IGACtivities Newsletter

of the International Global Atmospheric Chemistry Project

A Note From the IGAC Chair: *Guy Brasseur*

Atmospheric Chemistry and Climate

At the end of the 19th century, the Swedish Nobel laureate Svante Arrhenius suggested that a doubling in atmospheric carbon dioxide (from coal burning) would increase the Earth's mean temperature by 5-6 degrees Celsius. Numerous climate models have been developed in recent decades to study in more detail the atmosphere's response to such human-induced perturbations. Though early models focused entirely on CO_2, in the last 10-15 years climate researchers have recognized the importance of such chemically and radiatively active gases as methane, nitrous oxide, halocarbons and ozone. Some of these are produced by the biosphere, but their concentrations are all perturbed on the global scale by human activities. These greenhouse gases contribute to global warming.

Now a variety of aerosols are being recognized for their impact on climate. Sulfate aerosol particles, produced through coal burning, scatter back to space a significant fraction of incoming solar radiation, leading to potential regional cooling in industrialized/urbanized areas. In this issue of *IGACtivities*, Barry Huebert and Don Lenschow discuss several field campaigns designed to investigate the chemical and microphysical processes involved in the formation and fate of these aerosols. Because aerosol particles are believed to serve as condensation nuclei for the formation of clouds, aerosols of anthropogenic origin are expected to produce an indirect climate forcing through changes in the optical properties of the clouds. This difficult scientific issue is discussed in the following pages by John Seinfeld and Richard Flagan.

Other types of aerosols are also significant. Black carbon (soot) tends to absorb a fraction of the solar and terrestrial radiation, thus warming the atmosphere. Irina Sokolik describes in this issue how mineral dust affects the atmosphere's radiative balance and hence the climate system. Sea salt and organic aerosols are additional particles that need to be considered in comprehensive climate studies.

One of IGAC's foci is atmospheric aerosols. The importance of this issue for global change is becoming increasingly evident. Aerosols not only affect climate, they play an important role in the chemistry of the stratosphere and troposphere through a number of heterogeneous reactions, and have a significant impact on the intensity of photolytic radiation. They affect visibility, damage plants, and can be detrimental to health. But, while fouling the air, aerosols also create beautiful sunsets!

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Introduction

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"Greenhouse Forecasting Still Cloudy,"
—*Science*, 16 May 97

Is it just the headline writers, or do most people expect that scientists should be capable of providing a definite answer to every possible question about how the Earth works? I think this idea that scientists ought to know everything probably comes from primary school, where there is always one kid in class who seems to have every answer. He or she can work the science and math problems nobody else can, and all the other kids feel like he or she makes them look bad. Years later some of these smart kids become Earth scientists, and their classmates still expect them to know everything.

Only Earth science questions are a lot harder than those we had to answer in third grade. When society needs to know whether we are causing irreversible climate change and how it will affect rainfall in the world's grain belts, many people think these smart kids ought to be able to give them an answer. You and I know, however, that many geophysical problems are so vast and so complex that we may never have the tools to provide concrete solutions to them. For questions about aerosol forcing of climate change our understanding is improving, but even our most confident answers have large uncertainties.

Yet the mass media react as if it is scandalous when scientists disagree or when they admit to not knowing the answers to important environmental questions. The popular conception seems to be that the scientific community is a failure if it can't provide uniform, direct answers to questions about how changes in emissions would change climatic impacts.

Ironically, that kind of certainty is the exact opposite of how good science should work. Frank discussions about uncertainties motivate progress. When I need the best possible judgement about an issue, I try to ask someone who is skeptical about our level of understanding. The person who believes our present theories are good enough (even if they are) is not likely to be motivated to look as hard at the weakest points in hypotheses. The doubter, by contrast, will try to calculate just what the major uncertainty is, and how that can be reduced to give us greater confidence in the answer.

The U.S. National Research Council convened a Panel on Aerosol Radiative Forcing and Climate Change a few years ago to evaluate uncertainties in the role of aerosols in the climate system and to recommend the best strategies for reducing them. To reduce the uncertainties in calculated aerosol radiative climate forcing to 15% both globally and locally, the Panel (NRC, 1996) recommended six categories of research that need to be conducted in concert:

- Global climate model development;

- Process studies (modeling and small-scale field studies);

- Field studies: large, international field studies and systematic ship- and aircraft-based surveys of aerosol properties;

- Satellite observations and continuous measurements from surface-based research measurement networks;

- Technology development (excluding satellite development); and

- System integration.

