IGACtivities Newsletter

of the International Global Atmospheric Chemistry Project

Issue No. 29
December 2003

In this Issue

A Note from the IGAC Co-Chairs:
Tim Bates, Sandro Fuzzi, and Shaw Liu

This issue of IGACtivities describes the first three Tasks endorsed by the IGAC SSC under Phase II of IGAC. They are Air-Ice Chemical Interactions (AICI), Intercontinental Transport and Chemical Transformation (ITCT-Lagrangian-2k4), and Mega-Cities: Asia as a source of aerosols and oxidants (Mega-Cities). IGAC Phase II Tasks are organized around clearly defined science goals with finite project lifetimes. Previous IGAC Phase I Activities were terminated at the end of 2002, when IGAC Phase II officially commenced. Requirements for an IGAC Task can be found at (www.igac.noaa.gov/proposals.php).

We note that all three Tasks are just beginning and would like to encourage participation from scientists who could contribute to any of the Tasks described here. Interested scientists should contact the authors of the Task articles. Further information about the Tasks, including their plans for data dissemination and outreach, can be found on the IGAC web page (www.igac.noaa.gov).

One of IGAC's goals is to facilitate interaction between scientists in different disciplines relating to atmospheric chemistry. In particular, IGAC will be working closely with two Core Projects within IGBP that relate directly IGAC:

SOLAS (Surface Ocean Lower Atmosphere Study) is a new project that is addressing the ocean/atmosphere interface. (http://www.uea.ac.uk/env/solas/). In fact, the first Task in this newsletter, AICI, a joint Task between IGAC and SOLAS. We would like to encourage proposals for other multi-disciplinary Tasks.

ILEAPS (Integrated Land Earth Atmosphere Process Study) is a new project that is addressing the land/atmosphere interface. We would like to congratulate ILEAPS on the success of their first Open Science Meeting, held 29 September-3 October in Helsinki, Finland. (http://www.atm.helsinki.fi/ILEAPS/).

In addition, **SPARC** (Stratospheric Process and their Role in Climate), working under the auspices of the World Climate Research Program (WCRP), shares an interest in the links between chemistry and climate in the upper troposphere/lower stratosphere region (http://www.aero.jussieu.fr/~sparc/). A very successful joint SPARC-IGAC workshop was held in Giens, France, during 3-5 April 2003 to assess the current state of our understanding on some of the key issues related to climate-chemistry interactions. A report on the major findings of the workshop is planned as the main science feature for the next issue of the IGACtivities Newsletter.

This issue of IGACtivities is the first published by our new Taipei Project Office. We thank Sarah Doherty (Seattle Project Office) for her editorial assistance and Doris Chen (Taipei Project Office) for managing the production and dissemination of the newsletter.

GLOBAL CHANGE IGBP

CACGP

SCIENCE FEATURES

Air-Ice Chemical Interactions (AICI)

Contributed by **Eric Wolff** (ewwo@bas.ac.uk), *British Antarctic Survey, High Cross, Madingley Road, Cambridge CB3 0ET, UK, and* **Paul Shepson** (pshepson@purdue.edu), *Department of Chemistry, Purdue University, 560 Oval Drive, West Lafayette, Indiana 47907-2084, USA*

Introduction

It is now recognised that changes in one compartment of the Earth System can strongly affect the state of other compartments. Feedbacks can amplify or mitigate trends. As the Earth and its climate change, particularly in response to phenomena such as greenhouse gas increases, and stratospheric ozone depletion, it becomes increasingly important to understand the interactions between different parts of the system. This has been the basis for the new structure of IGBP, which studies the three main compartments (land, ocean, and atmosphere) and the exchanges and interactions between them. The cryosphere – ice, snow and permafrost – occurs in all three of these compartments. It forms a large proportion of the surface separating the land and ocean from the atmosphere: a seasonal maximum of 40% of land is covered by snow or ice, while several percent of the ocean is sea-ice covered (Figure 1).

The physical processes that involve ice are now being studied by the World Climate Research Program project CliC (Climate and Cryosphere). Until recently, it was generally assumed that the main biogeochemical role of ice was that it restricted exchanges between the more active ocean and land surfaces and the atmosphere. However, studies in recent years have revealed evidence that important chemical exchanges also occur between ice and the atmosphere, and it makes sense to study these processes and their consequences generically. AICI aims to do this, forming a bridge between scientists working on ice-covered surfaces in both polar regions and in the mid-latitudes, atmospheric scientists interested in processes occurring on ice particles, laboratory scientists studying the properties of ice, and modellers who need to parameterise processes that involve ice.

This project has been jointly endorsed by IGAC and SOLAS (Surface Ocean Lower Atmosphere Study) – both of which are IGBP Core Projects – but it is also relevant to ILEAPS (Integrated Land Ecosystem-Atmosphere Process Study), to PAGES (Past Global Changes), as well as to WCRP-CliC.

Scientific Context

Until recently, the snow and ice covered regions of the world received little interest from the atmospheric chemistry community. However, a number of field programmes, principally in the Arctic and Antarctic have revealed many unexpected and interesting phenomena, and opened up the possibility that the chemistry of the ice may control aspects of boundary layer chemistry over large regions of the world, rather than the other way round. In addition, for some species the exchanges between snow and the atmosphere, as well as post-depositional ice processes, have a decisive impact on the signal that is retrieved from ice cores: our best hope of reconstructing atmospheric chemistry from the pre-instrumental period.

Below we outline, as examples, two of the issues that have become apparent in recent years, as a preamble to setting questions that will be attacked within the lifespan of the AICI Task.

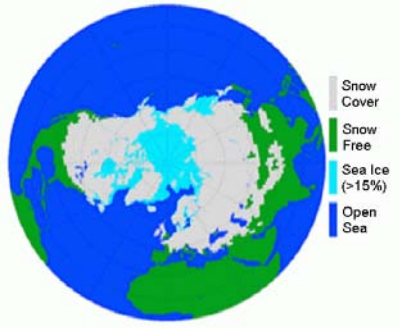

Figure 1 – Shown is the maximum northern hemisphere snow and sea ice cover from January, 1979. Image courtesy of the National Snow and Ice Data Center, University of Colorado, Boulder. Source: Northern Hemisphere EASE-Grid Weekly Snow Cover and Sea Ice Extent product.

Example 1 – tropospheric ozone depletion

The discovery of ozone depletion events within the polar marine boundary layer [*Bottenheim et al.*, 1986; *Oltmans and Komhyr*, 1986] came as a surprise. Since the initial finding, we have learnt much about the phenomenon, but we are still far from being able to predict the spatial or temporal distribution of the depletion, or to model the effects. The surprising magnitude of the effect points to weaknesses in our understanding of tropospheric ozone chemistry. This must be remedied, given its central role with respect to the atmosphere's oxidizing capacity, and its significant contribution to radiative forcing.

Tropospheric ozone depletion occurs in both the Arctic (Figure 2), sub-Arctic and the Antarctic [*Wessel et al.*, 1998]. In the Arctic marine boundary layer in springtime, depletion occurs from the surface up to ~1.5km [*Bottenheim et al.*, 2002b], and occurs over periods of days to weeks, with concentrations as low as 50 ppt at times. The depletion events are associated with halogens, with Br/BrO appearing to play an especially important role [e.g. *Barrie et al.*, 1988; *Foster et al.*, 2001]. Satellite observations of BrO, showing particularly high concentrations over sea ice zones in springtime [*Wagner et al.*, 2001], have reinforced current ideas about the general nature of the reactions leading to depletion, and suggested that sea ice is implicated. Recently it has been suggested that concentrated brines on new sea ice and in frost flowers [*Rankin et al.*, 2002] might be the source of the halogens. This would explain why a similar phenomenon is seen over salt lakes [*Stutz et al.*, 2002] and the Dead Sea [*Matveev et al.*, 2001] where very salty deposits are also available. Brines and frost flowers may also be responsible for a significant part of sea salt aerosol concentrations over the polar regions [*Rankin et al.*, 2002]. Among the chemical effects of the ozone depletion is a perturbation to the biogeochemistry of mercury, leading to enhanced atmospheric removal and deposition to Arctic ecosystems [*Lindberg et al.*, 2002; *Schroeder et al.*, 1998]; this process has also been reported from the Antarctic [*Ebinghaus et al.*, 2002]. Finally, it is also noteworthy that ozone depletion occurs in cirrus clouds; it is conceivable that these chemistries could be related [*Roumeau et al.*, 2000].

This work gives a picture of a phenomenon for which we have a skeleton understanding, but as yet no answers to some important questions. We do not yet understand why depletion occurs in the spring but not in the autumn; we have no confirmation that frost flowers are involved; we have not explored the likely effects on atmospheric boundary layer chemistry in full; we cannot completely identify the precursors to halogen atoms; and we have not established whether the phenomenon is involved in significant climate feedbacks.

