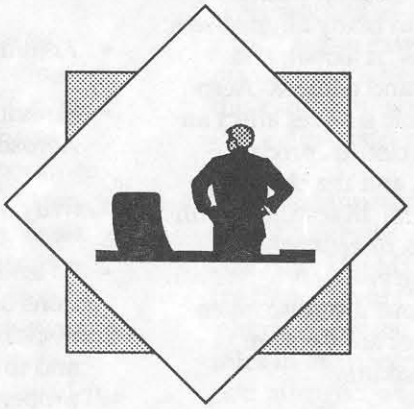
IGACtivities NewsLetter
of the International Global Atmospheric Chemistry Project

Issue No. 4, March 1996

In this issue



A Note From the IGAC Chair

Guy Brasseur

I would like to begin my first "Note from the Chair" by thanking Prof. Ron Prinn, who recently stepped down as Chair of the IGAC Scientific Steering Committee, for having led the Project so enthusiastically since its creation seven years ago. Ron's role in the development of IGAC, and in the planning and implementation of its successful Activities has been essential.

I am very honored to be Ron's successor. I already realize that the task will be extremely challenging, but I also know that with the support and help of the outstanding scientists who serve on the Steering Committee, and with the energy and dedication of our Activity Conveners and their committees, IGAC will address new interdisciplinary questions and accomplish exciting science. Most IGAC Activities are now underway, some early results have already been published and more efforts, including field campaigns, data analysis, and global modeling, will take place in the near future. You will read about many of them in future issues of IGAC*tivities*.

I very much hope that IGAC will be perceived increasingly as an organization that directly and efficiently serves the needs of individual scientists, research institutions and funding agencies. The Project will do the best it can within its means to facilitate exchanges of information between scientists at the international level, provide to the atmospheric-biospheric chemistry research community a forum for discussion of its scientific priorities, and help communicate the results of sometimes complex scientific investigations to decision makers. In this vein, this issue of IGAC*tivities* contains several articles related to aerosol science. After reading them I hope that you will agree that the new IGAC *Focus on Atmospheric Aerosols* provides a unique opportunity for linking microphysical and chemical approaches for studying aerosols in the context of global change.

GLOBAL CHANGE
IGBP

Introduction

Atmospheric Aerosol Research in IGAC

Contributed by P.V. Hobbs, University of Washington, USA, and A.Pszenny, IGAC Core Project Office, USA

Although aerosols are only a minor constituent of the earth's atmosphere, their importance is amplified by their ubiquity, the roles they play in many atmospheric processes, and their effects on local, regional, and global air quality and on weather and climate. Aerosols from natural and anthropogenic sources affect air quality, visibility, acid deposition, clouds, precipitation, the earth's radiation balance, and the chemistry of the troposphere and stratosphere. In common with trace gases, aerosols do not respect international boundaries. However, aerosols are very inhomogeneously mixed throughout the atmosphere. Thus, studies of the global effects of aerosols are inherently an international undertaking.

An understanding of atmospheric aerosols requires concerted efforts from many branches of atmospheric research, including gas, liquid and solid phase air chemistry—to describe the formation of particulate matter in the atmosphere; aerosol physics—to quantify the dynamic evolution of aerosol size distributions and their physical properties; cloud physics and cloud chemistry—to understand how aerosols affect clouds and are affected by clouds; radiative transfer theory—to describe the interaction of aerosols with radiation; and climate research—to incorporate the direct and indirect effects of aerosol forcing into climate models. Consequently, studies of aerosols cross many subdisciplines within the atmospheric sciences.

In recognition of the above facts, the decision was made in early 1995 to merge the International Global Aerosol Program (IGAP) with IGAC to form a new IGAC *Focus on Atmospheric Aerosols (FAA)*. This merger has enhanced significantly IGAC's potential to undertake a comprehensive research program on all aspects of atmospheric aerosol research. However, in view of the urgency to elucidate and quantify the role of aerosols in the earth's radiation balance, the initial principal objective of the FAA is "To improve understanding of the role of atmospheric aerosols in climate forcing and in the prediction of changes in global climate and geospheric-biospheric processes".

An implementation plan for the FAA is nearing completion and should be available later this year. Current plans call for the FAA to consist initially of four new IGAC Activities:

- Activity 8.1: Aerosol Characterization and Process Studies (ACAPS).
- Activity 8.2: Direct Aerosol Radiative Forcing (DARF).
- Activity 8.3: Aerosol-Cloud Interactions (ACI).
- Activity 8.4: Stratosphere and Upper-Tropospheric Aerosols (SUTA).

Activity 8.1 (ACAPS) has three main goals:

- To understand the spatial and temporal distributions of the chemical, physical, radiative and cloud nucleating properties of the atmospheric aerosol and to investigate the relationships between these properties.
- To determine the chemical, physical, and biological processes controlling the formation and fate of aerosols and how these processes affect the number size distribution, the chemical composition, and the radiative and cloud nucleating properties of aerosols.
- To quantify the role of aerosol-related heterogeneous processes in gas phase atmospheric chemistry and the major biogeochemical cycles.

The main goal of Activity 8.2 (DARF) is to:

- Determine, primarily through observations, the magnitude, uncertainty, chemical sources, and temporal and spatial variations of the direct radiative climate forcing by aerosol of various types (e.g., sulfates, organics, mineral dust).

The principal goals of Activity 8.3 (ACI) are:

- To improve understanding of the relationships between the physical and chemical properties of aerosols and the microphysical and radiative properties of the clouds that form on them.
- To explore the relationships between the physical and chemical properties of clouds and the radiative properties of the aerosols that are processed or formed by clouds.

The goals of Activity 8.4 (SUTA) are to answer the

following questions:

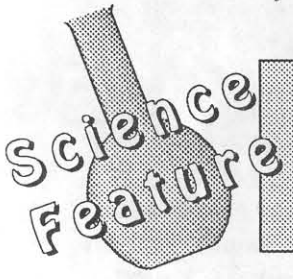
- What are the dispersal and decay mechanisms for stratospheric aerosols?
- Are stratospheric aerosol properties adequately characterized, especially their size?
- Can stratospheric aerosols serve as nuclei for cloud formation in the upper troposphere and stratosphere?
- Is there a "background" stratospheric aerosol, and what is its source or sources?
- Are the impacts of volcanic emissions on the stratosphere adequately characterized?
- Are stratospheric aerosols frozen or do they exist as supercooled liquid particles at low temperatures?

The FAA will also involve the following on-going aerosol-related IGAC Activities, which will be augmented where necessary to achieve FAA objectives:

- IGAC Activity 6.6: "Global Integration and Modeling (GIM)." The former IGAP Science Project entitled "Modeling the Effects of Aerosols on Radiative Forcing and Climate (MEARC)" will be a new Task under IGAC Activity 6.6.
- IGAC Activity 2.3: "Biomass Burning Experiment: Impact on the Atmosphere and Biosphere (BIBEX)." The former IGAP Science Project entitled "Biomass Burning Aerosols in the Tropics: Impact on Radiation Budget and Climate (BATIR)" will be an identifiable new component within BIBEX.
- IGAC Activity 3.1: "Polar Atmospheric and Snow Chemistry (PASC)." The former IGAP Science Project entitled "Sulfur and Carbonaceous Aerosols in the Arctic and their Radiative Forcing (SCAARF)" will be incorporated into PASC.

In support of the FAA, the IGAC Scientific Steering Committee has endorsed the following international field campaigns: *ACE-1*, which was carried out over the Southwest Pacific Ocean in late 1995, the *Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX)*, to be carried out off the East Coast of the United States in summer 1996, and *ACE-2* to be carried out over the Northeast Atlantic Ocean in summer 1997.

