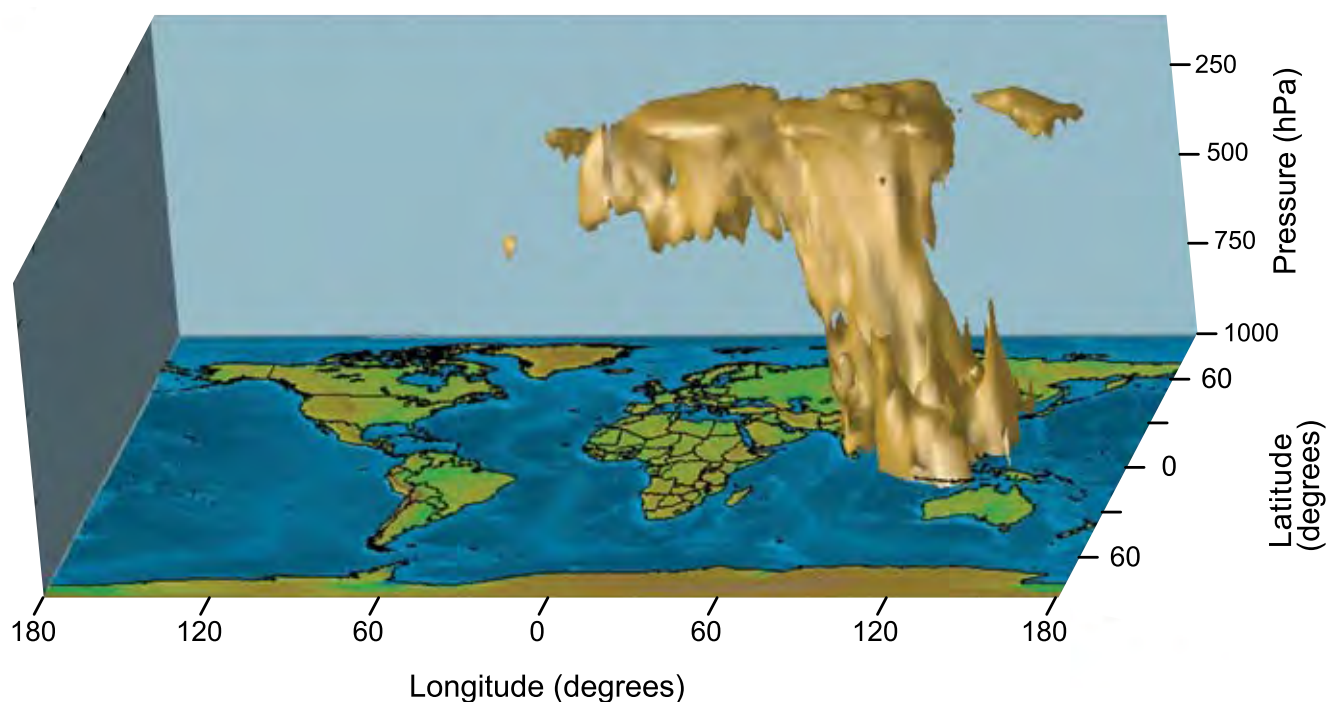
and anthropogenic sources of ozone to the Northern Hemisphere troposphere. A major task for IGAC is to undertake these quantitative analyses.

While tropospheric ozone and aerosols will continue to be a focus of IGAC research, other environmentally important components will also be a focus. For example, mercury is a priority pollutant in many international conventions (e.g. the United Nations Economic Commission for Europe Convention on Long-range Trans-boundary Air Pollution) and there is a growing awareness that a significant fraction of regional mercury deposition can originate outside the region (e.g. Travnikov, 2005; Jaffe et al., 2005; Seigneur et al., 2004; Lee et al., 2001). Formulating effective deposition control policies for mercury (and other priority pollutants) will require the development of source-receptor relationships on hemispheric and global scales. This in turn will require a better understanding of the global distribution of different mercury compounds in the atmosphere, and of the processes that control these distributions.