Interestingly, the Panel also found that few of these recommendations were being made for the first time. An extensive Appendix quotes from publications as far back as 1971 that have argued for similar work on many of these same issues. At issue now is how to finally implement these recommendations in the most cost-effective manner possible. Several implementation strategies are discussed in this issue of the IGAC newsletter.

Since the Panel last met in 1995, there have been advances in our understanding of the impact of aerosols on climate. IGAC has organized three major field programs, ACE–1 [Bates *et al.*, 1998], TARFOX [Russell *et al.*, 1999], and ACE–2 to study aerosol distributions, properties, and processing (in the clean marine troposphere, North American outflow, and European outflow, respectively). In part because of these

experiments we have developed a more quantitative understanding of aerosol radiative forcing, but we have also learned much more about how to study the problem most effectively. Our models are more realistic, we have developed new instruments for studying the problem, and our experimental strategies are improved. In the articles that follow, we evaluate some of this progress and reassess strategies for reducing uncertainty in the impact of aerosols on climate.

These papers describe strategies for addressing several of the issues the NRC Panel raised:

- Is the Lagrangian observational strategy a viable one for quantifying process rates in the atmosphere? (Huebert and Lenschow)

- How should we modify our approach to the indirect forcing question in view of our present understanding of aerosol/CCN/cloud droplet number relationships? (Seinfeld and Flagan)

- How does the evolution of mineral dust modify the impact of the resulting multi-component aerosol? (Sokolik)

Some review the results of recent IGAC experiments. All point us toward strategies we should be using in the future. Some of the strategies are discussed in the context of ACE-Asia, a new field campaign being planned.

The international community of aerosol researchers is turning its attention to Asia. The rapid evolution of emissions in the region creates an opportunity to do a stimulus-response experiment. As automotive emissions increase [Elliott et al., 1997], how will the photochemical system respond? When more coal is burned or biomass emissions are reduced, how will these changes modify the air the regions' population breathes and the deposition of particles to the ocean downwind?

These are global issues both because they affect the global climate and because in one form or another they are common to each continent. The only way to understand such common problems is to study them together. IGAC is structured to do just that, with a truly democratic structure that allows every country to include its ideas and expertise in the planning process. Through this vehicle, we are planning some experiments that will help everyone do a better job of estimating the impact of their emissions. The more countries, funding agencies, and research groups that decide to participate formally, the more powerful and useful the results will be to people everywhere.

The sophistication of Asia's atmospheric scientists makes it possible to mount a broad-reaching study of Asian aerosols and their ultimate fate over the Pacific. They will be working with scientists from Europe, North America, and Australia who will bring state-of-the-art aerosol instruments to study this interesting region. Several years of network operations will be supplemented by multi-platform intensive experiments during the spring period of outflow to the Pacific. In March and April of 2001, for instance, the goals will be to 1) survey the composition of aerosols at many latitudes, locations, and distances from the coast and 2) study the evolution of those aerosols with time and distance to quantify the processes that control their concentrations and effects. More information can be found at http://saga.pmel.noaa.gov/aceasia/.

Since you are reading this, you were probably one of those kids who stood out in grade school as better than most at doing science and math. If so, you now share the burden to provide policymakers in your country with reliable predictions about human-induced climate changes, including realistic error bars. But progress in understanding the Earth system is incremental by nature. When we do an experiment, we learn a little about the Earth and a lot about how to be more successful next time. Our hope is that the strategies summarized in the following papers will help us fine-tune the course of aerosol/climate research, so that our next experiments will convince even us that scientists are pretty smart.

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What have Lagrangian experiments accomplished?

