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Planning for Chemical Air-Sea Exchange Research

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The International Global Atmospheric Chemistry (IGAC) Programme has been created in response to growing international concern about rapid atmospheric chemical changes and their impact on mankind. This program, while emphasizing atmospheric composition and chemistry, recognizes that the Earth's atmosphere, oceans, land, and biota form an interacting system that collectively determine the global environment and its susceptibility to change. The IGAC Programme is intended to be a vital contributor to the broader interdisciplinary program of the International Geosphere Biosphere Programme (IGBP), providing the important atmospheric chemistry component and recognizing its linkages with the biosphere and human activities. The IGAC Programme is building on existing national programs by providing the international cooperation whereby essential scientific endeavors can be accomplished, even though they involve large demands for man power, technology, geographic coverage, or monetary resources beyond the capability of a single nation.

One of the six initial IGAC foci is the natural variability and anthropogenic perturbations of the marine atmosphere. Since the oceans cover about 70% of the planet and act as both a source and sink of many important atmospheric constituents, it is an essential area in which to study source, sink, and transformation processes in detail. IGAC therefore proposed a project to study Marine Aerosol and Gas Exchange, Atmospheric Chemistry and Climate (MAGE).

The goals of MAGE are to understand the

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chemical, biological, and physical mechanisms that control the exchange of trace gases and particulate materials between the atmosphere and the surface of the open ocean; to develop formulations of open ocean exchange processes for inclusion in global-scale climate and air chemistry models; and to extend the experimental knowledge of air-sea exchange to conditions of strong winds, rough seas, and spray. The MAGE Steering Committee's first meeting was held in Anaheim, Calif., in February 1990.

Key MAGE Issues

The ocean is a source and/or sink of many trace species that affect the radiative balance of the Earth either directly (CO_2, CH_4, N_2O) or indirectly (CO, hydrocarbons, halides, sulfur compounds) by altering the photochemistry of the marine atmosphere. The first step in studies of air-sea exchange is to document the oceanic and atmospheric concentrations of the climatically important trace species. These measurements can be used to assess the seasonal and regional sources and sinks of these species and to evaluate natural and anthropogenic contributions to annual and decadal trends in their oceanic and atmospheric distributions. Although these measurements can be used to calculate fluxes, they are not measured fluxes. We are technologically limited in being able to directly measure the emissions and deposition of the key species and therefore must seek additional methods to verify the flux calculations.

The second step in studies of air-sea exchange must be to improve our understanding and predictive capabilities of the physical and biogeochemical processes controlling the oceanic and atmospheric concentrations of these climatically important trace species. Studies of these processes, for example, the physical parameters influencing air-sea exchange, the biological production and consumption of climate gases and their precursors, marine photochemistry and its affect on chemical concentrations will require not only measurements of the trace species but also many ancillary physical, chemical, biological and meteorological parameters. The complexity of these studies require an integrated interdisciplinary approach often including simultaneous measurements from several research platforms. We must seek ways to develop the monetary and personnel resources to carry out these large studies.

Remote sulfur chemistry is a subject of particular interest to MAGE. Improving our understanding of the impact of DMS emissions on clouds is fundamental to improving climate models, since some models suggest that a 4% in the areal extent of marine stratocumulus clouds could have a cooling effect equivalent to (offsetting) a 30% increase in CO_2. Cloud-condensation nucleii (CCN) formed from DMS might be one of the significant controllers of cloud radiative properties. We are technologically limited in not being able to measure both the emissions of DMS and H_2S and the deposition of SO_2, MSA, and sulfate aerosol.

Continental outflow can affect the chemistry of the marine boundary layer (MBL) through the addition of anthropogenic pollutants. In what regions of the ocean is sulfate aerosol formation dominated by man-made SO_2, for instance, and where is DMS the major source? This answer would give us an idea of the maximum global change we could effect by reducing air pollution. The continents can also have a major impact on biota, since the atmospheric transport of nutrients derived from continental sources is thought to limit productivity in some marine regions. How might changes in land-use practices alter the productivity of the oceans? We cannot answer such questions without much better estimates of fixed-nitrogen and trace metal deposition.