Research Approaches

Recently, global chemical weather forecasts have begun to be computed using chemical transport models driven by meteorological forecasts; the chemical weather is the short-term state of the atmosphere in terms of the time-dependent distributions of trace gases and aerosols (e.g. Figure 13). These forecasts have been used extensively in the day-to-day flight planning for campaigns such as INDOEX, Transport and Chemical Evolution Over the Pacific and ACE-Asia; however, chemical weather forecasts also have potential health and agricultural applications. Global chemical weather forecasting is expected to become a major focus in atmospheric chemistry in the next decade, as it integrates improved emissions inventories, long-range transport, chemical transformations, deposition and assimilation of observations (especially satellite-based). The atmospheric chemistry community may profit from working in a more "operational" mode (like the meteorological community), and working towards measurable improvements in community-wide products. Careful coupling of global- and regional-scale forecasts will be required. Although there is already considerable experience with urban ozone forecasts, strong global interconnections indicate the need to embed regional models in a global forecasting framework.

Emissions from one region and ambient pollution levels in another are linked via atmospheric processes such as mixing, transport, chemical transformation and deposition. Forecasting – whether parametric or deterministic – of ambient pollution levels requires an understanding of how these processes influence pollution formation and distribution. Reliable regional chemical forecasts require an improved understanding of the entire ensemble of salient atmospheric processes as well as reliable inventories of pertinent emissions. On this issue, IGAC will work closely with GEIA – an activity of AIMES.

The models required for global chemical weather forecasting are similar to those required in Focus 2.1, and thus a similar model evaluation process is appropriate in order to quantify forecast reliability and to identify areas requiring improvement. Models should be evaluated in terms of their ability to model emissions, meteorology and transport, chemical transformation and deposition.

Obtaining information on atmospheric chemical constituents – to initialise models and evaluate chemical forecasts – will require a similar observing system as that which supports weather forecasting. These data will also enable diagnostic evaluation of complex grid models. Observational work will be coordinated with IGOS-P.

Focus 2.3: Anthropogenic Impacts on the Cleansing Capacity of the Atmosphere

The oxidising or cleansing capacity of the atmosphere is generally defined by hydroxyl radical levels, as these determine the rate at which atmospheric pollutants, greenhouse gases and short-lived replacement hydro-chlorofluorocarbons are removed from the atmosphere (Ehhalt, 1999; Prinn, 2003). To a large extent the oxidising capacity is controlled by concentrations of ozone, water vapour, ultra-violet radiation and trace gas levels. Due to the extremely high reactivity of the hydroxyl radical and its rapid cycling with hydrogen and hydroperoxyl radicals, its concentrations are very low and highly variable. Only since the mid-1990s has it been pos-

sible to measure hydroxyl radical concentrations directly with any degree of accuracy. Even so, because of their very high temporal and spatial variability it is still not possible to measure concentrations on a global scale so as to directly determine global concentration trends. The large-scale concentrations and trends can, however, be inferred indirectly from long-term global measurements and emission estimates of methyl chloroform (Figure 14), since the hydroxyl radical is the major destruction mechanism for this chemical (Prinn et al., 1992, 2001; Krol and Lelieveld, 2003; Montzka et al., 2000).