In this issue of IGAC*tivities* we present an early report of some of the results from ACE-1, and a summary of the plans for TARFOX. These features are preceded by a synopsis of highlights from an earlier IGAC aerosol research campaign: ASTEX/MAGE. Together these articles illustrate how IGAC research builds on past knowledge and experience, and why large, internationally coordinated programs are needed to answer many fundamental questions about global climate change.



Sorting Out Chemistry and Dynamics: The ASTEX/MAGE Experiment

Contributed by B.J. Huebert, University of Hawaii, USA

We've all had similar experiences: your daughter received her college diploma, but you can't even identify her in the photo because she was moving just as the shutter opened. Blurring eliminated the critical information from the picture. Exactly the same thing happens when you try to study aerosol dynamics in the atmosphere, since everything is moving. The air you want to take a picture of is advecting downwind, material is being added and removed at the surface, entrainment is mixing in different air from above, dispersion is causing horizontal mixing, and vertical wind-shear is slowing down air at the surface. How can you hope to tease out the oxidation rate of sulfur dioxide or the source strength of ammonia from such a cauldron? That's the challenge ASTEX/MAGE scientists undertook in the marine atmosphere near the Azores in 1992.

The Atlantic Stratocumulus Transition Experiment (ASTEX), one of the second series of FIRE international cloud-climatology experiments, took place in June of 1992 in the stratocumulus-capped marine boundary layer. The primary purpose of ASTEX was to study the factors influencing the formation and dissipation of marine clouds. The chemical experiment within ASTEX was organized by IGAC's *Marine Aerosol and Gas Exchange* (MAGE) Activity. Its objective was to study air-sea exchange and the formation and transformation of marine aerosols, in part by making

Lagrangian observations (moving the measuring systems to stay with the same air). Chemical instrumentation was deployed on two islands, one French and two U.S. ships, and three aircraft from the U.S. and U.K. (Huebert et al., 1996). Although the meteorological situation was complex, chemists benefited from the observations of dozens of other groups who were studying cloud physics, boundary layer dynamics, and radiative transfer (Albrecht et al., 1995). These ASTEX scientists characterized turbulent and large-scale air motions in and above the boundary layer, thus enabling MAGE scientists to quantify the impact dynamics had on chemical concentrations.

To study marine chemical processing, we developed a Lagrangian sampling strategy for repeatedly studying the same air parcel over a two-day period. This approach relied on the release of constant-density balloons which floated downwind with the parcel and radioed their GPS-derived locations to a relay of sampling aircraft. The balloons served as markers for the boundary layer airmass, which was continually being modified by chemical and energy fluxes at the surface, entrainment of free tropospheric air, wind shear within the boundary layer, horizontal dispersion, chemical reactions, and aerosol transformations.

Unfortunately, the balloons also served as excellent drizzle detectors, since a 0.1 mm thick layer of water deposited on their 1 m^2 upward facing surfaces (100 grams) was enough to drive them into the ocean. None of the balloons survived more than 7 hours in our first attempt. The second try was more successful: during the second Lagrangian (L2) one balloon (#7) remained aloft for over forty-two hours, seven research flights, and two sets of observations by ships (Businger et al., 1996; Huebert et al., 1996). Incidentally, the popular hypothesis that #7's success was due to the fact that it was the only balloon with a happy face drawn on it was recently disproved in ACE-1 (see following article by T.S. Bates and J.L. Gras), where three faceless "smart" balloons designed to adjust their own buoyancy were followed for 2 days by NCAR's C-130 aircraft.

While it is an oversimplification to say we studied "the same air" over a two day period, moving with the wind allowed us to see how dynamics modified a parcel of air. Conditions near the Azores made our job harder: the 2 km deep boundary layer was frequently separated into several sublayers, each of which had to be characterized. Mixing between layers and dilution by entrainment of free-tropospheric air were often the major causes of concentration changes. Using an entrainment velocity of 0.6 cm s^{-1}, Bretherton et al. (Bretherton and Siems, 1995) computed a time scale of about 4 days for replacing the L2 boundary layer with free-tropospheric air. Chemically this usually meant that our wet, aerosol-laden boundary layer air was diluted by drier, cleaner air from above. However, late in L2 a dusty Saharan airmass passed over us, entraining large mineral aerosols into the cloud layer. This addition of large aerosols produced a noticeable modification in the microphysics and dynamics of the cloud field (Martin et al., 1995), creating larger droplets and increasing the likelihood of drizzle. We could not have sorted this out from Eulerian observations alone.

We were also able to derive the sea-to-air flux of ammonia (NH_3) vapor from studying the NH_3 budget during L2 (Zhuang and Huebert, 1996). Biologically, this NH_3 emission represents an unexpected loss of a limiting nutrient from the surface ocean. It is also important from an atmospheric standpoint, because the climatic and visibility impact of sulfuric acid droplets change dramatically when they are converted into ammonium salts. The ammonium/non-seasalt sulfate (NSS) ratio in this polluted European airmass increased with time, while the total NSS decreased. Several terms (all in units of micromoles (μmol) NH_3 m^{-2} day^{-1}) dominated the budget (Figure 1): wet and dry deposition could have removed -4, while dilution by entrainment corresponded to a loss of -29, for a net removal flux of -33. Since the observed mixed layer concentration change (the net effect of all fluxes) was only -7, sea-to-air exchange must have provided a flux of +26 μmol NH_3 m^{-2} d^{-1}. Although substantial uncertainties remain, particularly in the estimation of the entrainment term, the Lagrangian strategy allowed us to measure enough of the air-motion terms to sort out a surface flux.

Chemical reaction rates were also determined by this approach. A group from the University of California at Irvine used repeated Lagrangian observations from a single flight (during which the plane was advecting with the balloons) to derive information on oxidation by free radicals (Wingenter et al., 1996). They used measurements of hydrocarbons and halocarbons versus altitude to characterize the impact that dilution by entrainment would have on concentrations. Then they employed the differing reactivity of several species to attack by hydroxyl and chlorine radicals and the observed concentration changes with time to solve for the concentrations of each radical. Significant levels of chlorine radicals could explain why several species disappear faster than expected from the marine atmosphere, but the attempts to directly measure their concentration have been controversial. Thus, this indirect observation of their importance is a big step forward. Here again, the Irvine group's ability to quantify the impact of mixing was crucial for separating out the changes caused by these oxidants.