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Some problems involving reactive trace species in the atmosphere are simply so complex that there is no substitute for observing their rates in the actual atmosphere. A good example is the oxidation of sulfur dioxide (SO_2) to non-sea salt sulfate (NSS). Four processes compete for SO_2, and each could be significant in various circumstances: 1) dry deposition to the Earth's surface in the marine boundary layer (MBL) may remove as much as half of the NSS produced; 2) homogeneous oxidation by OH is the only pathway that can produce the gas-phase H_2SO_4 needed to support the nucleation of new particles; 3) oxidation of SO_2 in cloud droplets is now thought to be the major source of NSS; and 4) finally, under some circumstances seasalt aerosol water may also be an important oxidation medium. Its alkalinity may keep it from rapidly acidifying, thereby permitting extra oxidation by ozone.

Existing models cannot predict to within 10% how much SO_2 is lost by each process. Some parameters are rather rough estimates; it is not hard to imagine that one of them could be off by a factor of 2, which would dramatically change the predicted relative importance of all four processes. Thus, if we want to know how much additional NSS would be formed by a 20% increase in SO_2 emissions from the US or Asia, for instance, we need more certainty than the models can supply. To gain that certainty we must measure the rates of these processes in the actual atmosphere.

This can be done in two ways. One is to compare the output of chemical transport models (CTMs) with a time-series of concentrations in the area of interest. High-quality, comparable time-series data from many locations can constrain many factors in a model. The other approach is to measure the change of concentrations with time in a single airmass by following the airmass with a suite of measuring instruments as it moves. This Lagrangian approach is extremely powerful but difficult to implement. It requires multiple aircraft (with double crews to fly successive flights), ships, forecasting personnel, and a significant infrastructure. Is it worth it? We can address this question by looking at the experience gained from a series of Lagrangian experiments conducted over the past 12 years during major field campaigns: DYCOMS, FIRE Stratocumulus, ASTEX/MAGE, ACE-1, and ACE-2.

DYCOMS

One approach to Lagrangian observations started with an interest in the differences between free troposphere (FT) and MBL concentrations of nitric acid (HNO_3) and SO_2. To aid the analysis, a simple cloud-capped quasi-steady-state MBL regime was selected—the marine stratocumulus region at the eastern edge of the summertime subtropical high, where large-scale subsidence produces a well-defined lid to the MBL. This simplifies evaluation of trace species budgets in a Lagrangian framework.

The budget of any species, S, can be described by a continuity equation [Lenschow et al., 1981; Kawa and Pearson, 1989]:

$$\frac{d\langle S \rangle}{dt} = J_0(S) - J_h(S) + F(S) - D(S)$$

Entrainment fluxes into the MBL, $J_h(S)$, can be parameterized in terms of the entrainment velocity, V_E (common to all species being mixed) and concentration differences across the capping inversion. Repeated measurements in the volume can define $d\langle S \rangle/dt$, the time-rate of change for each species. If the chemical formation and loss rates, $F(S)$ and $D(S)$, of this substance can be measured, the surface flux, $J_0(S)$, may be solved for as a residual. Alternatively, if the surface flux is measured, it may be possible to solve for the chemical formation and destruction rates.

One uncertainty in this analysis is determined by the magnitude of the concentration change during the observation period, $d\langle S \rangle/dt$, relative to the uncertainty in species measurement systems. For many species the limited flight duration in DYCOMS made it impossible to remain in the study airmass long enough to detect a change [Lenschow et al., 1988].

FIRE Stratocumulus IFO

To overcome this measurement time limitation it is necessary to return to the target airmass for multiple flights. But how can you be certain that you sample the same air during successive flights? Neutrally-buoyant free-floating balloons can, in principle, mark the airmass. During the FIRE stratocumulus IFO experiment near San Diego [Albrecht et al., 1988], balloons deployed from a small boat near shore were tracked from a light aircraft, but were lost when they drifted into controlled military airspace. We learned that we needed a deep-water ship to launch the balloons and a transmitter on the balloons that the research planes themselves could detect from at least 150 km away. This experiment also provided examples of "decoupled" MBLs [Paluch and Lenschow, 1991], which we found in later experiments to be a common occurrence over the ocean.