Figure 14. (a) One-, (b) two- and (c) three-year weighted-average estimated hydroxyl radical concentrations with one standard deviation error boxes inferred from methyl chloroform concentrations. These absolute concentrations, but not the relative hydroxyl radical variations, depend on the weighting and the model used in the calculation. Also shown are a polynomial fit to the one-year weighted averages and the results when different reference emissions are used (dotted and dashed lines). From Prinn et al. (2005).
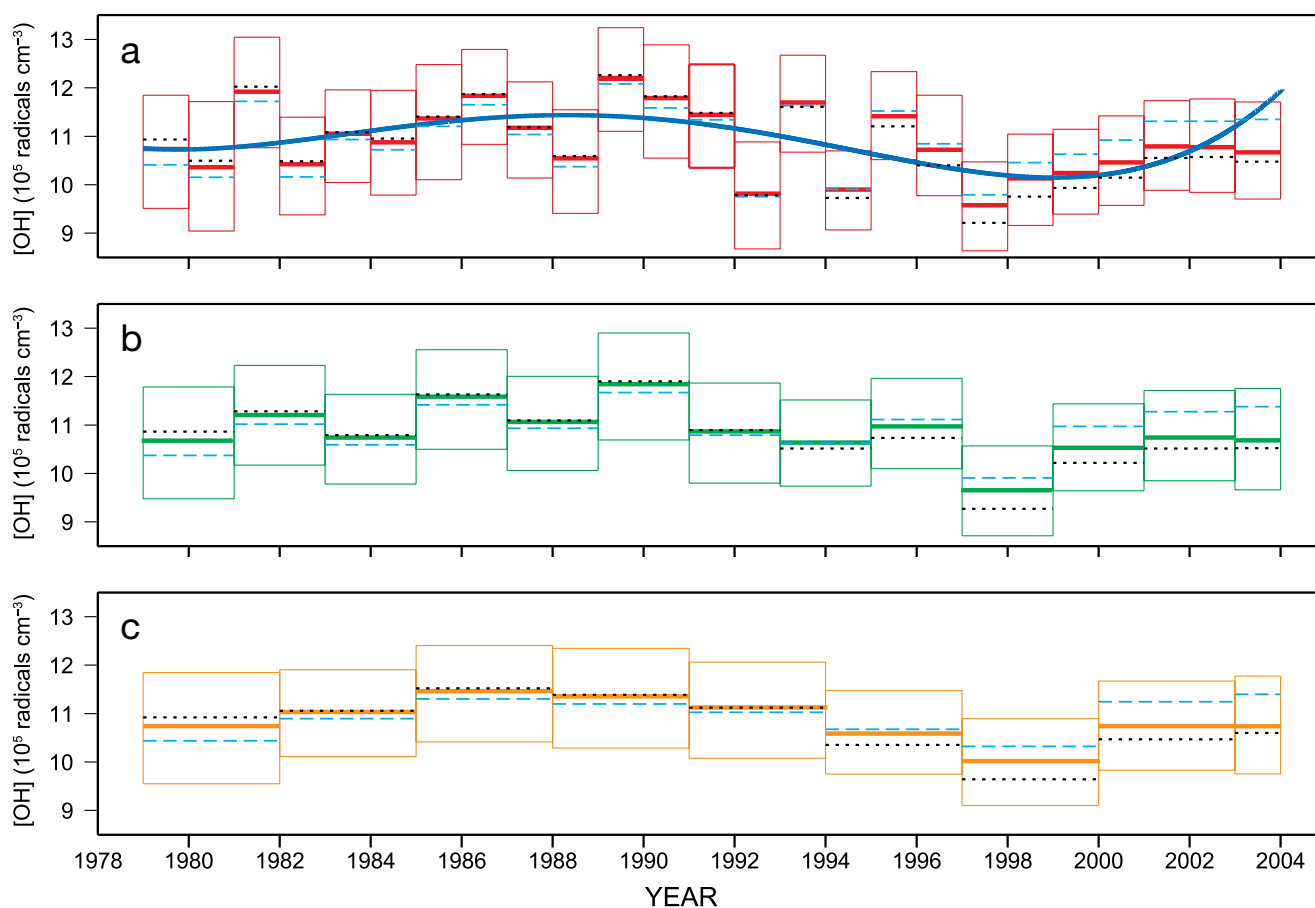
Concentration changes over the last two decades have been determined from measurements of tracer gases (such as methyl chloroform, chlorofluoromethane and carbon-14 labelled carbon monoxide), while concentration changes since pre-industrial times have been estimated using global chemistry transport models. Measurements of the tracer gas methyl chloroform indicate that the global weighted average concentration of the hydroxyl radical in the troposphere is $\sim10^6$ molecules cm^{-3} (Prinn et al., 2001; Krol and Lelieveld, 2003). More recently, Bousquet et al. (2005) used an inverse method with a three-dimensional model together with methyl chloroform measurements to reveal substantial year-to-year variations in global average concentrations between 1980 and 2000. In general, global models estimate a decrease of around ten percent over this period, with results sensitive to assumed changes in emissions of nitrogen oxides, carbon monoxide, methane and hydrocarbons. This contrasts with recent estimates based on the analysis of tracer gas measurements with inverse modelling techniques, which suggest no long-term trend over the past 25 years (Prinn et al., 2005; Manning et al., 2005). One difference between these studies is that stratospheric ozone depletion was not considered in most of the modelling studies. Stratospheric ozone depletion increases tropospheric ultra-violet radiation and hence increases hydroxyl radical concentrations. This mechanism may at least partially explain the observed changes in methane and carbon dioxide growth rates over the last twenty years. It is therefore still very unclear how hydroxyl radical levels changed in the past, and it is unclear how they will change in the future in response to changing anthropogenic emissions and climate change.