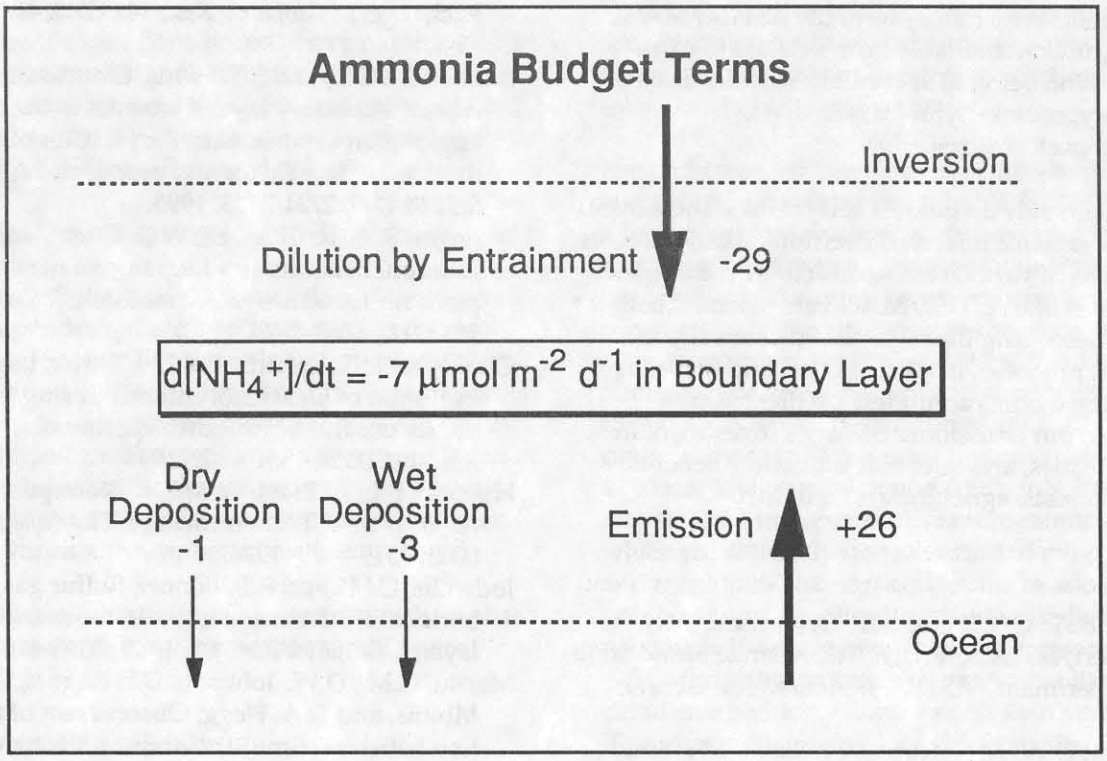


Figure 1. Schematic of the ammonia budget for the marine boundary layer during the second ASTEX/MAGE Lagrangian experiment.

Another group reversed the process and used aerosol measurements to derive exchange rates between the various layers of the decoupled boundary layer (Clarke et al., 1996). They confirmed that the dynamically-derived entrainment velocity of 0.6 cm s^{-1} for the main inversion was consistent with a simple aerosol mixing model, and concluded that the surface mixed layer entrained cloud-layer air with an effective entrainment velocity of 0.45 cm s^{-1}. They also identified a method whereby the ratio of volatile to nonvolatile nuclei can be used to characterize mixing between air masses with different histories. This is one of many examples in which chemical and aerosol measurements were able to constrain dynamics and provide the meteorological investigators with information they could not have derived from their usual suite of observations. Clarke's group also used the Lagrangian observations to demonstrate that no aerosol nucleation had occurred in the marine boundary layer during the course of L2.

The various platforms played complementary roles. Eleven-hour impactor samples from Santa Maria Island proved to be important for estimating particle removal rates. The aircraft were able to gather vertical profiles and keep up with the tagged air masses. The two ships made unique contributions because of their ability to move to locations of interest (like the starting and ending points of the Lagrangians), stay on station around the clock, make measurements very close to the surface, and support instruments with long sampling times. Jodwalis and Benner (1996) demonstrated that a new variance method can be used to measure air/sea sulfur fluxes, based on a fast total gaseous sulfur detector. In view of the need for ways to test the wind speed-based parameterizations of dimethylsulfide (DMS) emissions from which most submicrometer marine aerosols are derived, this is a valuable addition to our arsenal. The variance method generally found larger fluxes than estimates derived from simultaneous measurements of DMS in the water and air (Blomquist et al., 1996). From the other ship, Putaud and Nguyen (1996) used measurements of DMS concentration gradients to estimate fluxes. These complementary approaches improve our ability to derive a consensus among flux estimation techniques.

Of course, these examples are just a small part of what was learned about aerosols and their source materials during the ASTEX/MAGE program. A collection of MAGE papers has been published in the February 1996 issue of the Journal of Geophysical Research - Atmospheres and is available as a compilation from this author (huebert@soest.hawaii.edu). Most of the more dynamically-oriented ASTEX papers, many containing analyses based on the Lagrangian observational strategy, are contained in the August 15, 1995 issue of the Journal of the Atmospheric Sciences. As with all field programs, there is still much to be

learned from further analysis of the ASTEX/MAGE data set (publicly available from a database maintained by John Seinfeld at the California Institute of Technology; contact Lynn Russell, lynn@aeolus.che.caltech.edu).

Policymakers rely on models of dynamics and aerosol chemistry to make informed decisions about the costs and benefits of various emissions control strategies. Experiments like ASTEX/MAGE are essential both for properly describing the physics and chemistry of individual processes in these models and for seeing whether the models accurately predict nature's response to our emissions. Society's investment in ships, airplanes, and scientists ultimately benefits fisheries, forests, agriculture, and industry.

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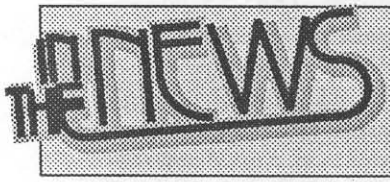

IGAC's *First Aerosol Characterization Experiment* (ACE-1) Field Program Completed

Contributed by T.S. Bates, NOAA Pacific Marine Environmental Laboratory, USA and J.L. Gras, CSIRO Division of Atmospheric Research, Australia

Hypothesis testing is the acknowledged cornerstone of scientific research, and nowhere could the testing of hypotheses be more critical than in assessing the future climate of our planet and man's impact on that future. Whilst the past couple of decades have seen an explosion in our ability to generate predictive climate models, in a number of critical areas this has not been matched by a corresponding ability to adequately describe our physical and chemical environment. In one particular area, the impact of aerosol particles, this lack of knowledge severely limits our ability to reduce the modeled uncertainty to an acceptable level.

The basic processes whereby aerosol particles affect climate are reasonably well known: direct effects are the result of scattering and absorption of solar radiation by particles whereas indirect effects are primarily those due to modification of cloud optical properties by particles that serve as nucleation sites for cloud droplets. Based on what are arguably the best model estimates for aerosol effects on climate (IPCC, 1996), the direct effect of anthropogenic aerosols causes a climate forcing of about -0.5 W m^{-2} whilst the indirect effect through cloud modification is less certain but of similar magnitude. Reducing this uncertainty is

imperative for models to accurately predict future climate scenarios.

To a substantial degree these uncertainties are the result of inadequacies in our knowledge of the global distribution of aerosol properties and the processes that control these properties. Aerosol particles are intrinsically difficult to study and to categorize. They have a multitude of sources from both natural processes and nearly all human activities. Unlike gases such as carbon dioxide (CO_2) where one molecule behaves exactly like every other molecule, aerosol particles have a continuous spectrum of sizes from a few nanometers to tens of micrometers. Their chemical composition, radiative importance and cloud nucleating ability all vary with size. To complicate matters, there is continuous evolution of the spectrum of sizes as aerosol particles respond to and modify their environment.

The function of climate models is to calculate regional or global climate forcing due to a particular entity (e.g., CO_2, methane, sulfate aerosol particles). What perhaps is often overlooked in this rush to calculate climate forcing is that models themselves are based on a series of hypotheses that may or may not always be well founded. These include hypotheses regarding our ability to accurately describe the important features of the existing atmosphere. From an aerosol perspective this includes our ability to describe accurately as a function of space and time those aerosol properties that are important directly in the global radiative balance or those properties which may alter the optical properties of clouds. It also includes the presumption that we can accurately model the radiative and cloud nucleating effects of particles in the real world, not to mention seemingly simple atmospheric processes which generate significant variability on scales the models cannot resolve. To this point in time, for example, simply understanding the processes that control particle production and accurately modeling the rate of particle production have been singularly elusive. Challenging these hypotheses requires observational data that verify and expand existing aerosol climatologies and allow direct evaluation of parameterizations used to calculate radiative properties and effects. More realistic descriptions of aerosols in climate models require a better understanding of the processes that determine the size distribution, chemical composition and state of mixing of the atmospheric aerosol.