Finally, we recognize that marine aerosols are important factors in the global radiation budget, independent of their role as CCN. Existing models of aerosol deposition are very sensitive to such factors as size, which unfortunately changes dramatically over the normal humidity gradients in the MBL. Improved deposition measurements are needed to estimate the lifetimes of these radiatively important particles, so that we can model the impact of changes in source terms.

We see two areas in which MAGE can contribute to studies of air-sea exchange. The first is to promote the development of new measurement technologies, while the second is to organize field projects to both test the new technologies and to facilitate large interdisciplinary research studies. We have outlined below several areas where technological improvements will aid air-sea exchange studies and we have identified three field opportunities where IGAC/MAGE can take advantage of ongoing programs to promote studies of air-sea exchange.

Technique Development

Unfortunately, air-sea exchange research is seriously limited by technology. Most of the important fluxes have never been measured in the field. We believe that improvements in the understanding of marine aerosol and gas exchange can only be achieved through the development of new measurement technology. We hope that the field programs discussed below will serve as test beds for several promising new techniques.

If one were to measure both the diurnally varying atmospheric concentration of DMS and its loss (reaction) rate, the surface source of DMS could be computed. We propose to use this approach, applied in a Lagrangian framework, to estimate marine surface fluxes. In the case of DMS, the loss processes are not yet well enough known to improve our flux estimates by much, but simultaneous measurements of oxidants and related sulfur species combined with the results of ongoing laboratory studies offer the potential for significant progress. Once we have demonstrated our ability to do a Lagrangian sulfur experiment, we would like to conduct a release of DMS to quantify its reaction rates and pathways and to observe its impact on cloud microphysical properties.

To do such a Lagrangian experiment, one must measure ambient concentrations of SO_2, DMS, aerosols (sulfate, ammonium, and MSA), and the ionic species in liquid cloudwater (preferably as a function of droplet size). However, even concentration measurements are not routine for most of these species. Development work is still needed to achieve 1–2 pptv SO_2 sensitivity on a 5-min integrated sample, for instance, since 5–15 pptv SO_2 concentrations are common in the boundary layer far from continents.

Airborne measurements of entrainment across the MBL top can be combined with time-derivatives of concentrations to estimate surface fluxes. By doing such experiments in a Lagrangian mode, it is possible to eliminate concentration changes resulting from advection. We hope to use "smart" constant-level balloons, which can adjust their bouyancy to maintain one altitude, to tag MBL air masses, so that we can sample in the same parcel on two successive days from an aircraft. Once this technology has been demonstrated in the field operations described below, it can be used to ensure the success of a major release experiment.

The eddy correlation technique produces the most reliable flux measurements, but it requires a fast chemical sensor, capable of 1-Hz measurements from towers or 10-Hz rates from aircraft. Recent rapid advances in the field of mass spectrometry suggest that it should be possible to develop a mass spectrometer that would be capable of measuring DMS at 1 Hz or faster. Although an atmospheric pressure-ionization triple-quadrupole MS should be capable of this kind of speed, there are also smaller, lighter, and thus more airworthy, systems that also offer promise. This would represent an incredible advance, since one of the most interesting fluxes could be measured directly, permitting us to "calibrate" the thin film measurements on which our current estimates are based. We feel that this development should be given extremely high priority because of its central importance. It is likely that this same technology can be extended to direct flux measurements of other important species, as well. Other promising micrometeorological methods, such as eddy accumulation, should also be pursued.