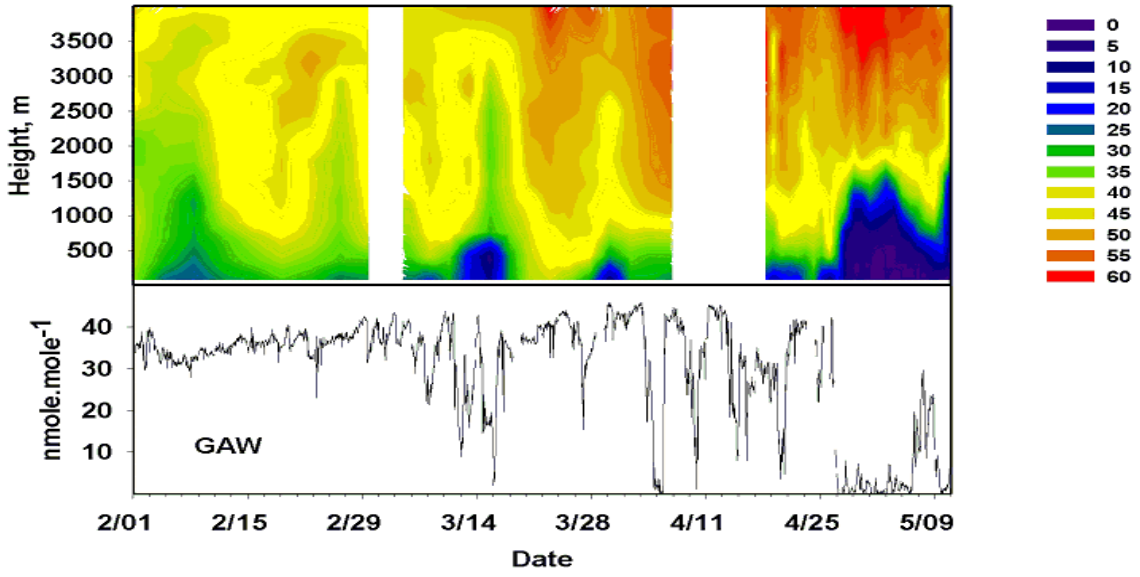

Figure 2 – From Bottenheim et al., 2002b. Shown are tropospheric ozone concentrations for the winter and spring of 2000 at the Alert Baseline Observatory in Alaska. This station is part of the Global Atmosphere Watch network of the WMO. Shown are concentrations from the lowest 4km of the atmosphere, measured using sondes, (top panel; in nmole·mole^{-1}) and surface ozone concentrations (bottom panel).

Example 2 – snow photochemistry

Until recently, no consideration was given to the concept that the snowpack could itself be affected by photochemistry. However, a series of field [e.g. *Honrath et al.*, 1999; *Jones et al.*, 2000] and laboratory [e.g. *Cotter et al.*, 2003; *Dubowski et al.*, 2001; *Honrath et al.*, 2000] experiments have established beyond doubt that NO_x, as the first example, is produced photochemically from snow, with nitrate as the main precursor. In retrospect, this is not surprising – light penetrates at least the top few cm of snowpack, and many hydrophilic UV-absorbing chemicals are present in concentrations that are high relative to those in the atmosphere [*Wolff et al.*, 2002].

Comparisons with model results have shown that emissions out of the snowpack can overwhelmingly control the chemistry of the sunlit Summer boundary layer over snow, particularly in cases where there is little mixing due to a surface inversion. For example, NO was present at South Pole in concentrations over 200 pptv during December 1998 [*Davis et al.*, 2001], when model calculations excluding a snow source would have suggested 1-5 pptv. Very high OH concentrations [*Mauldin et al.*, 2001] were also observed, and the NO_x rich environment may have been responsible for this, in turn influencing the entire range of chemistry. The full implications have not yet been explored, but ozone production over a large part of the Antarctic plateau is one probable consequence [*Crawford et al.*, 2001].

Many other chemicals seem to be produced photochemically from the snowpack – there is evidence so far for at least aldehydes such as HCHO [*Sumner and Shepson*, 1999; *Sumner et al.*, 2002], CO [*Haan et al.*, 2001], C_2H_2 [*Bottenheim et al.*, 2002a] and HONO [*Zhou et al.*, 2001]. In addition, there is extensive physical exchange of important species such as HCHO and H_2O_2 [*Hutterli et al.*, 2001; *Hutterli et al.*, 1999]. Until now, only the existence of the phenomenon, and some tentative flux estimates [e.g. *Beine et al.*, 2002; *Honrath et al.*, 2002; *Jones et al.*, 2001] are available. Much work is needed, in the laboratory and the field, to understand the factors controlling the release of each chemical. Only then will it be possible, through modelling studies, to assess whether the processes have any large-scale importance. However, it is already clear that such snowpack chemistry will significantly affect both ice phase and boundary layer concentrations for photo-labile species. With our current state of knowledge, we assume that similar emission processes are occurring over mid-latitude and seasonal snowcovers, so that there is the potential for significant alteration of the boundary layer chemistry over substantial parts of the globe, including urban areas.

As far as we know, the phenomena described above occur under natural conditions and have always been present. However, changes in UV radiation, in concentrations of pollutants, in snow cover, and in temperature, could all affect the extent of the processes. For example, the tropospheric ozone production reported at the South Pole [*Crawford et al.*, 2001] seems to have increased over the last 30 years; this could be partly a result of increased NO_x production that would be predicted from increased UV radiation reaching the surface (because of stratospheric ozone depletion) [*Jones and Wolff*, 2003]; indeed we would expect all the photochemical reactions in snow to be enhanced in this way. The chemistry of halogen activation, as well as emission of species such as HONO, may be pH-dependent and thus impacted by acidic deposition from anthropogenic sources.

One of the key expectations of a warming world (under increased greenhouse gas concentrations) is that the cryosphere will shrink. In the near-term, seasonal snow cover and sea ice extent are expected to diminish. This will both reduce the scope for the snow-atmosphere interactions (such as those described above) to occur, and enhance the interactions of the atmosphere and the increased land and ocean areas. As an example, if the springtime depletion of tropospheric ozone is indeed linked to brine layers on new sea ice, then any change in the production rate of new sea ice will alter the locations and extent of this phenomenon. There are indications that Arctic ozone depletion events, and concomitant deposition of Hg, have increased in recent decades [*Lindberg et al.*, 2002; *Tarasick and Bottenheim*, 2002]. We would also anticipate a change in the concentration of sea salt aerosol, with further possible direct and indirect climate effects.

Surface uptake by ice is highly temperature dependent, so that climate change can effect the deposition of contaminants (such as persistent organic pollutants (POPs)) to the polar regions. Even the nature of the surface of ice is highly temperature dependent, and this may affect the nature of ice surface chemistry [*Cho et al.*, 2002]. Assessments of possible feedbacks such as these are central to understanding the effects of global change.

Scientific Objectives

IGAC has two over-arching science questions, and each has a corresponding question directly relevant to the AICI Task:

- What is the role of atmospheric chemistry in amplifying or damping climate change?
  - **How will changing amounts of sea ice, snow cover, and atmospheric ice alter atmospheric chemistry and composition and are there important feedbacks to climate?**

- Within the Earth System, what effects do changing regional emissions and depositions…have on air quality and the chemical composition of the planetary boundary layer?
  - **What are the present regional emissions and losses over snow, ice and sea ice, and how could they alter with changing climate?**

For the sea ice component, these same issues address various SOLAS activities, such as: sea-salt particle formation and transformations; trace gas emissions and photochemical feedbacks; and understanding halogen release from sea ice.

Obviously the two science questions we have highlighted in bold, as well as the associated IGAC questions, are similarly large, and unlikely to be satisfactorily answered in the suggested ~3-year timeframe for an IGAC Task. We have therefore established a more limited set of goals that we will undertake in the first phase of the AICI Task:

- To document the full range of processes and emitted trace gases that arise at the air-ice interface, and how they depend on environmental conditions.

- To quantify the fluxes of trace gases and aerosol between atmosphere and ice under a range of field experimental conditions

- To determine the main factors that control the fluxes of trace gases between air and ice, using both field data and carefully designed laboratory experiments

- To build simple models that include processes in the upper firn and lower atmosphere, to assess the effects of air-ice fluxes

- To make a first assessment of the scale of these processes, and assess the significance for climate or atmospheric chemistry within the global troposphere, the atmospheric boundary layer, or regionally, and in urban environments.

- To develop simple parameterisations of the fluxes and processes studied for incorporation into sophisticated global chemical models

Based on the outcome of this first phase of AICI, we envisage a second AICI project in which more quantitative fluxes and parameterisations will be made, and the effects of changing climate and cryosphere will be rigorously assessed, through a combination of regional scale field studies, with associated laboratory and modeling activities.

Task Activities

Addressing the goals outlined above will require a combined programme of field measurements, laboratory measurements, satellite data retrieval, and modelling studies. The field work poses a particular challenge because of the remoteness of the polar sites, so that only through a range of international co-operative activities can sufficient data be obtained. The organisation of Arctic and Antarctic logistics has previously dictated that most studies were led by a single country, with participation from others; we regard the future integration of the different experiments and leveraging of multinational resources to be an important added value that AICI can provide. The following elements will be essential, some of which are already in place, and some of which we will encourage:

- **Comprehensive studies in snow/ice covered areas of a range of types, locations, and time periods.** Some studies will focus on particular times of year, but year-round studies will also be important so that we can be sure all relevant processes are being observed. Profiling studies, involving aircraft, tethered balloons, and towers, will be needed, as well as ground-based studies. The range of environments should include:

  - Clean permanent inland ice sheet locations, where snow photochemistry can most simply be studied (ANTCI at South Pole and Geosummit in Greenland are examples of studies that are already funded and whose

value could be enhanced by the addition of AICI activities).