Reducing the uncertainties in estimates of aerosol forcing of climate requires a global perspective that can only be achieved by combined international efforts in an interdisciplinary and systematic approach. The *Aerosol Characterization Experiments* (ACE), which are a component of IGAC's *Focus on Atmospheric Aerosols*, are designed to investigate aerosol properties and processes in key areas of the globe. The complexity of these processes requires a combination of atmospheric chemistry, aerosol physics, atmospheric radiation, cloud microphysics, oceanography, biology, meteorology and boundary layer dynamics. The large number of research platforms and investigators needed for these experiments requires an international effort.

ACE-1 was the first of a series of these studies and was focused on the minimally-polluted marine atmosphere. This environment provided an opportunity to study the chemical, physical, radiative and cloud nucleating properties of the natural aerosol system and thus provided a background from which to compare and quantify any anthropogenic perturbation. The experiment took place from October to December of 1995 and included transects across the Pacific Ocean and a month long intensive campaign south of Australia. The experiment involved the efforts of over 100 research scientists from 11 countries and coordinated measurements from the NCAR C-130 aircraft, the NOAA research vessel *Discoverer*, the Australian fisheries research vessel *Southern Surveyor*, and land based stations in New Zealand and at Cape Grim and Macquarie Island, Australia (see Figure 2).

Although the ultimate success of the experiment can only be fully gauged after a more complete data analysis period, it is very clear from the preliminary analyses that most of the experiment's initial objectives were met or exceeded. Some of the initial highlights of the experimental results are described below. A more detailed description of ACE-1 can be found on http://saga.pmel.noaa.gov.

- Measurements of the chemical, physical, radiative and cloud nucleating properties of the aerosols were made using state of the art instrumentation in a wide range of environments. This included mainly the background marine atmosphere, but also volcanic plumes of Kilauea, Hawaii and Mt. Ruapehu, New Zealand, and some biomass burning-and anthropogenically-affected air masses from Australia. These data will now form the basis for closure studies that assess our ability to calculate radiative and cloud nucleating properties of the aerosol from its fundamental properties, the chemical and physical size distributions. This detailed data set will also be used to develop and test aerosol parameterizations in regional and global climate models.
- One of the more exciting aspects of this type of experiment is the real-time observations that directly address current hypotheses - for example where new particle production occurs in the

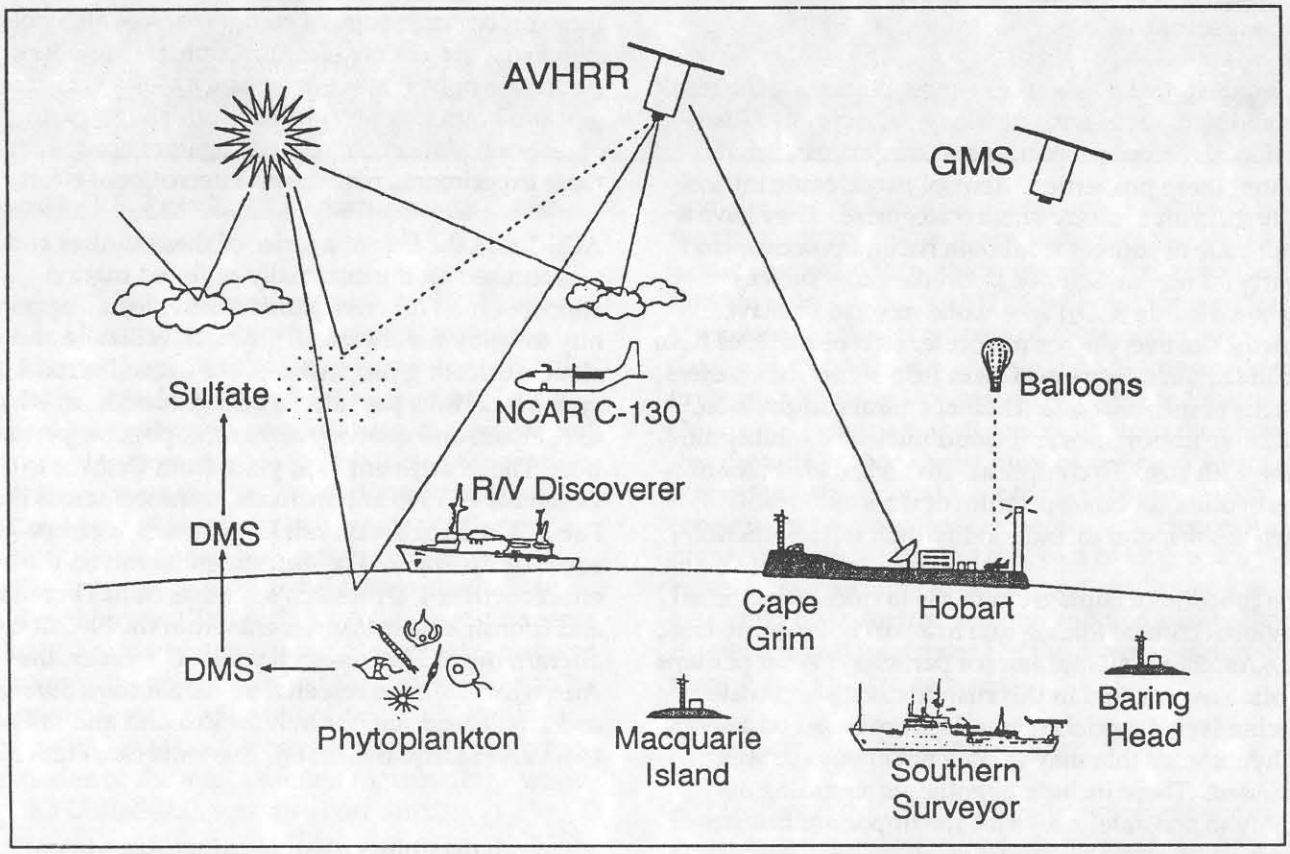

Figure 2. Schematic representation of the ACE-1 intensive study area and measurement platforms.

atmosphere. Simultaneous measurements during ACE-1 of both the precursor gases (sulfur dioxide (SO_2), sulfuric acid and ammonia) and ultrafine particle concentrations confirmed the formation of new particles in the outflow from cumulus clouds that has been observed elsewhere (e.g., Radke and Hobbs, 1991; Perry and Hobbs, 1994). New particles were also observed to mix into the marine boundary layer after cold frontal passages and during periods of synoptic-scale subsidence and convective mixing between the free troposphere and the marine boundary layer. The ultrafine particles were regularly observed during these meteorological events on *Discoverer* (see Figure 3) and at Cape Grim and Macquarie Island. It is clear from these data that particle formation occurs on large spatial and vertical scales and is not confined to regions of precursor gas emissions.

- Detailed measurements were made of the biological and chemical characteristics of the water masses south and west of Tasmania to assess both the rate and amount of dimethylsulfide (DMS) released from the ocean to the atmosphere and the processes controlling the concentrations of DMS.