Research Approaches

Three approaches are required to assess change in the oxidising capacity of the atmosphere. Firstly, global chemical models must be based on better emissions inventories and be evaluated with global data. Secondly, *in situ* and satellite-based measurements of key atmospheric components should be compared with palaeo data from ice cores describing the past. And thirdly, coupled chemistry-climate models are needed to simulate past and present oxidising capacity, and to simulate future chemical forcing, climate response and chemistry-climate feedbacks.

Improved emission inventories to reduce the uncertainties in global chemical models should focus on ozone precursors, for which data are poor – particularly with respect to characterising temporal variation. A better understanding of chemistry-climate interactions would also improve model estimates. For example, it is currently unclear how nitrogen

oxides produced by lightning respond to changes in convection. Other knowledge gaps include understanding how terrestrial ecosystem changes directly and indirectly affect the oxidising capacity of the atmosphere, and understanding how these atmospheric changes feed back to ecosystems. On this latter issue IGAC will collaborate closely with iLEAPS. Once again, reducing model uncertainties will require continued evaluation of all aspects of model performance by comparisons with data and model intercomparison.

Observational requirements include, in particular, continued long-term monitoring of trace gases such as halocarbons, carbon monoxide, methane and ozone. New monitoring stations are required in data-sparse regions, and measurements are required of key trace gases for which trends are extremely uncertain, such as nitrogen oxides and VOC. Long-term measurements are required in the free troposphere, and this will be addressed to a certain extent by new satellite-derived data. Measurements from commercial aircraft are one option – for example, the MOZAIC ozone measurements of 1994–2002 – but the development of new sonde instrumentation should also be encouraged. Dedicated observational campaigns are necessary in areas for which current understanding is poor and which are sensitive to climate change – for example west Africa and Eurasia. Measurements of a variety of chemical components (including hydrogen and the hydroperoxyl radical) have been shown to provide stringent tests of tropospheric photochemistry understanding, while tracer measurements allow evaluation and improvement of model transport schemes. In evaluating chemistry-transport and chemistry-climate models and possible oxidising changes, historical chemical data – for example, methane in ice cores – provide useful constraints. Further extraction and measurement of gases related to ozone, the hydroxyl radical and other oxidants is envisioned.

Combining new modelling techniques – such as data assimilation and inverse modelling – with new observations will provide new insights into past, present and future changes in the oxidising capacity of the atmosphere. As well as quantifying changes in regional anthropogenic emissions, Earth System models with chemistry-climate links should be used to study links between anthropogenic climate change and natural changes in emissions, atmospheric dynamics and the water vapour cycle. In is expected that over the coming decade chemistry-climate model development and application in Earth System studies will lead to an improved understanding of the oxidising capacity of the atmosphere and how it is changing.

Implementation Strategy

The IGAC scientific agenda requires a global implementation strategy within an Earth System framework, such that activities are targeted to allow assessment of Earth System responses and feedbacks. IGAC research will be implemented by Tasks, Workshops and focused Initiatives. These three mechanisms aim to achieve a balance between bottom-up and top-down activities. IGAC will also add value by synthesising new or existing activities in atmospheric chemistry to help resolve problems of global dimensions and importance. A specific focus will be on coordination of national projects with international efforts, thereby providing a synergy that produces information and understanding beyond the level that could be achieved by single nationally-driven efforts.