- Coastal marine ice sheet sites, where both snow photochemistry under different climatic and snow conditions, as well as gas phase and aerosol processes associated with new sea ice surfaces, can be studied. The year-round CHABLIS project at Halley Bay (Antarctica), ongoing studies at sites such as Neumayer and Dumont d'Urville (Antarctica), Alert, Barrow and Ny Ålesund (Arctic) are examples of this category. Again additional activities could be added to these existing sites.

- Studies within the sea ice zone, using for example a ship or platform, are ideal for studying processes over sea ice. The proposed US SOLAS OASIS study falls into this category.

- Studies over Alpine and seasonal snow-cover environments are necessary to assess how widely applicable the polar findings are and to determine the importance of regional scale anthropogenic inputs to the snowpack and the resultant impact of the subsequent photochemical processes.

- Studies in urban snow-impacted environments, which are likely to have much higher concentrations of key ice-surface reactants – e.g. nitrate, organic matter, etc.

- **Laboratory experiments to assess the fundamental processes and controlling factors.** These are likely to include use of idealised artificial ice experiments, to determine quantum yields and uptake coefficients, diffusion coefficients, reaction rates and equilibrium constants, as well as controlled studies of natural snow to isolate the factors controlling fluxes.
- **Use of satellite data to determine the spatial and seasonal distribution of key species and environments.** For example, it is expected that far more specific indicators of sea ice type (including algorithms for frost flower extent) will be developed, which can be correlated with the occurrence of BrO or of ozone loss.

- The field studies above will necessarily be limited to fairly local areas, and the satellite data will mainly look at columns and will be less able to assess the extent of phenomena (such as boundary layer ozone depletion) at ground level. **To gain a full picture of the spatial extent, it will be necessary to develop autonomous instrument packages that could produce a low resolution year-round record of key species at unattended sites.** Such platforms would be a highly appropriate contribution to the planned International Polar Year (IPY; 2007-08). While IPY will not fall into this phase of AICI, the package development, which could be used by many nations, will.

- **Improvements in instrumentation** will be needed to make the necessary measurements at the low concentrations, and under the difficult conditions, encountered at some sites.

- **Simple models of the processes within the upper snowpack and the lower atmosphere** will be used to assess: 1) the full range of likely impacts of the fluxes observed, 2) the possible influence on the larger-scale atmosphere, and 3) influences on the material finally preserved in the ice core record. 1-D and 3-D chemical transport models will ultimately need to incorporate these ice-atmosphere interactions.

AICI does not include field studies of ice in the upper troposphere (e.g. cirrus), but we recognise that such studies, by others, can benefit from and contribute to advances in the other areas discussed here.

The role of the AICI Task Team is to coordinate international research and education activities along the lines described and to efficiently leverage multinational research, human, and logistics resources with the assistance of IGAC and SOLAS. The AICI Steering Committee has representation from key participating countries, and critical science areas. The function of the SC is to disseminate information about activities falling within the AICI remit, to coordinate such activities, and to encourage activities that are not currently funded or planned.

Some of the field activities necessary for AICI are already funded and planned, which is fortunate given the long lead time of polar field studies, and the need for many of them to run for a year or more. The data from several of these field campaigns should become available throughout the task period. In order to stimulate linking activities between them, as well as further field and modelling studies, we will take advantage of the "Air-ice interactions" session at the IGAC conference in Christchurch, New Zealand

in September 2004 (*see "Announcements" at the end of this Newsletter*) and will hold AICI meetings in conjunction with it. We also plan to hold small workshops where the data from the campaigns, laboratory and other work can be discussed, during 2005-6. The task will close with an AICI symposium and a related journal special issue late in 2006, at which future plans, and the need for a further task, will be discussed.

References

Barrie, L.A., J.W. Bottenheim, R.C. Schnell, P.J. Crutzen, and R.A. Rasmussen, Ozone Destruction and Photochemical-Reactions at Polar Sunrise in the Lower Arctic Atmosphere, *Nature*, *334*, 138-141, 1988.

Beine, H.J., R.E. Honrath, F. Domine, W.R. Simpson, and J.D. Fuentes, NOx during background and ozone depletion periods at Alert: Fluxes above the snow surface, *J. Geophys. Res.*, *107*, 4584, doi:10.1029/2002JD002082, 2002.

Bottenheim, J.W., H. Boudries, P.C. Brickell, and E. Atlas, Alkenes in the Arctic boundary layer at Alert, Nunavut, Canada, *Atmos. Env.*, *36*, 2585-2594, 2002a.

Bottenheim, J.W., J.D. Fuentes, D.W. Tarasick, and K.G. Anlauf, Ozone in the Arctic lower troposphere during winter and spring 2000 (ALERT2000), *Atmos. Env.*, *36*, 2535-2544, 2002b.

Bottenheim, J.W., A.G. Gallant, and K.A. Brice, Measurements of Noy Species and O-3 at 82-Degrees-N Latitude, *Geophys. Res. Let.*, *13*, 113-116, 1986.

Cho, H., P.B. Shepson, L.A. Barrie, J.P. Cowin, and R. Zaveri, NMR investigation of the quasi-brine layer in ice/brine mixtures, *J. Phys. Chem. B*, *106*, 11226-11232, 2002.

Cotter, E.S., A.E. Jones, E.W. Wolff, and S.J.-B. Bauguitte, What controls photochemical NO and NO_2 production from Antarctic snow? Laboratory investigation assessing the wavelength and temperature dependence, *J. Geophys. Res.*, *108*, 4147, doi:10.1029/2002JD002602, 2003.

Crawford, J.H., D.D. Davis, G. Chen, M. Buhr, S. Oltmans, R. Weller, L. Mauldin, F. Eisele, R. Shetter, B. Lefer, R. Arimoto, and A. Hogan, Evidence for photochemical production of ozone at the South Pole surface, *Geophys. Res. Let.*, *28*, 3641-3644, 2001.

Davis, D., J.B. Nowak, G. Chen, M. Buhr, R. Arimoto, A. Hogan, F. Eisele, L. Mauldin, D. Tanner, R. Shetter, B. Lefer, and P. McMurry, Unexpected high levels of NO observed at South Pole, *Geophys. Res. Let.*, *28*, 3625-3628, 2001.

Dubowski, Y., A.J. Colussi, and M.R. Hoffmann, Nitrogen dioxide release in the 302 nm band photolysis of spray-frozen aqueous nitrate solutions. Atmospheric implications, *J. Phys. Chem. A*, *105*, 4928-4932, 2001.

Ebinghaus, R., H.H. Kock, C. Temme, J.W. Einax, A.G. Lowe, A. Richter, J.P. Burrows, and W.H. Schroeder, Antarctic springtime depletion of atmospheric mercury, *Env. Sci.& Tech.*, *36*, 1238-1244, 2002.

Foster, K.L., R.A. Plastridge, J.W. Bottenheim, P.B. Shepson, B.J. Finlayson-Pitts, and C.W. Spicer, The role of Br-2 and BrCl in surface ozone destruction at polar sunrise, *Science*, *291*, 471-474, 2001.

Haan, D., Y. Zuo, V. Gros, and C.A.M. Brenninkmeijer, Photochemical production of carbon monoxide in snow, *J. Atmos. Chem.*, *40*, 217-230, 2001.

Honrath, R.E., S. Guo, M.C. Peterson, M.P. Dziobak, J.E. Dibb, and M.A. Arsenault, Photochemical production of gas phase NOx from ice crystal NO3, *J. Geophys. Res.*, *105*, 24183-24190, 2000.

Honrath, R.E., Y. Lu, M.C. Peterson, J.E. Dibb, M.A. Arsenault, N.J. Cullen, and K. Steffen, Vertical fluxes of NOx, HONO, and HNO3 above the snowpack at Summit, Greenland, *Atmos. Env.*, *36*, 2629-2640, 2002.

Honrath, R.E., M.C. Peterson, S. Guo, J.E. Dibb, P.B. Shepson, and B. Campbell, Evidence of NOx production within or upon ice particles in the Greenland snowpack, *Geophys. Res. Let.*, *26* , 695-698, 1999.

Hutterli, M.A., J.R. McConnell, R.W. Stewart, H.-W. Jacobi, and R.C. Bales, Impact of temperature-driven cycling of hydrogen peroxide (H_2O_2) between air and snow on the planetary boundary layer, *J. Geophys. Res.*, *106*, 15395-15404, 2001.

Hutterli, M.A., R. Rothlisberger, and R.C. Bales, Atmosphere-to-snow-to-firn transfer studies of HCHO at Summit, Greenland, *Geophys. Res. Let.*, *26*, 1691-1694, 1999.

Jones, A.E., R. Weller, P.S. Anderson, H.-W. Jacobi, E.W. Wolff, O. Schrems, and H. Miller, Measurements of NO_x emissions from the Antarctic snowpack, *Geophys. Res. Let.*, *28*, 1499-1502, 2001.