- The value of Lagrangian studies for deconvoluting the effects of dynamics and chemistry has been shown clearly by the ASTEX/MAGE experiment (see preceding article by B.J. Huebert). Whilst such an experiment in the "roaring forties" where wind and weather conditions are known for their severity must be considered high-risk, the potential pay-off is large. With the aid of outstanding meteorological support from the Australian Bureau of Meteorology two successful Lagrangian experiments were conducted during ACE-1. As with the ASTEX/MAGE studies, the data will be used to quantify the rates and efficiencies of sulfur gas oxidation and the evolution of the aerosol over the two-day periods of these experiments. Additional shipboard measurements of DMS and SO_2 diurnal cycles will be used to constrain oxidation rates and efficiencies.

- Clear-sky, column-integrated particle scattering experiments were carried out over a wide range of atmospheric optical depths by including not only background marine atmosphere near Tasmania but the volcanic plume of Kilauea.

The first ACE-1 data workshop is scheduled for 24-28 June 1996 to review preliminary results from the

experiment and to integrate the combined data set. The understanding gained in ACE-1 will be used to study progressively more complex environments. The Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX, see following article by P.B. Russell) will focus on clear-column closure experiments off the east coast of the U.S. in July 1996. ACE-2 will extend the process and closure studies to the eastern North Atlantic Ocean in June/July 1997 and focus on the anthropogenic aerosols from the European continent and desert dust from the African continent.

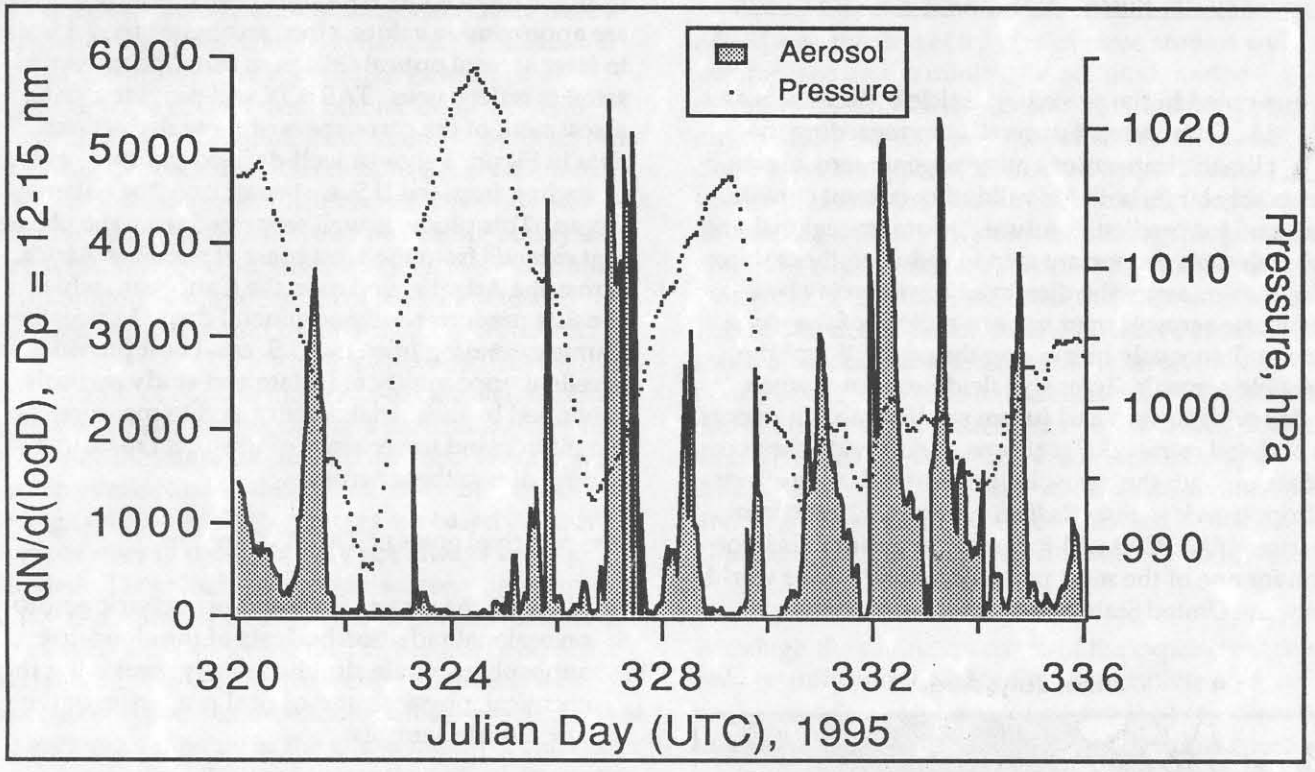

Figure 3. Particle number concentration (12 < Dp < 15 nm) and barometric pressure for a 15-day period during the ACE-1 intensive. (Data collected aboard *Discoverer* by V. Kapustin.)

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THE NEWS: Next Step: The *Tropospheric Aerosol Radiative Forcing Observational Experiment* (TARFOX)

Contributed by P.B. Russell, NASA Ames Research Center, USA

As noted in the preceding article by T.S. Bates and J.L. Gras, current uncertainties regarding the climatic impacts of anthropogenic aerosols are unacceptably large, both for validating current climate models and for predicting future climate on regional and global scales. An important step in reducing these uncertainties is to measure the direct radiative forcing by tropospheric aerosols over various regions of the globe while simultaneously measuring the properties of the responsible aerosols. Intensive field programs, when extended with current and future satellite measurements and validated retrieval algorithms, can provide the necessary data on both the aerosols and their radiative effects. The Tropo*spheric Aerosol Radiative Forcing Observational Experiment* (TARFOX) will focus on providing this information for one of the most polluted regions of the world, namely, the United States eastern seaboard.

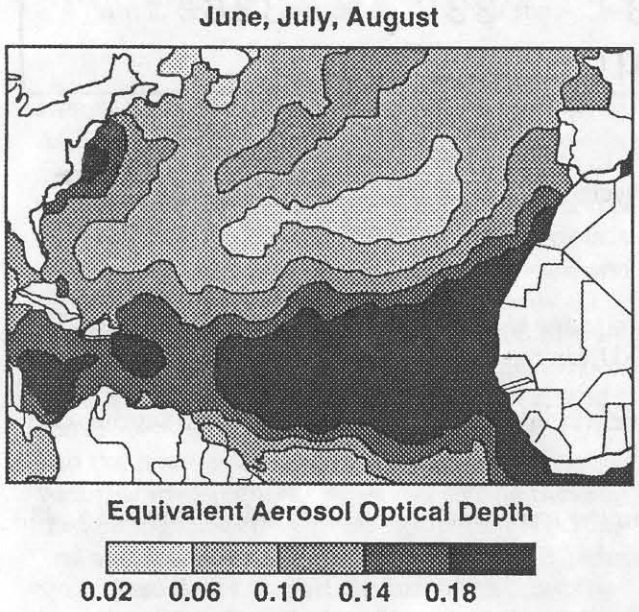


Figure 4: June/July/August map of aerosol optical depth derived from NOAA/AVHRR satellite reflectance data over the oceans (Husar and Stowe, http://capita.wustl.edu/CAPITA/CapitaReports/TropoAerosol/trop2.html).

To illustrate the coverage possible with satellite remote sensing of aerosol properties, Figure 4 shows a map of the mean June/July/August aerosol optical thickness derived from the NOAA/AVHRR operational product. These are approximate values, since an aerosol model is used to infer aerosol optical thickness from the measured satellite reflectances. TARFOX will provide a critical assessment of the correctness of these deductions. The data in Figure 4 show a well-defined plume of aerosol extending from the U.S. east coast over the Atlantic Ocean. This plume is well separated from the plume that extends from the west coast of northern Africa, across the Atlantic, and over the Caribbean, which consists predominantly of mineral dust. Hence, the plume extending from the U.S. east coast provides an excellent opportunity to isolate and study aerosols generated by industrial activity, and to measure the magnitude and uncertainty of the direct radiative forcing due to these aerosols.