ASTEX/MAGE

In ASTEX/MAGE, we were fortunate to have two ships—a balloon-launch ship at the start of the trajectories and a second ship to intercept them at the end. In addition to balloon launching, ships are great platforms for making continuous measurements of atmospheric and oceanic variables, and aerosol chemical size distributions. During the first Lagrangian, water accumulation due to drizzle from marine stratocumulus sank all the GPS-equipped balloons within 7 hours. Since observing the modification of aerosol by rain is an important objective, the marker balloons need to be able to survive modest rainfall.

For our second Lagrangian experiment, the balloons' ballast was put in a plastic bottle dangling from a line beneath the balloon. Using this approach in a heavily polluted European airmass, one balloon was tracked for 42 hours and sampled during seven research flights, four by NCAR's Electra (Figure 1). The MBL, which was about 2 km deep, contained a weak inversion at about 0.6 km which decoupled the upper part from the surface-based mixed layer (SBML), except for occasional intermittent transport. Unfortunately, our sampling strategy (the number and height of flight legs) was predicated on the assumption of a well-mixed MBL with a well-defined lid, as in DYCOMS. In retrospect, we should have made a point of chemically characterizing the separate layers much better than we did, but our understanding of decoupled layers was still in its infancy. The combined observations from two aircraft produced a picture of the evolution of a deep two-layered MBL as it moves over warmer water that has been extremely useful for understanding their dynamics and evolution [Bretherton *et al.*, 1995].

Three significant chemical evolution papers resulted from this Lagrangian experiment. In one, based on essentially a single flight drifting with the airmass, Wingenter *et al.* [1996] found that the loss rate of some hydrocarbons was faster than OH alone could explain. The UC-Irvine group attributed the additional loss to atomic chlorine. Clarke *et al.* [1996] studied the evolution of aerosol populations in several layers, quantifying the impacts of mixing, coagulation and removal on aerosol properties. Zhuang and Huebert [1996] looked at the ammonia budget and quantified an ammonia source at the ocean's surface. Marine-fixed nitrogen is redistributed via the atmosphere, potentially moving hundreds or thousands of kilometers before being washed out in another region.

In each of these studies, our ability to quantify chemical and physical processes was limited by our ability to quantify the impact of vertical mixing. It is critical, therefore, that we improve our technologies for measuring the vertical transport and mixing of airmasses. One essential measurement is the rate of transport, or entrainment of mass into (and sometimes out of) the MBL across the capping inversion. A direct method for obtaining this measurement is to measure the flux profile of a tracer species through the MBL and extrapolate the profile to the top. The entrainment velocity, V_E, is obtained from the ratio of the flux to the jump in tracer concentration across the top. Kawa and Pearson [1989] used a fast-response ozone instrument to measure the ozone flux across the inversion by eddy correlation in DYCOMS. Unfortunately, ozone is not the optimal tracer for this measurement.

A schematic illustration of species concentration and flux profiles in the relatively simple DYCOMS regime is shown in Figure 2. Two different types of species are illustrated. The top panel shows the behavior of ozone, which has a small sink at the surface and a source above
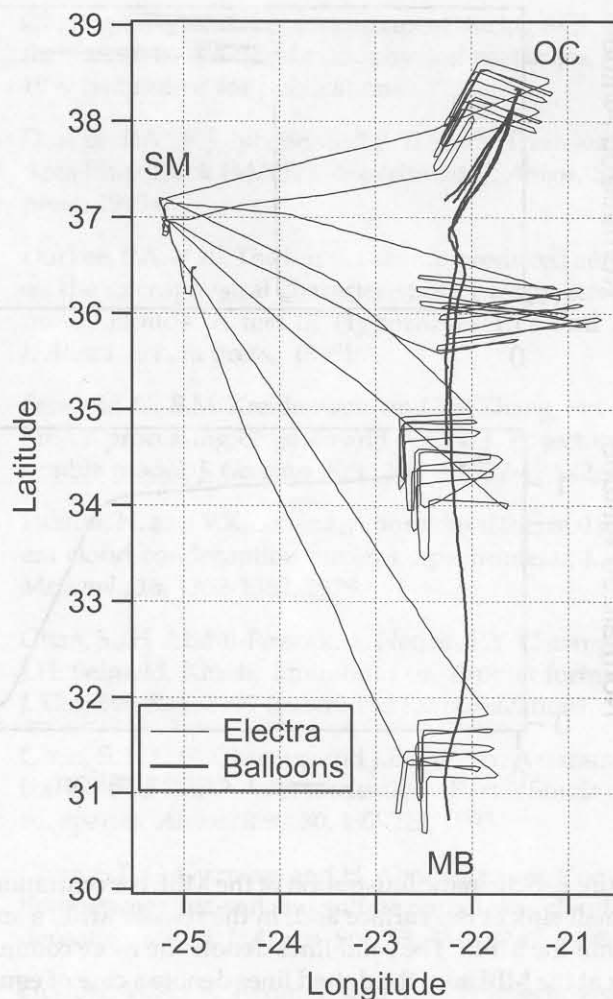