IGAC Tasks are activities that address specific scientific questions, require an international research framework and promote wide participation. Tasks will include field campaigns, modelling intercomparisons, measurement intercomparisons, measurement networks and international laboratory studies. Task proposals may be prepared by researchers within the IGAC community or may emerge from IGAC workshops. Task proposals should not exceed eight pages and may be submitted at any time to the International Project Office (IPO). Proposals should outline (i) the specific research questions to be addressed that will contribute to fulfilling the Science Plan above; (ii) the research timetable (normally over 3–5 years) and completion date; (iii) a quality assurance and data plan (that includes free access to task data); (iv) educational and capacity building efforts; (v) expected peer-reviewed papers in, or special issues of, targeted journals; and (vi) the names and contact details for the Task coordinators.

Proposals will be circulated by the IPO to the IGAC Scientific Steering Committee (SSC) for review and approval. The SSC will assign an SSC member as a contact point for each approved Task. Tasks will be reviewed at the annual SSC meeting on the basis of summaries submitted by Task coordinators to the IPO six weeks in advance of the meeting. Task summaries should not exceed three pages and should articulate (i) scientific highlights for the year; (ii) Task products in line with the approved proposal (with copies appended); and (iii) proposed changes to the Task.

The SSC will serve as an international advisory board for IGAC, and will advocate for national and international funding of IGAC Tasks. The SCC will help IGAC interact with other IGBP projects and other relevant international research programmes and provide a channel of communication amongst scientists in different countries. SSC members will provide project advocacy and promotion and will enlist wide international participation in the project. Each SSC member will also liaise with one relevant project or programme, including – where possible – attending the annual meeting of the project/programme.

IGAC Workshops are used to address focused issues identified by the SSC for rapid attention. Workshops may be sponsored or cosponsored in support of IGAC Tasks. Workshops will assemble leading researchers on the issue to produce a white paper, planning document or review paper, or to initiate a new Task.

IGAC Initiatives are started by the SSC, sometimes in collaboration with partner projects, to address important, current issues that require the synthesis and integration of research across disciplines or geographic boundaries, and that would not necessarily be addressed by the community without outside organisational assistance, oversight and support.

Communication

IGAC will continue to promote science communication via the project newsletter, website and open science conferences. The IGAC Newsletter (IGACtivities) was initiated in 1995 and now has a distribution of almost 4,000 across one hundred countries. It communicates syntheses of IGAC science, and reports on IGAC Tasks, Workshops and Initiatives and other relevant research. The IGAC website (www.igac.noaa.gov) will continue to be a key source of project news and information, and a repository for project publications.

Communication within the IGAC community, the generation of new research ideas, and the engagement of young and developing country scientists are all fostered

by the bi-annual IGAC open science conferences. The IPO will continue to raise funds specifically to support attendance at the meeting. In alternate years, the IGAC conference will be held jointly with CACGP.

Project Offices

IGAC operations will be coordinated by the IPO and supporting regional offices. Initially, the IPO will be based at the Pacific Marine Environmental Laboratory of the US National Oceanic and Atmospheric Administration (Seattle, United States). The IPO will coordinate with IGBP and act as the Secretariat for SSC and other IGAC meetings; it will be the central communications hub for IGAC.

Regional offices will initially be at the Academia Siniaca (China-Taipei) and at the European Atmospheric Composition Change European Network of Excellence (ACCENT) office at the Institute of Atmospheric Sciences and Climate of the Italian National Research Council (Bologna, Italy). All IGAC offices will promote and facilitate international atmospheric chemistry research and help enlist wide international participation in IGAC. Additionally, they will assist the SSC to plan and facilitate new research and to collate information on, and maintain connections with, relevant national and international programmes. They will facilitate communication amongst IGAC scientists and with other global change scientists, and secure financial support for the office.

Linkages with Other Projects

IGAC will contribute to, and work closely with Global Emissions Inventory Activity (GEIA) which was initiated in the first phase of IGAC and is now an activity of AIMES. IGAC will identify important gaps in emissions inventories and encourage evaluation by models and observations. In this area of research, IGAC will also interact with Surface Ocean – Lower Atmosphere Study (SOLAS) and Integrated Land Ecosystem – Atmosphere Processes Study (iLEAPS).