The principal goals of TARFOX are to:

- Measure the direct effects of tropospheric aerosols on regional radiation budgets of the cloud-free atmosphere, while simultaneously measuring the chemical, physical, and optical properties of the responsible aerosols.

- Perform a radiative "column closure" study in one of the most polluted regions of the globe - the U.S. eastern seaboard. Use the degree of closure (i.e., consistency) among different measurements and analyses of aerosol properties and effects to (1) assess and reduce uncertainties in predicted aerosol climate effects, and (2) guide planning for future aerosol studies (such as ACE-2).

- Extend these measurements and analyses to other periods and regions with similar aerosol properties, using validated satellite remote sensing methods.

To achieve these goals, the following principal questions have to be answered:

- What are the perturbations in the radiation budget in regions strongly affected by anthropogenic aerosols?

- Do calculations based on *in situ* and remote measurements of the properties of these aerosols agree with the measured perturbations?

- What aerosol properties (e.g., chemical composition, size distribution) primarily control the perturbations?

Answering the questions posed above requires an extensive theoretical radiative transfer modeling effort, an intensive field measurement program, and integrated analyses. The specific implementation tasks of TARFOX are discussed below.

Task 1. Radiative Forcing Sensitivity Studies

An accurate theoretical determination of the wavelength-dependent, direct radiative forcing due to tropospheric aerosols in a vertically inhomogeneous, absorbing and scattering atmosphere, must be carried out. These investigations should employ standard meteorological profiles together with a compilation of various natural and anthropogenic aerosol optical properties to examine the sensitivity of the radiative fluxes at various atmospheric levels to:

- Aerosol single-scattering parameters (such as phase function, single-scattering albedo, and optical depth)
- Vertical profiles of aerosols
- Spatial (e.g., latitudinal) and temporal (e.g., seasonal) variations due to changes in solar insolation, aerosol characteristics, and surface albedo

These computations should use measured aerosol radiative properties. There may be limitations in their use, but identifying these will help define the types of measurements that must be carried out in field programs. These same computations will be used to complete column-closure experiments by comparing the radiation perturbations measured at the top, bottom, and other levels of the atmosphere with calculations based on *in situ* and remote sensing measurements of the properties of the aerosol in the atmospheric column.

Task 2. Field Measurements

The direct, clear-sky radiative forcing (i.e., change in net radiative flux) caused by aerosols is related to the aerosol optical thickness through such factors as the underlying surface albedo and the aerosol chemical composition and size distribution. This forcing can be measured by surface and airborne radiometers and derived from satellite-measured radiances using retrieval algorithms. Similarly, optical thickness can be measured by surface and airborne photometers and radiometers, and also derived from satellite-measured radiances. Regressing measured radiative flux changes versus measured optical depths will provide an empirical measure of the sensitivity of radiative forcing to aerosol optical thickness. An important TARFOX objective is to compare this empirical sensitivity to results of computations that use aerosol chemical, physical, and optical properties obtained from simultaneous measurements.

Figure 5 (see next page) summarizes the platforms and measurements planned for TARFOX. Because the effects of aerosol scattering on upwelling radiative flux are generally stronger over low-albedo surfaces, the satellite and aircraft measurements will focus on ocean areas, while also tying in the more continuous measurements at nearby land sites. To observe the strongest possible effects of anthropogenic aerosols, the field study will be conducted off the U.S. east coast, in the area of the aerosol plume shown in Figure 4. The intensive field period (IFP), July 10-31, 1996, was selected using several years of satellite imagery, which show that this is the period of minimum clouds and maximum haze optical depth. In addition to the polar-orbiting satellites listed in Figure 5, TARFOX will use half-hourly images from the geosynchronous GOES-8 satellite to define regions most likely to be cloud-free and hazy during polar satellite overpasses. This information will be used in directing TARFOX aircraft, in selecting periods for intensive ground observations, and in post-mission analyses. The primary aircraft base for TARFOX will be the NASA Facility at Wallops Island, Virginia, with the possibility of some flights to Bermuda.

Daily satellite measurements during the IFP may provide sufficient aerosol variability to infer the background (natural) tropospheric aerosol optical thickness from the minima observed. The difference between the IFP mean aerosol optical thickness and the background value, when multiplied by the sensitivity factor described above, will yield a regional estimate of tropospheric aerosol radiative forcing for these summertime conditions. However, ascertaining what portion of this forcing is due to anthropogenic aerosols will require careful analysis of other simultaneous data on aerosol properties, precursor gases, tracers, radiatively active gases (e.g., water vapor), and surface albedo. Also, independent measurements of optical depths and radiative fluxes will be required to test and improve the satellite retrievals.

The airborne and surface measurements are designed to provide this simultaneous information. The imaging spectrometers on the ER-2 (MAS and AVIRIS in Figure 5) will provide retrievals of aerosol and surface properties with finer spatial, temporal, and spectral resolution than the satellite measurements. The ER-2 lidar (LASE in Figure 5) will provide vertical profiles of water vapor, aerosols, and clouds from the surface to 20 km. Flights of the medium- and low-altitude aircraft (C-130, C-131A, Pelican) will be