Figure 1. Electra flights, ship locations, and balloon tracks (long, nearly north-south lines) during ASTEX/MAGE Lagrangian #2. SM is the airport at Santa Maria; OC is the location of the balloon-launch ship, R/V *Oceanus*; and MB is the location of the R/V *Malcolm Baldrige*.
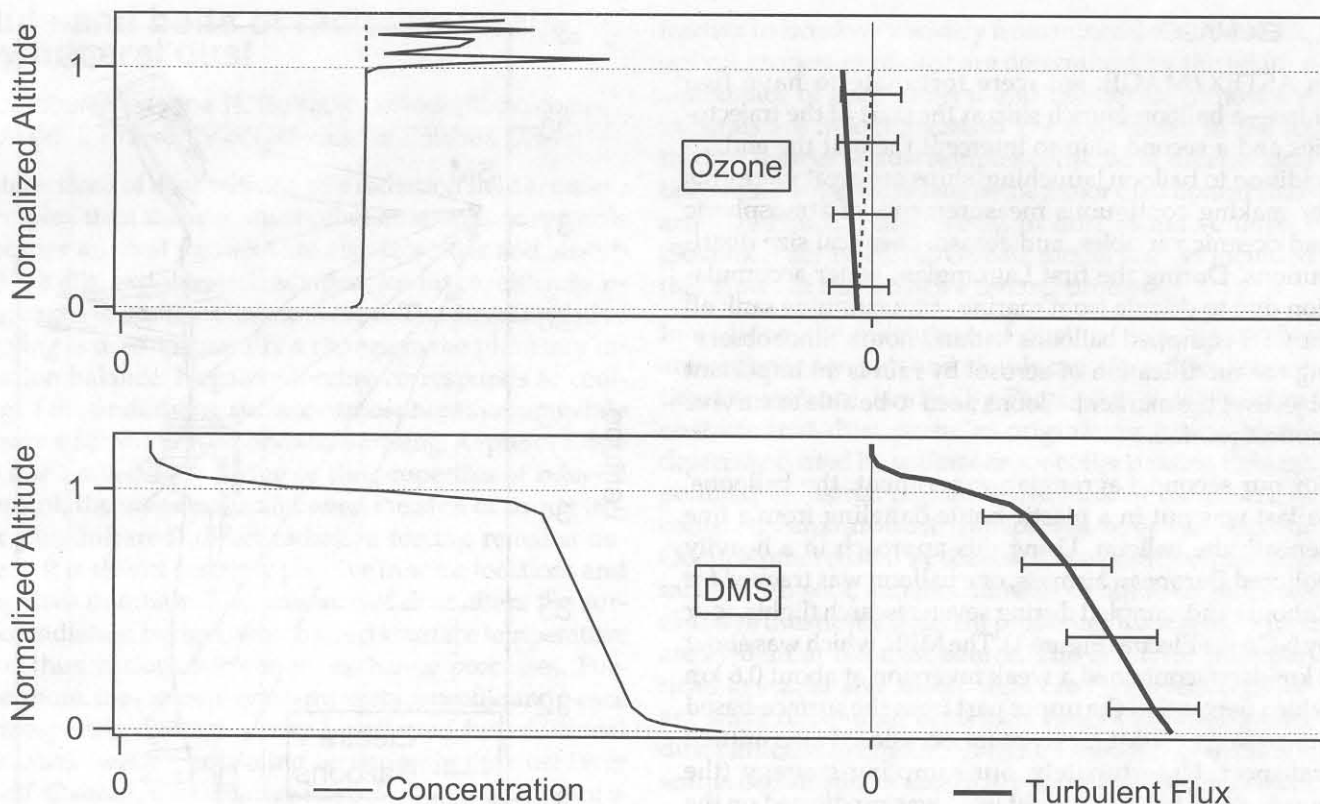
Figure 2. Schematic illustration of the MBL concentration and flux of ozone (top panel), a species with a source aloft, a small sink at the surface and, in the remote MBL, a small (i.e., time scale of a week or more) photochemical sink within the MBL. The solid lines denote the more common case of greater ozone concentration aloft and a negative flux at the MBL top; the dotted lines denote a case of equal concentration within and above the MBL, where ozone is useless as a tracer of entrainment. The bottom panel illustrates concentration and flux of DMS, a species with a source at the surface and a photochemical lifetime in the atmosphere of about a day. This results in a minimal concentration above the MBL and thus a relatively large entrainment flux and well-defined and easily measured jump across the MBL top.