It is difficult to employ micrometeorological techniques such as eddy correlation and gradient measurements from marine platforms because the instrument-support structure, such as a ship, distorts the airflow one needs to measure. We urgently need work on ways of resolving this problem. Among the possibilities are bouy-mounted towers, in which much of the structure is below the waterline and an inertial navigation system would be used to subtract the motions of the sensors from the total observed (relative) air motions. Airships also offer possible advantages, since they could deploy aerodynamically small instrument platforms on cables below their gondolas. Their low airspeed may also present a significant advantage for aerosol measurements. Coastal towers, drilling platforms, long forward-reaching structures for ships, and even submarines may serve as platforms for long-term, relative to aircraft, marine micrometeorological flux measurements. Although atmospheric aerosols have not received nearly the attention or support that gas-phase chemistry has, they are central to understanding many global-change questions. Since it is now apparent from recent experiments on the National Center for Atmospheric Research's Electra that many of the previous airborne aerosol measurements may have suffered from serious sampling artifacts, we feel that intensive development work should be concentrated on the sampling and chemical analysis of aerosols. Without reliable airborne size-resolved chemical measure-

ments, we cannot hope to prepare credible budgets for sulfur or other species with important condensed phases, let alone estimate aerosol loss rates and humidity-dependent radiative impacts. Similar sampling problems for CCN analyzers and liquid cloudwater instruments also merit extra attention, both for the study of cloud microphysics and because of the importance of clouds as processors of aerosols and gases.

We strongly support IGAC's philosophy that the intercomparison of related techniques should be used wherever possible to confirm the utility of any of these measurement strategies. Measurements for thin-film flux estimates should routinely accompany any of the newer measurements described above. Only by deploying as many related techniques as possible can we hope to improve our confidence in the results from each measurement system. In recent years, the atmospheric chemistry community has made major advances in understanding by using field intercomparisons, and we intend to continue in this mode.

Field Programs

IGAC/MAGE has identified three field opportunities, which are discussed below.

JGOFS/IGAC Experiment in the Equatorial Pacific Ocean, November 1991

The U.S. JGOFS (Joint Global Ocean Flux Study) program is planning a major field program in the equatorial Pacific Ocean (140°W, 9°N to 15°S) in the fall of 1991 and spring of 1992 to study the processes controlling the cycling of carbon and related biogenic elements in the oceans. The cruises will allow a substantial number of biological and chemical measurements designed to define many important oceanographic processes, for example, new production, foodweb structure and function, vertical flux of biogenic elements, and remineralization of biogenic compounds. Many of the processes being studied by JGOFS will be important factors in the oceanic production and consumption of biogenic trace gases and hence will affect the rate of gas exchange across the air-sea interface.

IGAC/MAGE is planning to conduct a suite of trace gas and aerosol measurements that will complement the JGOFS oceanographic studies by looking at the cycling of trace gases within the ocean and the exchange of these biogenic compounds with the atmosphere. Plans are underway to measure the concentrations and biogeochemical cycling of the major carbon, sulfur, nitrogen, oxygen and halogen species in the ocean and overlying atmosphere, probably in October and November of 1991.

The IGAC/MAGE committee is seeking both ship (NOAA, UNOLS) and aircraft (NCAR Electra) platforms in order to measure the biogenically and climatically important trace gases and aerosols. Scientists interested in participating in JGOFS/MAGE should contact, for ship measurements, T. Bates, NOAA/Pacific Marine Environmental Laboratory, Seattle, WA 98115; or for aircraft measurements, A. Bandy, Department of Chemistry, Drexel University, Philadelphia, PA 19104.

ASTEX/FIRE Experiment in the Tropical Atlantic, June 1992

ASTEX (Atlantic Stratocumulus Transition Experiment) is a part of the second series of FIRE international cloud-climatology experiments. The primary purpose of ASTEX is to study the factors influencing the formation and dissipation of marine clouds. There are plans to have meteorological and chemical instrumentation on both Santa Maria island in the Azores and Porto Santo in Madiera, as well as on a ship west of the Canaries, which is located so as to complete an equilateral triangle. Aircraft from at least three countries will also be sampling in and around the study area. ASTEX is being designed to overlap with several other international oceanographic and atmospheric programs that will share the eastern Atlantic in the summer of 1992.