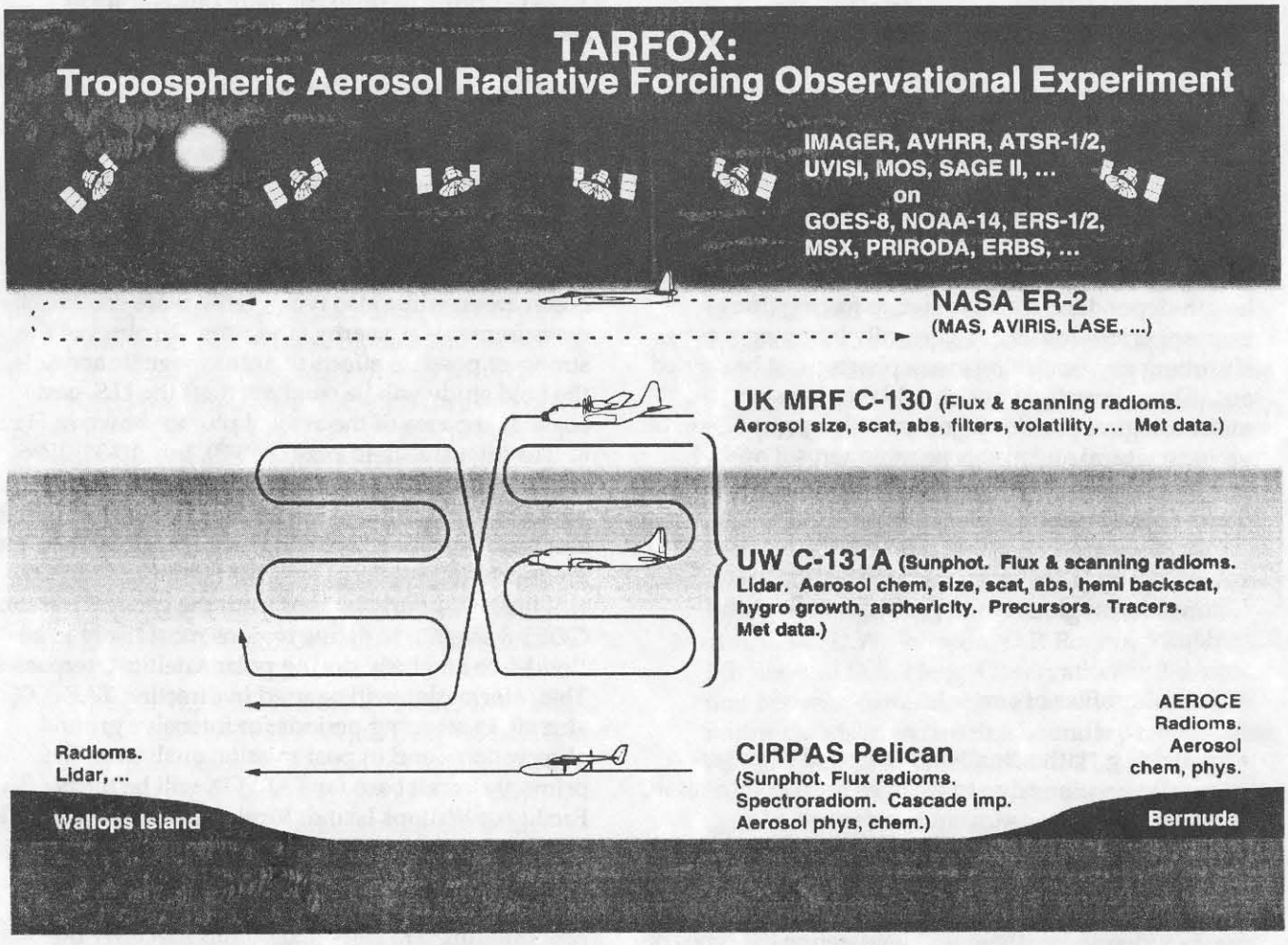


Figure 5: Schematic overview of TARFOX platforms and measurements.

embedded in these lidar vertical profiles. The lidar profiles will show the vertical context (including any unsampled aerosol or cloud layers) of *in situ* samples and radiative flux and optical depth measurements.

Figure 5 also summarizes the measurements to be made by each of the medium- and low-altitude aircraft. Flights will be directed to satellite-determined hazy, cloud-free regions, or will span contrasting hazy and clean areas. Careful flight coordination will permit: (1) simultaneous radiative flux measurements above and below aerosol layers while the layers themselves are sampled, (2) simultaneous *in situ* aerosol measurements at different heights, to document differences within the column, (3) simultaneous optical depth measurements from below and within or above sampled aerosol layers, to separate tropospheric haze from overlying optical depths, and (4) vertical profiles of *in situ* samples, to provide column properties for comparison to photometer- and radiometer-derived optical depths.

Surface measurements will provide additional information at fixed locations, with greater temporal coverage. Locations will include long-term monitoring sites operated by the Atmosphere-Ocean Chemistry Experiment (AEROCE) network in Bermuda and by NOAA/ERL, NASA/GSFC and the WMO Global Atmospheric Watch (GAW) program at other North Atlantic sites, plus special TARFOX measurements at Wallops Island.

Task 3. Integrated Analyses

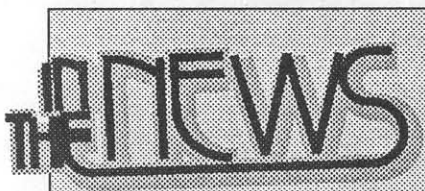
The combined ground, air, and space data sets will permit a wide variety of closure analyses. Specifically, measurements of aerosol light-scattering, absorption, and forward/backscatter ratios at a given height will be used to derive the basic quantities needed to determine the effects of aerosols on solar radiation, namely, the aerosol extinction, single-scattering albedo, and asymmetry factor. "Internal closure" will be assessed by comparing extinction coefficients, single-scatter albedos, and asymmetry factors deduced in this way with those derived from the simultaneous *in situ* measurements of particle size distribu-

tions and chemical compositions. Analyses will seek to apportion the measured extinction by chemical species and hence (although more problematically) by source. The main tool for this analysis will be multiple linear regression. At each altitude in the vertical profiles of aerosol properties, the measured aerosol scattering and absorption coefficients will be regressed onto the co-measured mass concentrations of the various chemical components of the aerosol. This will yield the chemically specific mass scattering efficiencies and hence the relative contribution of each component to the light scattering at that altitude. Weighted vertical integration of these scattering budgets will then yield a chemical "budget." This statistical approach has proven fruitful in visibility studies, which are similar in this aspect to TARFOX.

"External column closure" will be assessed by vertically integrating the two types of aircraft-determined extinction and comparing the resulting optical depths with those simultaneously derived from the airborne sun photometers, satellite radiometers, and ER-2 imaging spectrometers. Another aspect of column closure will be comparisons of aerosol radiative forcing, or radiative flux changes, determined by: (1) airborne flux radiometer measurements, (2) satellite flux retrievals from radiance measurements, and (3) flux calculations from (a) *in situ* measured aerosol scattering, absorption, and asymmetry factors, (b) the same properties derived from size distribution and composition measurements, and (c) sun photometer- and satellite-derived optical depths with ancillary single-scattering albedos and asymmetry factors. The sensitivity of radiative forcing to changes in aerosol optical thickness will be derived from the detailed *in situ* measurements and compared to the empirical sensitivity obtained by regressing satellite-derived radiative forcing vs. satellite-derived optical depth.

Such closure analyses will yield critically needed assessments and reductions of the uncertainties in deriving anthropogenic aerosol radiative forcing for use in climate models. The closure analyses that use satellite optical depth and flux results will provide tests and, where necessary, improvements of the satellite retrieval algorithms. The resulting validated algorithms will permit extensions of the TARFOX results beyond the TARFOX period and to other areas dominated by similar aerosols (e.g. the European Atlantic coast). Lessons learned and remaining questions will provide a useful background for ACE-2, which will study both direct and indirect effects of not only the European industrial aerosol plume, but also the African mineral dust plume.

More information on TARFOX is available on http://www-space.arc.nasa.gov/~tarfox, or in the TARFOX Science and Implementation Plan, available from the author (philip_russell@qmgate.arc.nasa.gov).



IGAC, START, and AGU Complete First Short Course in Atmospheric Chemistry in Latin America

Contributed by C.J. Gilman and E.W. Bierly, American Geophysical Union, USA

The first short course in atmospheric chemistry sponsored by IGAC's Atmospheric Chemistry and Environmental Education in Global Change (ACE^{ED}) Activity, the System for Analysis, Research and Training (START), and the American Geophysical Union (AGU) was given at the University of Buenos Aires, Instituto de Química Física de los Materiales, Medio Ambiente y Energia (INQUIMAE), from October 30 to November 10, 1995. The course was entitled *"Instrumentation and Measurement Methodologies in Atmospheric Chemistry Research"*. The coordinators of the course were Prof. Enrique San Roman of INQUIMAE and Prof. Volker Mohnen, State University of New York at Albany. Initial funding came from the atmospheric chemistry program in the Division of Atmospheric Sciences of the U.S. National Science Foundation and from the Inter-American Institute for Global Change Research (IAI). Supplemental funds were received from the IAI that were used to pay for the travel and per diem costs of the participants. This single factor made the difference between a fully successful course and one that would have been only marginally adequate.

The course was attended by 35 participants from 7 Latin American countries. The course was given as part of a "proof of concept" to determine if it would be possible to begin to build an atmospheric chemistry capacity in countries where baseline stations from the World Meteorological Organization's Global Atmosphere Watch (WMO/GAW) either were just built, being built, or about to be built. Argentina was selected as the country in which to start because the WMO/GAW station at Ushuaia was recently completed and there was a recognition that atmospheric

chemistry needs could not be fulfilled locally simply because there was little capacity in the subject area.