IGAC will collaborate closely with Stratospheric Processes and Their Role in Climate (SPARC) to develop research strategies to thoroughly investigate the processes controlling the distribution of chemical species in the atmosphere. Similarly, IGAC will collaborate with iLEAPS and with the Global Energy and Water Cycle Experiment (GEWEX) on investigations of the con-

centration, size and composition of aerosols that act as cloud condensation nuclei.

Palaeo climate records indicate wide variations in natural aerosol loadings and trace gas concentrations. However, to estimate the climatic impacts of these changes it is necessary to know the radiative effect of present levels of natural aerosols. Attribution of present Aerosol Optical Depth (AOD) to natural and anthropogenic aerosol components will lay the groundwork for such palaeo climate analyses. In this research IGAC will collaborate closely with the Past Global Changes (PAGES) project.

Many aspects of IGAC science are relevant to both climate and air quality, with the latter in particular bridging the biophysical and human dimension sciences. In this area IGAC will link to the Urbanisation project of the International Human Dimensions Programme on Global Environmental Change (IHDP). Vast amounts of aerosol data are routinely gathered around the world to monitor local air quality and to guide local emission control strategies so as to minimise the effects on human health. IGAC will collaborate with IHDP to provide a forum for linking these monitoring programmes in order to assess regional and global-scale effects. Similarly, mega-city plumes can provide excellent calibration targets for satellite sensors, due to high concentrations and strong gradients. Chemical characterisation of mega-city plumes will assist long-range transport and transformation studies. IGAC will foster the acquisition of high quality and consistent data across diverse regions, benefiting both the climate and air quality research communities.

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Acronym List

ACCENT	Atmospheric Composition Change: the European Network of Excellence
ACE	Aerosol Characterization Experiment
AIMES	Analysis, Integration and Modelling of the Earth System
AOD	aerosol optical depth
APARE	East Asia–North Pacific Regional Experiment
CACGP	Commission on Atmospheric Chemistry and Global Pollution
ENVISAT	Environment Satellite
ESSP	Earth System Science Partnership
ESA	European Space Agency
GEIA	Global Emissions Inventory Activity
GWP	global warming potential
HCFC	hydro-chlorofluorocarbon
IAMAS	International Association of Meteorology and Atmospheric Sciences
IGAC	International Global Atmospheric Chemistry
IGBP	International Geosphere-Biosphere Programme
IGOS-P	Integrated Global Observing Strategy Partnership
IHDP	International Human Dimensions Programme on Global Environmental Change
iLEAPS	Integrated Land Ecosystem – Atmosphere Processes Study
INDOEX	Indian Ocean Experiment

IPCC	Inter-governmental Panel on Climate Change
IPO	International Project Office
ITCT	Intercontinental Transport and Chemical Transformation
LiDAR	Light Detection and Ranging
MOZAIC	Measurements of Ozone and Water Vapour by In-service Airbus Aircraft
NARE	North Atlantic Regional Experiment
NASA	US National Aeronautic and Space Administration
ppbv	parts-per-billion by volume
SAFARI	Southern African Fire-Atmosphere Research Initiative
SeaWiFS	Sea-viewing Wide Field-of-view Sensor
SHADOZ	Southern Hemisphere Additional Ozonesondes
SOLAS	Surface Ocean – Lower Atmosphere Study
SPARC	Stratospheric Processes and their Role in Climate
SSC	Scientific Steering Committee
TRACE-A	Tropospheric and Atmospheric Chemistry near the Equator-Atlantic
TTL	tropical tropopause layer
UNFCCC	United Nations Framework Convention on Climate Change
VOC	volatile organic compounds
WCRP	World Climate Research Programme
WMO	World Meteorological Organization

IGAC

The International Global Atmospheric Chemistry project is a multidisciplinary project of the International Geosphere-Biosphere Programme (IGBP) and the Commission on Atmospheric Chemistry and Global Pollution (CACGP). IGBP is an interdisciplinary bodies of the International Council for Science (ICSU). CACGP is one of the commissions of the International Association of Meteorology and Atmospheric Sciences (IAMAS), which is turn is one of the associations of the International Union of Geodesy and Geophysics (IUGG) – an international scientific union of ICSU.

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