Chemical investigations during ASTEX will address several issues. First is the impact of aerosol chemistry on the microphysics of cloud formation and dissipation and climate. Are anthropogenic aerosols affecting either the extent of cloudiness or the marine radiation budget? Of course marine sulfur cycling is central to such questions, so we plan to make comprehensive measurements of sulfur compounds and the photochemical oxidants with which they react. Nitrogen oxide measurements will also be used to help quantify continental outflow and nutrient deposition to coastal waters.

The centerpiece of our aerosol and gas exchange studies in ASTEX is intended to be a Lagrangian experiment using smart constant level balloons, alongside traditional thin-film measurements. We hope to use the NCAR Electra to sample near the balloon launch point, about 500 km upwind of our airfield on the first day, and then use satellite tracking to find the balloon, and the same airmass, a similar distance downwind 24 hours later. Scientists interested in participating in ASTEX/MAGE should contact B. Huebert, Center for Atmospheric Chemistry Studies, Graduate School of Ocenaography, University of Rhode Island, Narragansett, RI 02882; tel. 401-792-6616; fax 401-792-6899.

PSI Experiment in the Southeast Pacific (Easter Island), April/May 1993

The Pacific Sulfur/Stratus Investigation (PSI) is designed to define the role of marine biogenic sulfur emissions in the formation of non-seasalt (NSS) sulfate aerosol particles and cloud concentration nuclei (CCN), and to quantify the effects of these species on cloud albedo and climate. PSI is an interdisciplinary/interagency program using ship, aircraft, satellite, and shore-based measurements, to describe the biological, chemical, and physical processing of sulfur gases into CCN. It will employ measurements of atmospheric gas phase, condensed phase and cloud water chemistry, aerosol size distribution, CCN population, cloud microphysics, cloud albedo, and meteorology.

A major field experiment is being planned for the beginning of 1993 in the southeast Pacific Ocean. This is a region distant from both the northern hemisphere background sulfur burden and southern hemisphere continental sulfur emissions and is a region where the stratus clouds cause a net cooling of the Earth. This experiment is an excellent opportunity for both the IGAC MAGE and the IGAC CCN/Cloud projects to join efforts in studying the remote background sulfur cycle and its effect on CCN, clouds and climate. We hope to be able to do a DMS release experiment here, using our Lagrangian experience from ASTEX to track the released plume. Scientists interested in participating in PSI/MAGE should contact T. Bates or R. Charlson.

Participation

IGAC and MAGE are intended to be inclusive programs, which welcome the participation of all interested scientists. Anyone interested in participating in any part of the projects described above should contact either the scientists identified as planners of individual experiments or the MAGE Steering Committee Convener, Barry J. Huebert. An open meeting of the steering committee will be held in conjunction with the 7th International Symposium of the Commission on Atmospheric Chemistry and Global Pollution (CACGP) Meeting in Chamrousse, France, this September.

Copies of the 1989 IGAC Programme report, edited by I. Galbally, outlining the entire range of IGAC plans, can be obtained from Ronald G. Prinn, IGAC Steering Committee Chairperson, Dept. of EAPS, MIT 54–1824, Cambridge, MA 02139.

News

Free Report From Liquefaction Workshop

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The technical report *Proceedings From the 2nd U.S.-Japan Workshop on Liquefaction, Large Ground Deformation and Their Effects on Lifelines* is available free of charge from the National Center for Earthquake Engineering Research, headquartered at the State University of New York at Buffalo. The 499-page proceedings contain more than 30 reports on case studies of liquefaction and earthquake-induced ground deformation from previous earthquakes in the U.S. and Japan.

The proceedings are the result of the second in a three-part series of workshops sponsored by NCEER and the Association for the Development of Earthquake Prediction (Japan). The workshops are part of a U.S.-Japan cooperative research program between the sponsoring organizations, which is believed to be the first sustained international effort on ground deformation. The goal of the series, which continues through 1990, is to develop international guidelines for improved engineering solutions and assessments related to earthquake-induced ground deformation.

Free copies may be obtained by calling NCEER at 716-636-3391 and asking for Technical Report 89-0032.