The course was open to anyone who applied, who spoke Spanish, and had a desire to take part in the course because of ongoing work that might be related or because of a need to know that might occur in the near future. As a result, the participants were a very heterogeneous group ranging from technicians operating sampling equipment to professors desiring background so they could add related material to courses they already were teaching or would be teaching soon.

The lecturers were drawn partly from ACE[ED]'s *Volunteer Teaching Corps* and also came from diverse backgrounds. The four from the U.S. represented disciplines of chemistry, oceanography, aerospace engineering, and civil and environmental engineering. The one lecturer from Brazil is a physicist. The four from the University of Buenos Aires, School of Science, represented physics, chemistry, and atmospheric sciences.

IGAC*tivities* NewsLetter

Edited by Alex Pszenny and Elaine Robbins

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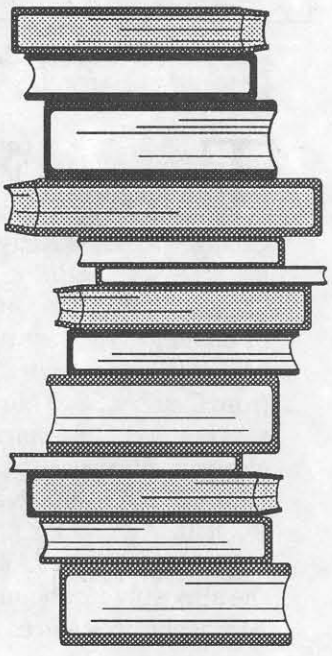
Thanks to Linda Kubrick for Logos and assistance with graphics.

The syllabus for the course consisted of an introduction and description of atmospheric chemistry, measuring systems used for sampling the atmosphere and their problems, absorption and emission spectroscopy including the underlying physical principles, gas chromatography including underlying principles, aerosol and precipitation chemistry, meteorological concepts needed to interpret the chemical measurements, computerized data acquisition, fundamentals of quality assurance and control, and data assessment. An evaluation of the course was submitted by 24 of the participants. They felt that the course attained its objectives, that the concepts were well developed, the lectures were well presented, the level of the course was good, though perhaps a bit long, and that exercises presented were useful. They indicated that they would attend a similar course or an extension were it to be offered.

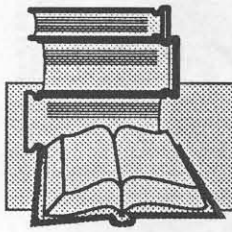
Because there was so much excitement among the participants and among the faculty and staff at INQUIMAE, there is good reason to believe that what began at this short course needs to be followed up within a reasonable amount of time to ensure achieving the goal of developing a strong Latin American capacity in atmospheric chemistry.

[Editor's Note: A second short course on *"Photo Chemical Air Quality Modeling"* for Latin American scientists will be held at the University of Chile, Santiago, 20-31 May 1996. Topics to be covered by members of ACE[ED]'s *Volunteer Teaching Corps* include:

- Fundamental process components of air quality simulation models
- Overview of operational approaches used in ozone air quality management
- Description and application of photochemical models
- Fundamentals, description, and applications of observationally-based modeling approaches



The registration deadline for this course has already passed, but there is a possibility that the course may be offered again at the Federal University of Bahia, Brazil, in late 1996.]



Publications of Interest to the IGAC Community

Most of IGAC's Activities now underway and research results are beginning to appear in the peer-reviewed scientific literature. We provide here some examples of larger efforts which, like ASTEX/MAGE (see "Science Feature"), have produced or are planning special journal issues.

- The Northern Wetlands Experiment (NOWES), coordinated by the Canadian Institute for Research in Atmospheric Chemistry (CIRAC) in collaboration with the National Aeronautics and Space Administration (NASA), assessed the importance of northern wetlands as a source/sink of biogenic gases to the atmosphere. The primary emphasis was on the exchange of CH_4 between wetland ecosystems and the atmosphere, but the fluxes of N_2O, NO_X, VOC, DMS, and CO_2 were also examined. NOWES was an early campaign under IGAC's *High-Latitude Ecosystems as Sources and Sinks of Trace Gases* (HESS) Activity. Results were published in a special issue of the *Journal of Geophysical Research (JGR) - Atmospheres* in January 1994.

- Three campaigns were conducted as part of the Southern Tropical Atlantic Regional Experiment (STARE) within IGAC's *Biomass Burning in the Tropics: Impact on the Atmosphere and Biosphere* (BIBEX) Activity. More than 100 publications have been published in or submitted to major journals. Many of these related to the Transport and Atmospheric Chemistry Near the Equator - Atlantic (TRACE-A) and the Southern African Fire-Atmospheric Research Initiative (SAFARI-92) campaigns will appear in special sections of *JGR - Atmospheres* in 1996.

- Fire of Savannas/Dynamique et Chimie Atmosphérique en Forêt Equatoriale (FOS/DECAFE), another early BIBEX campaign, studied gas and particle emissions from savanna fires in the Ivory Coast of Africa in 1991. Results appeared in a special issue of the *Journal of Atmospheric Chemistry* in October 1995.

- IGAC's *East Asia-North Pacific Reginal Experiment* (APARE) Activity aims to understand processes controlling atmospheric chemistry in the rapidly developing East Asian region and the effects of airborne pollutants (ozone in particular) on the marine region downwind. An APARE/IGAC special issue of *Terrestrial, Atmospheric and Oceanic Sciences* appeared in September of 1995 containing the proceedings from an "International Conference on Regional Environmental Changes in East Asia" that was held in December of 1993. In addition, the Pacific Exploratory Mission (PEM) - West, one of the PEM series of experiments executed by NASA, provided airborne observations of ozone and its precursors as well as sulfur species over the western Pacific using extensively instrumented aircraft as the primary tool. Results from the first field phase of this campaign appeared in the January 1996 issue of *JGR - Atmospheres*.

- Several campaigns have been sponsored by IGAC's *Global Atmospheric Chemistry Survey* (GLOCHEM) Activity. The Mauna Loa Obsevatory Photochemistry Experiment (MLOPEX) made concurrent measurements of the key species that play controlling roles in the photochemical transformation of ozone, odd nitrogen, and odd hydrogen species in the remote free troposphere. Results from these campaigns will be published soon in *JGR - Atmospheres*. Additional measurements of chemically active gases in the free troposphere were made during the Second Tropospheric Ozone (TROPOZ-II) campaign in January-February 1992. Ozone and water vapor in the mid- and upper troposphere are also being measured from automatic instrumentation aboard five long-range commercial aircraft under the Measurements of Ozone on Airbus In-Service Aircraft (MOZAIC) project.

- The *North Atlantic Regional Experiment* (NARE) Activity conducted a series of intensive measurement campaigns between 1991 and 1994. A special issue of *JGR - Atmospheres* is in preparation.

- The Polar Sunrise Experiment 1992 (PSE-92) conducted in the Canadian Arctic as part of IGAC's *Polar Atmospheric and Snow Chemistry* (PASC) Activity aimed to better understand the role of marine halogens and chemical destruction mechanisms in lower tropospheric ozone depletion at polar sunrise. A special section of *JGR - Atmospheres* containing the results was published in December 1994.

- The Atlantic Stratocumulus Transition Experiment (ASTEX) conducted by IGAC's *Marine Aerosol and Gas Exchange* (MAGE) Activity in 1992 improved our capability for studying cloud chemistry interaction and the air-sea fluxes that affect them. Initial results were published in the February 1996 issue of *JGR - Atmospheres*.