Jones, A.E., R. Weller, E.W. Wolff, and H.-W. Jacobi, Speciation and rate of photochemical NO and NO_2 production in Antarctic snow, *Geophys. Res. Let.*, *27*, 345-348, 2000.

Jones, A.E., and E.W. Wolff, An analysis of the oxidation potential of the South Pole boundary layer and the influence of stratospheric ozone depletion, *J. Geophys. Res.*, 108 (D18), 4565, doi:10.1029/2003JD003379, 2003.

Lindberg, S.E., S. Brooks, C.J. Lin, K.J. Scott, M.S. Landis, R.K. Stevens, M. Goodsite, and A. Rich-

ter, Dynamic oxidation of gaseous mercury in the Arctic troposphere at polar sunrise, *Env. Sci. & Tech.*, *36*, 1245-1256, 2002.

Matveev, V., M. Peleg, D. Rosen, D.S. Tov-Alper, K. Hebestreit, J. Stutz, U. Platt, D. Blake, and M. Luria, Bromine oxide - ozone interaction over the Dead Sea, *J. Geophys. Res.*, *106*, 10375-10387, 2001.

Mauldin, R.L., F.L. Eisele, D.J. Tanner, E. Kosciuch, R. Shetter, B. Lefer, S.R. Hall, J.B. Nowak, M. Buhr, G. Chen, P. Wang, and D. Davis, Measurements of OH, H2SO4, and MSA at the South Pole during ISCAT, *Geophys. Res. Let.*, *28*, 3629-3632, 2001.

Oltmans, S.J., and W.D. Komhyr, Surface ozone distributions and variations from 1973-1984 measurements at the NOAA Geophysical Monitoring for Climate Change baseline observatories, *J. Geophys. Res.*, *91*, 5229-5236, 1986.

Rankin, A.M., E.W. Wolff, and S. Martin, Frost flowers - implications for tropospheric chemistry and ice core interpretation, *J. Geophys. Res.*, *107*, 4683, doi:10.1029/2002JD002492, 2002.

Roumeau, S., P. Bremaud, E. Riviere, S. Baldy, and J.L. Baray, Tropical cirrus clouds: a possible sink for ozone, *Geophys. Res. Let.*, *27*, 2233-2236, 2000.

Schroeder, W.H., K.G. Anlauf, L.A. Barrie, J.Y. Lu, A. Steffen, D.R. Schneeberger, and T. Berg, Arctic springtime depletion of mercury, *Nature*, *394*, 331-332, 1998.

Stutz, J., R. Ackermann, J.D. Fast, and L. Barrie, Atmospheric reactive chlorine and bromine at the Great Salt Lake, Utah, *Geophys. Res. Let.*, *29*, art. no.-1380, 2002.

Sumner, A.L., and P.B. Shepson, Snowpack production of formaldehyde and its effect on the Arctic troposphere, *Nature*, *398*, 230-233, 1999.

Sumner, A.L., P.B. Shepson, A.M. Grannas, J.W. Bottenheim, K.G. Anlauf, D. Worthy, W.H. Schroeder, A. Steffen, F. Domine, S. Perrier, and S. Houdier, Atmospheric chemistry of formaldehyde in the Arctic troposphere at Polar Sunrise, and the influence of the snowpack, *Atmos. Env.*, *36*, 2553-2562, 2002.

Tarasick, D.W., and J.W. Bottenheim, Surface ozone depletion episodes in the Arctic and Antarctic from historical ozonesonde records, *Atmos. Chem, & Phys.*, *2*, 197-205, 2002.

Wagner, T., C. Leue, M. Wenig, K. Pfeilsticker, and U. Platt, Spatial and temporal distribution of enhanced boundary layer BrO concentrations measured by the GOME instrument aboard ERS-2, *J. Geophys. Res.*, *106*, 24225-24235, 2001.

Wessel, S., S. Aoki, P. Winkler, R. Weller, A. Herber, H. Gernandt, and O. Schrems, Tropospheric ozone depletion in polar regions - A comparison of observations in the Arctic and Antarctic, *Tellus B*, *50*, 34-50, 1998.

Wolff, E.W., A.E. Jones, T.J. Martin, and T.C. Grenfell, Modelling photochemical NOx production and nitrate loss in the upper snowpack of Antarctica, *J. Geophys. Res.*, *29*, 1944, doi:10.1029/2002GL015823, 2002.

Zhou, X.L., H.J. Beine, R.E. Honrath, J.D. Fuentes, W. Simpson, P.B. Shepson, and J.W. Bottenheim, Snowpack photochemical production of HONO: a major source of OH in the Arctic boundary layer in springtime, *Geophys. Res. Let.*, *28*, 4087-4090, 2001.

Intercontinental Transport and Chemical Transformation (ITCT-Lagrangian-2k4)

Contributed by **David Parrish** (parrish@al.noaa.gov), *NOAA Aeronomy Lab, 325 Broadway R/AL7, Boulder, CO 80305-3328, USA, and* **Kathy Law** (kathy.law @aero.jussieu.fr), *IPSL Service Aéronomie Boite 102,Université Pierre et Marie Curie, 4 Place Jussieu, 75252 Paris Cedex 05, France*

Introduction

An overarching goal of the ITCT program is to understand the chemical transformation and removal processes of aerosols, oxidants and their precursors during the intercontinental transport process. To make this understanding possible, measurements from a Lagrangian platform would be ideal; i.e., a platform that moves with an air mass during the total transport process. Such an ideal is not possible due to the limited range and endurance of existing aircraft. A practical approximation to this ideal, is a "psuedo-Lagrangian" study, where one or more aircraft make multiple, sequential sampling flights into the same air mass during the time required for the intercontinental transport of the air mass. Such a pseudo-Lagrangian study constitutes the IGAC Task described here.

In the Summer of 2004 a large international group of scientists will conduct a field program in the North Atlantic region. The program will focus on the study of emissions of aerosol and ozone precursors over

North America, their chemical transformations and removal during transport to and over the North Atlantic, and their impact upon Europe. The program is currently being organized and funded by several agencies including NOAA (U.S.) through the NEAQS - ITCT 2004 program (http://www.al.noaa.gov/2004/), and NASA (U.S.) through the INTEX-NA program (http://cloud1.arc.nasa.gov/intex-na/). The European contribution, Intercontinental Transport of Pollutants (ITOP;http://www-users.york.ac.uk/~chem89/European_ITOP_homepage), includes contributions from a UK university consortium (funded by NERC), DLR (Germany) and CNRS (France). Each of these programs has its own regionally focused goals and deployments, but together they provide coverage from the source regions on North America, through the transport pathways over the North Atlantic, and over the receptor regions of Europe. The ITCT-Lagrangian-2k4 Task is an organizational and analysis effort that will, within the individual project goals, coordinate the disparate programs into a pseudo-Lagrangian framework. That is, we will combine data from the multiple observational platforms collected at different stages during the transit of a polluted air mass across the North Atlantic.

Scientific Objectives

The ultimate goal of ITCT is the direct observation of the evolution of the aerosols, oxidants and their precursors from emission over North America, trans-Atlantic transformation and transport, and impact on aerosol and oxidant levels over Europe. Within this overall objective there are several distinct sub-objectives:

- **Determination of the photochemical oxidant and aerosol formation potentials in polluted air masses originating in North American emission regions and their chemical evolution as they are transported out over the North Atlantic.** This will involve quantification of physical and chemical loss processes of emitted species over North America, and detailed characterization of radical chemistry, VOCs and their degradation properties, the NOx/NOy budget and physical and chemical aerosol properties in the exported air masses. The contribution of natural sources (e.g. wild fires, biogenics, stratosphere, lightning) to oxidant and aerosol distributions will also be investigated.

- **Characterization of the dynamical processes responsible for pollutant transport out of the North American planetary boundary layer** (cyclonic air streams, fronts, convection, land- sea breezes, orographic effects) and the importance of mixing/layering processes in the long-range transport and chemical transformation of pollutant plumes as they cross the North Atlantic.

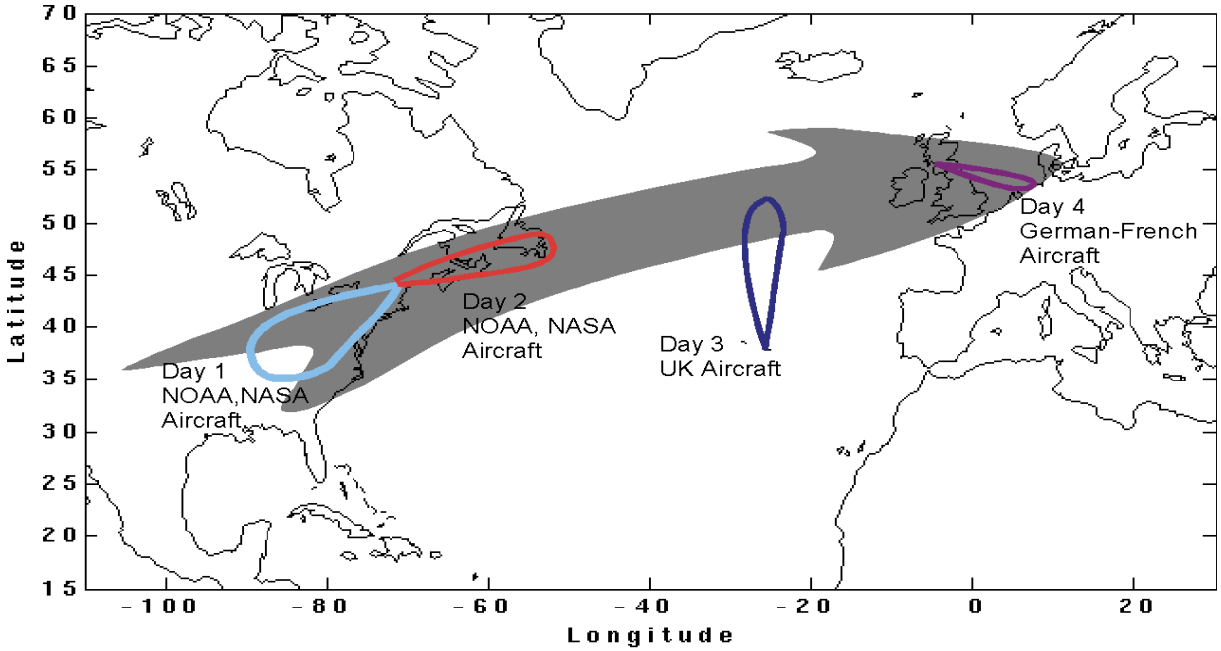

Figure 1 - The ITCT Task team will help coordinate multiple platforms participating in the Summer 2004 North Atlantic campaign so that the evolution of a given airmass (gray arrow) can be measured in a pseudo-Lagrangian manner.

- **Quantification of the export of North American pollutants to the background atmosphere, their subsequent fate over the North Atlantic and beyond (e.g. over Europe) and possible impact on climate.** This encompasses questions such as: are polluted layers producing or destroying ozone? Do these ageing air masses contain significant levels of anthropogenic aerosols and how is their chemical composition changing as they age?

- **Determination of the possible import of North American, and possibly Asian or other, pollutants into the boundary layer over Europe** where they may be contributing to background levels of pollutants and regional air quality.

The observational platforms and modeling activities currently proposed for the Summer 2004 campaign are quite extensive. In the source region and over the Western North Atlantic, NOAA will operate the WP-3D aircraft (with in situ gas phase and aerosol instrumentation plus radiation measurements), a remote sensing aircraft (with an ozone and aerosol lidar) and the Ronald H. Brown research vessel (with in situ and remote gas-phase and aerosol instrumentation plus radiation measurements). In that same region, NASA will operate the DC-8 and the P3-B, both with in situ and remote gas-phase and aerosol instrumentation. Within the framework of the UK's ITOP program the BAe-146 will operate over the central Atlantic from the Azores, and the German DLR and French CNRS Falcons will operate over the Eastern Atlantic and Western Europe with the aim of completing the pseudo-Lagrangian experiment. The UK aircraft will be equipped with a full range of chemical and aerosol measurements. On the European side, an ozone (and aerosol) lidar will be used to locate polluted layers already sampled by the North American and/or the UK aircraft. The DLR Falcon will then make more detailed chemical and aerosol measurements in these plumes.

These seven aircraft will also be coordinated with as many as five other research aircraft over North America. These include: the Canadian MSC Convair 580 and the Cal Tech/ONR/CIRPAS Twin Otter conducting detailed aerosol and cloud microphysical studies; the NSF/Harvard/COBRA program (operating the Wyoming King Air) conducting a carbon budget study; the U.S. Dept. of Energy G-1; and the University of Maryland Aztec aircraft. This suite of aircraft will be coordinated with a variety of ground sites including the AIRMAP network in New Hampshire, Harvard Forest in Massachusetts, a site on the southern tip of Nova Scotia, several other U.S. state networks, the PICO-NARE site on the Azores, and the German ATMOFAST lidar and high altitude surface sites.

It is hoped that other programs making measurements on commercial aircraft (i.e. MOZAIC, CARIBIC) will also participate in this task by making data available for the period of the measurement intensive. In the case of MOZAIC at least 1-2 flights are made daily across the Atlantic between European cities and, for example, New York or Boston collecting high temporal resolution data on O_3, CO and NOy concentrations. CARIBIC makes less frequent flights (1-2 per month) but with a more comprehensive instrument package that includes measurements of NOx, non-methane hydrocarbons, halogenated species and aerosols. Data from current and planned satellite platforms will contribute to the post-campaign analyses, and in some cases will be used for flight planning. Table 1 lists the satellite data sets that provide measurements of tropospheric species and biomass fire information. In addition, the near-real time visible and infrared imagery from GOES and METEOSAT and the winds derived from this imagery will provide flight-planning guidance. Satellite data that have been particularly useful for flight planning include aerosol products from TOMS, SEAWIFS, and MODIS, O_3 products from TOMS, and the fire data from MODIS. In 2004 we plan to utilize GOME and SCIAMACHY NO_2 columns and perhaps MOPPIT CO columns as well.

Many theoretical groups will be involved in forecasting for flight planning and in modeling for interpretation of results. These include global models (e.g. Harvard, NOAA/U, Iowa, U. Cambridge, MPI-Mainz, CNRS), regional models (U. New Hampshire, Environment Canada) and Lagrangian trajectory models (NOAA, U. Cambridge/Leeds, CNRS).

Coordination of a program to synthesize results from a pseudo-Lagrangian experiment over the North Atlantic requires an international research framework appropriate for IGAC; that coordination is central to the ITCT Task.

A goal of IGAC is to understand how atmospheric chemistry might evolve under climate change. However, the field project being used as the basis for the ITCT-Lagrangian-2k4 Task will, by necessity, cover only a very limited time window – approximately 6 weeks in Summer. The choice of season wasmade for several reasons: over North America it is the time of maximum photochemical production of oxidants and aerosols, of maximum biogenic emis-

Instrument	Species	Vertical Resolution in Troposphere	Current Status
TOMS	O_3, H_2O, Aerosol	Column	Operational since 1979
GOME	O_3, NO_2, CH_2O, SO_2	Column	Operating, but problems with data transmission
MOPPIT	CO, CH_4	Column & ~2 levels	Injured, but operating
SEAWIFS	Aerosol	Column	Operational since 1997
MODIS	Aerosol, fire hot spots	Column	Operational; near real time data available
MISR	Aerosol	Column	On Terra with MODIS
BIRD	Fire hot spots	----	Launched 2001
SCIAMACHY	O_3, CO, NO_2, CH_2O, SO_2, Aerosol	Column, Limb	Operational; near real time data available
MIPAS	O_3, H_2O, CO, HNO_3	Limb data in UT	Operational on Envisat
AURA-TES	O_3, H_2O, CO, NO HNO_3, SO_2	2-4 km resolution	Scheduled to be launched in early 2004
AURA-OMI	O_3, NO_2, CH_2O, SO_2, Aerosol	Column	Scheduled to be launched in early 2004

Table 1 - A suite of satellites are expected to be making measurements over the region of the Summer 2004 campaign that will be used in the ITCT Task.

sions of hydrocarbons that provide much of the fuel for this photochemistry, of enhanced stagnation episodes that allow the primary emissions, oxidants and aerosols to collect over the source regions, and a time of consistent northeastward transport of continental air masses to the North Atlantic troposphere. This narrow time window necessarily limits the investigation of natural variability on time scales longer than weeks. Contrasts with previous North Atlantic Regional Experiment (NARE) studies, which were conducted in the early Spring, late Summer and early Fall, will provide some information on seasonal variability.

However, in the interpretation of the results, it is necessary to consider the possibility of significant inter-annual variability. Importantly, the largest source of year-to-year variability of winter weather in the extra-tropical Northern Hemisphere is the North Atlantic Oscillation. Its influence on the variability of transport in the Summer over the North Atlantic is relatively weak, but still significant. To assess inter-annual variability, longer-term datasets, such as those from ozone sonde networks, AERONET and programs on commercial aircraft (MOZAIC, CARIBIC) will be used together with analysis of model results. Analysis of satellite data will also make an important contribution to our understanding of inter-annual variability of long-range transport of pollutants over the North Atlantic region.

Task Activities

The ITCT-Lagrangian-2k4 Task team has established a steering group (SG) to oversee pre-campaign planning, development of a coordinated flight strategy and post-campaign data analysis. The SG will include the task coordinators, representatives from the participating aircraft and forecasting and modeling groups, and other interested scientific participants.

The organization and analysis outlined above comprises four steps: review of previous results, instrument comparison activities (to ensure that measurements on the disparate platforms can be accurately integrated without confounding measurement uncertainties), flight coordination during the field deploy-

ment, and post-deployment analysis.

1. Retrospective Analysis of Previous P-seudo-Lagrangian Results.

During the NARE 1993 and NARE 1997 studies aircraft were operated on both the North American and European sides of the Atlantic. It was not the primary aim of these experiments to perform a Lagrangian study. These missions were more exploratory in nature, but events may have occurred during both studies when anthropogenically influenced air masses were sampled before and after transport across the Atlantic. However, examination of the data sets for such cases has been rather limited. The first effort of the ITCT-Lagrangian-2k4 task is to coordinate more extensive analysis. There are two periods to be examined. First, during NARE 1993 the NCAR King Air, operated in the Gulf of Maine by NOAA, characterized an air mass leaving the U.S. Trajectory calculations suggest the British C-130 may have sampled this same air mass during its return flight from Halifax to Britain. Second, during NARE 1997 the NOAA WP-3D characterized air masses off the U.S. east coast. In one case the British C-130 flying from the Azores may have sampled a characterized air mass after substantial transport. The DLR Falcon flying over Europe may also have sampled one of these air masses over Europe. Analysis of other datasets (e.g. MOZAIC, lidar, aircraft) may also reveal further interesting insights into the transport pathways and chemical signatures of pollutant plumes transported across the North Atlantic. The analysis of these earlier data has two primary goals: *First*, to see if measured levels of relatively inert tracers, such as carbon monoxide and long-lived NMHCs, support the indications from trajectory calculations that the same air mass was sampled. *Second*, to see if indications of processing can be discerned in the measured levels of more reactive species such as shorterlived NMHCs, oxides of nitrogen, ozone, and other oxidants. The successes and problems identified in these analyses of the early studies will serve as a valuable guide to the field implementation during Summer 2004.

2. Instrument Comparison Activities.

For the pseudo-Lagrangian approach to be successful, it is essential that the aircraft involved make measurements that are equivalent within quantified uncertainties. Quantifying measurement uncertainty establishes an objective, defensible basis upon which the pseudo-Lagrangian analysis can be built. In effect, a unified observation system is created. Comparison exercises will take place before, during, and after the 2004 field mission. The three general phases envisioned for the comparisons are outlined below, followed by a proposed strategy for their implementation.

Evaluation of standards (pre, during, post-mission): Comparison of compressed gas standards should be performed at least once for the different in-situ gas phase instruments on the participating platforms for NO, CO, CO_2, SO_2 and volatile organic compounds (VOCs). Comparison of ozone standards should also be performed. Effort will also be made to evaluate instrumental sensitivities to HNO_3 between the aircraft and shipborne instrumentation by sampling from a characterized permeation device. Also, Ion Chromotography (IC) standards will be exchanged between investigators utilizing IC aerosol composition measurement systems such as the Particle In Liquid Sample (PILS) system.

Direct comparison of measurements (pre, during, post-mission): Prior to field deployment, running instruments in the lab or in the field side-by-side is an excellent way to test performance. This is more easily done for some instruments than others, so this will be up to the various investigators to arrange as desired. During the mission joint flights of two aircraft, overflights of the ship, Ronald H. Brown, and overflights of ground sites will provide data from which instrument performance may be critically assessed. Such overflights provide an opportunity to compare a large variety of gas-phase, aerosol-phase, meteorological, and radiative parameters.

Indirect comparison of measurements (during and post-mission) While sampling in close physical proximity provides useful information, other opportunities exist to evaluate instrument performance by examining data taken during normal flight procedures. For example, the CO/CO_2 ratio should be approximately conserved for some time during transport over water, suggesting that aircraft and ship data in an urban plume might be usefully compared in this regard. Further, free tropospheric ozone levels should be comparable between the in situ and remote measurements if taken within a well-defined volume and relative short time of one another. These comparisons-of-opportunity can provide useful additional data with which to evaluate instrumental performance between the various participating platforms.

Proposed Strategy:

- *Species*: With regard to the pseudo-Lagrangian analysis, the measurements of most importance to compare include the following gas-phase oxidant and aerosol precursor and tracer species: CO, CO_2, NOx,

NOy, O_3, SO_2 and VOCs including oxygenates. For aerosols, likely comparisons include size-resolved number density and chemical composition as measured by PILS and aerosol mass spectrometer systems. Although perhaps secondary to the pseudo-Lagrangian analysis, the opportunity will be taken to compare the measurements of other important species as well, including the HOx family (OH, HO_2, and RO_2), peroxides and carbonaceous aerosol.

- *Organization:* A small group with representation from each major organizational group (NASA, NOAA, etc.) will attend to logistical details for comparisons. IGAC will play an important role in the organization of this group and its activities.

- *Formality:* After a side-by-side comparison, data sets will be reduced by the investigators and submitted independently to a referee. Data sets will include estimated uncertainties, allowing for a proper quantitative comparison. Once all data sets for a specific comparison are submitted they will be released to all study participants. Referees will encourage comparison participants to look for non-recoverable errors.

- *Develop comparison matrix:* A "transfer standard" concept will be used so that each platform is tied to the others through at least comparison with another, mutual platform. Ideally, each comparison will occur at least twice, and will be made over a range of important parameters (i.e., altitude, water vapor, possibly interfering pollutants.) Wingtip-to-wingtip aircraft comparisons, aircraft fly-bys of surface stations (ground-based or ship), and US-vs-European comparisons will all be employed.

3. Flight Coordination.

The pseudo-Lagrangian approach to trans-Atlantic transport involves three arenas of flight operations: over the source and outflow region of North America, over the mid-Atlantic and over the inflow region of Europe. Flight coordination will differ in each of these regions. On the North American side of the Atlantic, ITCT-Lagrangian-2k4 will not attempt to formulate flight plans or coordinate flights of the various aircraft. Each of the aircraft will have a variety of program goals to meet and will conduct flights according to those goals. However, these goals are such that they ensure that air masses with strong anthropogenic influence leaving North America will be well characterized during multiple aircraft flights as well as via measurements from other platforms. The responsibility of the ITCT-Lagrangian-2k4 team will be to closely monitor the flights that are made in the source region, to monitor forecast trajectories for these air masses, and to alert aircraft in the central and eastern Atlantic of possible interception opportunities. On the European side, efforts will be coordinated under the ITOP umbrella. A coordinated strategy for flight planning will be developed in collaboration with North American participants.

The ITCT-Lagrangian-2k4 team will coordinate the development of planning tools and procedures for identifying potential events that aircraft over the mid-Atlantic and Europe can mutually intercept. Analysis of trajectories and tracer calculations over periods of several years will be part of the process used to establish the most suitable locations for the mid-Atlantic and eastern Atlantic flights. The Task team will be sure that a strategy for coordinating flights is part of the overall planning for the Summer 2004 study.

4. Post-Campaign Analysis.

A combination of data analysis and modeling will be used to address the scientific objectives of individual programs and also the wider objectives of ITCT-Lagrangian-2k4. The SG will communicate with the separate science teams to assure that the Lagrangian-related data sets are available to all of the science teams involved, and they will coordinate the analysis of the data from the pseudo-Lagrangian point of view. Other data such as those collected on commercial aircraft, by sondes/lidar, and by satellites will also become part of the ITCT-Lagrangian-2k4 analysis data set where they have utility. The SG will organize joint workshops to discuss the results and identify possible papers to describe key findings.

Mega-Cities: Asia as a source of aerosols and oxidants

Contributed by **Yutaka Kondo** (kondo@atmos.rcast.u-tokyo.ac.jp), *Research Center for Advanced Science and Technology, University of Tokyo, 4-6-1 Komaba, Meguro, Tokyo 153-8904, Japan, and* **Young J. Kim** (yjkim@kjist.ac.kr), *Advanced Environment Monitoring Research Center, Kwangju Institute of Science and Technology, 1 Oryong-dong, Gwangju 500-712, Korea*

Introduction

Aerosols and oxidants are important atmospheric components both due to their impact on air quality and because they are radiatively important, affecting climate on a regional and sometimes global scale. Mega-cities are significant sources for aerosols and oxidants, so it is critical that we understand the key chemical and physical processes controlling the concentrations of these species in these regions.

To date, most large-scale, coordinated measurements of aerosols and oxidants (such as those made under the IGAC Aerosol Characterization Experiment, or "ACE", campaigns; c.f. *Quinn and Bates* article in IGACtivities No. 28) have taken place in regions well downwind of sources. Such campaigns have allowed us to characterize aerosols and oxidants after they have evolved in the atmosphere for one or more days. While this information is useful for understanding the large-scale impact of pollutants on a region, there are two weaknesses to this approach: 1) The airmasses measured during these campaigns have often mixed with natural background gases and aerosols (such as dust, sea salt, biogenic organics, and biogenic SO_4^{2-}) or are composed of pollutants from several regions, making it difficult to determine the anthropogenic fraction of what is being measured, and 2) Important chemical transformations occur rapidly after emission. It is only by making measurements near the sources that we will be able to make the link between source emissions and regional-scale air quality and climate impacts. Further, an increased understanding of aerosol and organic chemistry in mega-city regions would lead to more accurate numerical model predictions of future changes in air quality and radiative forcing in urban centers and surrounding regions.

Here we describe an IGAC Task, Mega-Cities :Asia, which will study the concentrations and chemical characteristics of aerosols and organics in a series of Asian mega-city clusters. We hope that this is the first in a series of IGAC Tasks that will study the many mega-city clusters around the globe that have a significant impact on air quality and climate and that Mega-Cities:Asia can be a model for the formation of the other Mega-city Tasks. As a start to this process, one of the sessions at the 2004 IGAC Conference in Christchurch, New Zealand (see "Announcements" at the end of this Newsletter) will be "Mega-cities". We look forward to sharing ideas and results with other researchers working on this problem and hope to use this session to initiate collaborations.

Scientific Objectives

The main objective of the Mega-cities:Asia Task is to investigate the contribution of pollutant emissions from mega-cities to local and regional including Asian Pacific air quality and to the radiation budget. Investigation of trends in emissions is also of interest. These data will be used to improve models' ability to reproduce current distributions of aerosols (inorganic aerosol, organic aerosol, and black carbon) and oxidants in Asian mega-cities and to improve their accuracy in predicting future changes in air quality. These will be achieved by:

- **Characterizing the temporal and spatial changes of aerosols, oxidants, and their precursors,** primarily by surface measurements near urban centers and their downwind (Figure 1). Temporal variations to be determined include diurnal variation; variations associated with synoptic scale disturbances; and seasonal variation.

- **Characterizing the composition, mixing state, and physical properties of aerosols in urban air.** This includes determining the hygroscopic and radiative properties of urban aerosol. This information is important for understanding the impact of aerosols on the radiation budget.

- **Validation of emission inventories of trace gases (e.g., NO_x, SO_2, NH_3, and VOCs) through comparisons of ratios of concentrations of trace species observed in urban air.**

This task is focused on the direct impacts of human activities on the atmosphere in Asian mega-cities and their surroundings. The influence of human activities

should change depending on natural conditions, including meteorological conditions, humidity, and solar radiation. These relationships will be investigated by studying seasonal variations of pollutants and the associated meteorological conditions.

We note that this task is closely related with the Atmospheric Brown Cloud (ABC) project that is currently being planned as a follow-on to the 1999 Indian Ocean Experiement (INDOEX) campaign. ABC will concentrate on regional effects on locations downwind of large sources. The Mega-cites Asia Task will be helpful to the ABC study in providing more detailed information on atmospheric composition in the source regions.

Task Activities

The first step to achieving these scientific objectives is the establishment a network of measurements in Asian mega-cities and their surroundings. To this end, the Mega-cities:Asia Task team will:

1) Generate a database of the measurements of aerosols, ozone, and their precursors that are currently being made (location, investigators, measurement methods, period, etc.) and post them on a centralized web site at the University of Tokyo. If there is a need, we will also post them on other web sites.

2) Encourage the various investigators to employ a uniform set of measurement techniques in an effort to obtain comparable data sets of high quality.

3) Inter-compare common measurements being made in different cities. Where possible directly compare instruments, using a single instrument as a "transfer standard" that is taken to each mega-city for comparison on-site.

4) Develop an implementation plan for how the various data sets from the Asian mega-cities could be combined and analyzed to deduce regional impacts.

5) Establish an explicit plan for data archiving and a protocol for making data public.

We have identified six Mega-cities or City Clusters that could be studied under this Task:

Japan: Tokyo
South China: Hong Kong-Guangzhou-Macau cluster
Taiwan: Taipei
Korea: Gwangju and Seoul
East China: Shanghai-Nanjing-Hangzhou cluster
North China: Beijing-Tianjin-Hebei cluster

For the first four of these, measurements are already planned or underway that are sufficient to act as a basis for the Mega-cities study. The studies in the South China and Tokyo region started in 2003 and so will comprise the initial studies for inclusion in the Mega-cities:Asia Task.

Details of the four planned or existing studies are given below:

a) IMPACT: Integrated Measurement Program for Aerosol and oxidant Chemistry in Tokyo

Observational Site:
Research Center for Advanced Science and Technology, University of Tokyo
Observational Periods:
Starting in April 2003
Parameters observed:
Meteorological parameters:
Temperature, winds, humidity
Radiation:
J (O^1D), J (NO_2), (spectroscopic actinic flux measurements)
Gases:
NO, NO_2, NO_y (chemiluminescence), HNO_3 (CIMS), PAN (in preparation), NMHCs, GC-FID, Oxy-Organics (PTR-MS), O_3 (UV absorption), CO (IR gas correlation), SO_2, (UV fluorescence), CO_2 (NDIR), NH_3 (in preparation)
Aerosols:
Chemical composition (aerosol mass spectrometer, ion chromatography), organic aerosols (thermal -optical method), mass loading of PM 1.0 (TEOM), size distribution (DMA).

b) Integrated Measurement Program for Aerosols and Oxidants in Pearl River Delta (Hong Kong-Guangzhou-Macau cluster)

Observational site:
a number of sites in Pearl River Delta, including Guangzhou city and rural areas
Observational periods:
starting in 2004, with pilot measurement starting in October, 2003
Parameters observed:
Meteorological parameters:
temperature, winds, humidity, radiation, light scattering
Gases:
NO, NO_2, NO_y (chemiluminescence), HNO_3 (SJAC), VOCs (GC-FID), O_3 (UV absorption),

CO (IR gas correlation), SO_2 (UV fluorescence), NH_3 (SJAC), H_2O_2 (in preparation), OH (in preparation)

Aerosols:

ion chemical composition (ion chromatography), elements (ICP), OC/EC (Sunset), organic aerosols speciation (GC/MS), mass loading of PM2.5 (TEOM), size distribution (MOUDI)

c) Aerosol studies in Taipei, Taiwan

Chemical and physical properties of aerosols have been made once per month since March, 2000 at four sites around Taipei. One is at Yangming Mountain, which is a remote mountain site (~ 1 km above sea level) north of Taipei. Measurements are also made continuously at two sites in the Taipei metropolitan area. The fourth site, Shi-men, is at the northern tip of Taiwan. This is on the windward side of the Asian winter monsoon, making it ideal for assessing the impact of long-range transport. All four sites are located at or near the Taiwan EPA's air quality monitoring stations, which can provide routine measurements of key air pollutants as well as meteorological parameters. Specific instruments at each site are listed below:

Yangming Mountain

Parameters observed:

Aerosols:

Chemical composition: Ions (Ion chromatography), elements (ICP/XRF), OC/EC (Thermal Optical Reflection), mass loading of PM 2.5 (Andersen Dichotomous sampler), mass loading of PM 10 (Andersen Dichotomous sampler)

Taipei Metropolitan sites

Parameters observed:

Aerosols:

chemical composition: sulfate (R&P 8400S), nitrate (R&P 8400N), OC/EC (R&P5400), black carbon (Magee Scientific AE-31 Aethalometer), total PAHs (EcoChem PAS 2000), mass loading of PM 2.5 (R&P 1400a), size distribution (SMPS 3936L22), size distribution (PMS PCASP-X), light-scattering coefficient (TSI 3563)

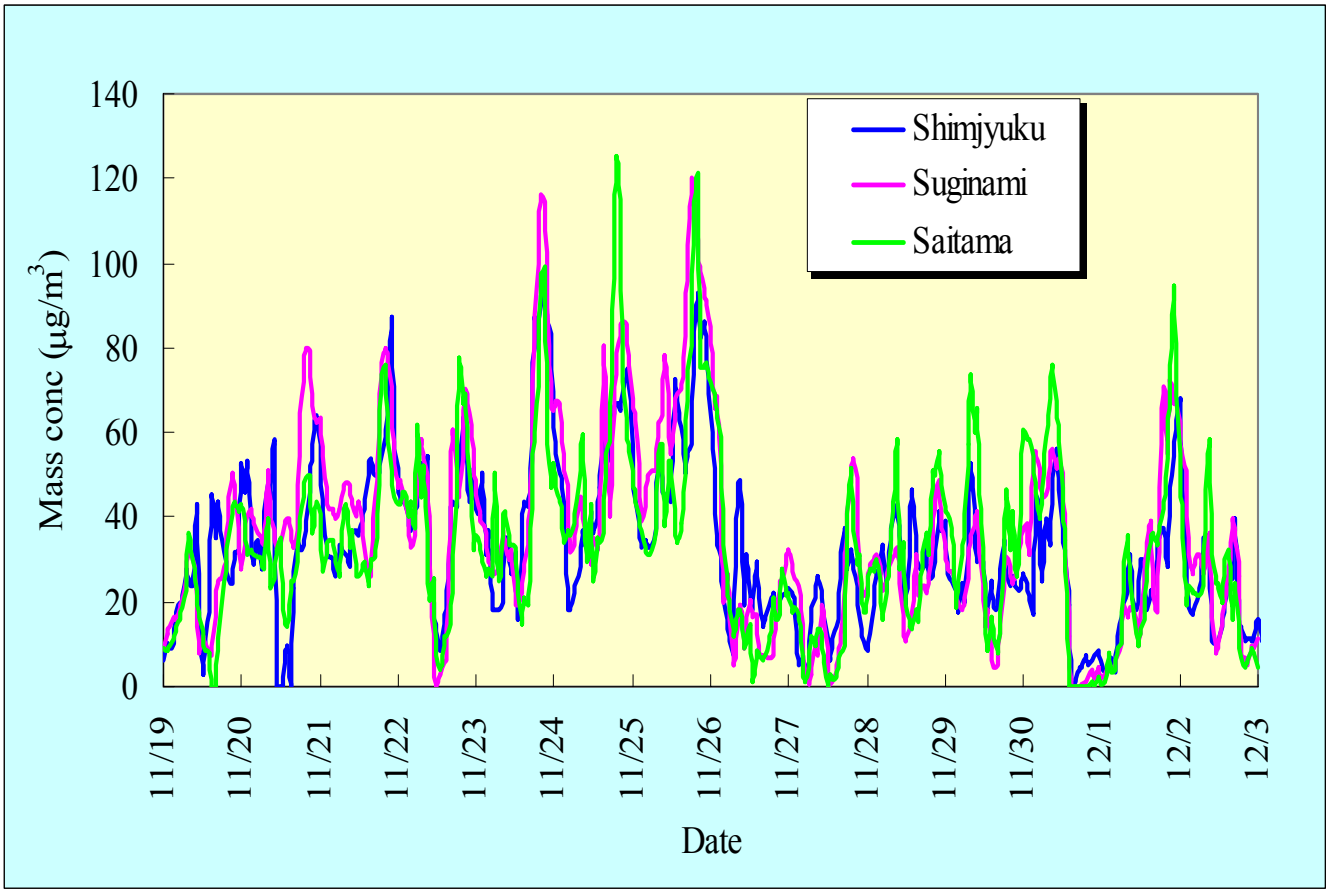

Figure 1 – Variations in the mass concentration from the Tokyo metropolitan area in 2001 are shown for stations near the center of the city (Shimjyuku), ~10km west of the city (Suginami) and ~30km north of the city (Saitama).

Shi-men site

Parameters observed:

Aerosols:

chemical composition: ions (ion chromatography), elements (ICP/XRF), OC/EC (Thermal Optical Reflectance), mass loading of PM 2.5 (R&P 2300), mass loading of PM 10 (R&P 2300 and TEOM), size distribution (MOUDI), size distribution (PMS PCASP-X), light-scattering coefficient (TSI 3563 nephelometer).

d) Aerosol studies in Korea

Possible Mega-cities or City Clusters to be studied:

Gwangju and Seoul, Korea

Period:

2003-2006

Gwangju Site (K-JIST/ADEMRC)

Parameters observed:

Aerosols:

Aerosol vertical profile (Multi-channel LIDAR), size distribution (MOUDI, aerosol profiler), ions (ion chromatography), elements (ICP/XRF), OC/EC (Thermal Optical Reflection carbon analyzer), mass loading of PM (WINS sampler, URG PM10 & PM2.5 samplers, Dichotomous sampler)

Radiation:

CIMEL Sunphotometer, MFR-7, UV-MFR-7, UV-A, UV-B

Optical measurement:

LP-DOAS (SO_2, O_3, NO_2, HONO, HCHO, BTEX), light extinction (Transmissometer), light scattering (nephelometer), light absorption (aethalometer)

Seoul Site

Parameters observed:

Aerosols:

size distribution (MOUDI), ions (ion chromatography), elements (ICP/XRF), OC/EC (Thermal Optical Reflection carbon analyzer), mass loading of PM (URG PM10 & PM2.5 samplers, Dichotomous Sampler)

Optical measurements:

LP-DOAS (SO_2, O_3, NO_2, HONO, HCHO, BTEX), light extinction (Transmissometer), light scattering (nephelometer), light absorption (aethalometer)

The IMPACT and Pearl River Delta measurements are underway and are expected to continue into 2005. Measurements in Taiwan are ongoing on a nearly continuous basis. The Mega-city Task team will coordinate joint intensive campaigns in order to characterize the regional impact of emissions from each mega-city. Because these campaigns are not fully concurrent and because they are currently being conducted independent of one another, the Mega-city Task team will host a data workshop each year that will bring these groups together. This will help ensure that measurements continue to be made in as uniform a manner as possible and will facilitate collaboration between researchers working in different mega-cities. Each study group will of course publish results as part of their individual campaigns. The Task team will make sure that, in addition, studies that integrate the various data sets will also be published.

Key to the integration of these data sets will be not only the uniformity of the measurements but also a careful assessment of the uncertainties in the measurement of key species:

Organic aerosol and black carbon - No international standard measurement techniques exist for measuring OC and EC. Thus, the University of Tokyo group will do a comparison of the different techniques being used and will transfer this knowledge to the participating groups. This effort is scheduled to commence in 2005.

NO_x, NO_y and CO – It is most important that the measurement of these two key species be carefully calibrated. There are several ways that accuracies might be improved: 1) assurance of the calibration gas; 2) proper zeroing; 3) a careful check of the conversion efficiency for NO_y.

The activities discussed above constitute the first phase of what we hope will be a multi-stage Mega-cities:Asia Task. These activities were chosen because they are achievable in a reasonable timeframe (i.e. ~3 years) and because they lay the necessary groundwork for producing a unified data set needed to analyze the impact of Mega-cities on a regional or global scale.

ANNOUNCEMENTS

The 8th Scientific Conference of the International Global Atmospheric Chemistry Project (IGAC)

September 4-9 2004
Christchurch, New Zealand

The focus of the conference will be Atmospheric Chemistry in the Environment.

Themes will include atmospheric chemistry in a variety of distinct regions such as the marine boundary layer, stratosphere, cryosphere, and urban areas as well as trans-boundary transport effects and global biogeochemical cycling. The Local Organizing Committee looks forward to welcoming you to this scientifically rewarding event held in the beautiful city of Christchurch in the spectacular South Island of New Zealand.

Abstract submission deadline: 29 February 2004

Invited Speakers will include:

- ❖Susan Solomon
- ❖"Ravi" Ravishankara
- ❖Phil Boyd
- ❖Paulo Artaxo

Sessions and conveners:

- ❖Effect of aerosols on clouds and the hydrological cycle (*Ulrike Lohmann*)
- ❖Aerosol chemistry (*Patricia Quinn*)
- ❖Climate-chemistry interaction (IGAC & SPARC related) (*Céline Mari*)
- ❖Observing the atmosphere (*John Burrows*)
- ❖Air-ice interactions including firn air and air bubble chemistry (*Eric Wolff*)
- ❖Reactive halogens in the troposphere (*Ulrich Platt*)
- ❖Atmosphere-ocean interactions (IGAC & SOLAS related) (*Mike Harvey*)
- ❖Trans-boundary transport and transformation (*David Parrish*)
- ❖Mega-cities (*Makoto Koike & Laura Gallardo-Klenner*)
- ❖Biomass burning, dust and light-absorbing aerosols (*Sandro Fuzzi*)
- ❖Toxic pollutants in the atmosphere (*Ilia Ilyin*)
- ❖Oxidizing capacity of the atmosphere (*Kathy Law*)
- ❖Emission and deposition fluxes (IGAC & ILEAPS related) (*Phil Rasch*)
- ❖Other aspects of atmospheric chemistry (*Timothy Bates*)

All information relating to the call for abstracts, registration, accommodation, deadlines, conference program etc. is available on the website:
http://www.igaconference2004.co.nz

SPARC 3rd General Assembly

*(**S**tratospheric **P**rocesses **A**nd their **R**ole in **C**limate)*
1-6 August, 2004
Victoria, British Columbia, Canada
Abstract submission deadline: 15 June 2004
http://sparc.seos.uvic.ca

SOLAS 2004 Open Science Conference

*(**S**urface **O**cean **L**ower **A**tmosphere **S**tudy)*
13-16 October, 2004
Halifax, Nova Scotia, Canada
Abstract submission deadline: 31 January 2004
http://www.uea.ac.uk/env/solas/ss04.html

2004 Gordon Research Conference

*(**B**iogenic **H**ydrocarbons and **T**he **A**tmosphere)*
2-7 May, 2004
Il Ciocco, Barga, Italy
http://www.grc.org

Please help us keep our mailing list up tp date:
_____Please note my new address
_____Please also send IGAC*tivities* to my collegue

Please send your contact information either by email or by postal mail to:
IGAC Core Project Office
NOAA-PMEL
7600 Sand Point Way
Seattle, WA 98115-6349 USA
email:igac.seattle@noaa.gov

Nam ___ Organization: _____

Stree _____

_____ State: _____ Zip: _ _

Telephone: _____ Fax: _____ E-Mail: _____

IGAC*tivities* Newsletter

Editors: Tim Bates (USA), Sandro Fuzzi (ITA), and Shaw Liu (CHN-Taipei)
Editorial manager: Doris Chen
Editing: Sarah Doherty
IGAC logo: Linda Kubrick

Published by The IGAC Core Project Office
RESEARCH CENTER FOR ENVIRONMENTAL CHANGE
ACADEMIA SINICA
128 Academia Rd. Sec. 2
P.O. Box 1-55 NanKang
Taipei, 11529 Taiwan

Tel: (+886+2) 2783-9910 Ext. 280
Fax: (+886+2) 2783-3584
E-mail: igac.taipei@earth.sinica.edu.tw

中華郵政台北誌字第 137 號執照登記為雜誌交寄
發行人：劉紹臣
發行所：中央研究院環境變遷研究中心
發行地址：台北市 115 南港區研究院路二段 128 號 1-55 號信箱

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