the MBL. As a result, the ozone concentration is higher above the MBL, where significant spatial variability exists, than within it. This leads to a variable (and sometimes very small) jump across the top which is difficult to measure accurately, and also a variable flux across the top. This can also lead to horizontal variability in the mean ozone concentration within the MBL. Thus, ozone can sometimes be useless as a tracer of entrainment.

Dimethylsulfide (DMS) is an ideal molecule for quantifying vertical exchange in the MBL. Concentration and flux profiles of DMS are shown in the bottom panel of Figure 2. DMS comes only from the surface (so there are virtually always quantifiable differences between layers), is negligibly soluble in clouds, and has a photochemical lifetime of only a few days so it decays in layers not in contact with the surface. If we could directly measure the DMS flux by eddy correlation, we could directly calculate entrainment velocities between layers. Other species fluxes could be accurately calculated by analogy, using the DMS-derived V_E and easily-measured concen-

tration differences across inversions. The challenge, then, is to develop a fast (10 Hz) DMS system for directly measuring the DMS flux from aircraft. Two fast DMS techniques offer promise: fluorine-induced chemiluminescence and atmospheric pressure ionization mass spectrometry; neither has been demonstrated in the field. Both should be given a high priority for development, since they would markedly increase the accuracy of Lagrangian-derived process rates.

ACE-1

Several improvements were made to the technology in ACE-1 [Bates et al., 1998], although we had only one aircraft and were thus unable to observe the airmass continuously. The balloons were made "smart," so that they could adjust their own buoyancy in response to drizzle. We were careful to map out layers in the target airmasses and to get useful samples in each. Also, we developed a strategy for measuring V_E by three independent techniques, each of which can be implemented using the same

flight track [Lenschow et al., 1999]: First is the inversion flux/concentration-jump technique described above. Second is evaluating budgets of trace species for which the chemical source/sinks and surface fluxes are known, and solving for the entrainment flux. Third is the divergence technique. By flying constant-level sampling legs in roughly 60 km diameter circles, the divergence of the air mass can be computed directly as the closed integral of the cross-track winds around the circles. Mean vertical velocity can then be calculated by integrating the divergence with height above the surface. A key development that made this approach possible is the advent of GPS for improved accuracy measuring horizontal airplane velocity. Lidar observations of the inversion height permit a calculation of V_E.

The 1998 NCAR Publication Prize was awarded to several ACE-1 PIs for implementing these techniques to determine V_E. Russell et al. [1998] measured the submicron aerosol size distribution with a radial DMA and found different shapes of this distribution in adjoining layers of the atmosphere. They then used the changes in the aerosol spectrum in the SBML to estimate entrainment of air from the overlying air (which they called the buffer layer, BuL) into the SBML. Similarly, they used changes in the DMS concentration in the BuL to estimate entrainment of air into that layer from the SBML. Combining this with calculations based on the divergence and flux techniques gave them estimates of three entrainment rates: entrainment from the SBML to the BuL, from BuL to the SBML, and from the FT to the BuL.

During Lagrangian A the northeast side of our circular legs began to develop cloudiness and was clearly different from their southwest side. This heterogeneity provided evidence about the evolution of aerosol in a cloudy region [Varner, 1997]. This confirmed the importance of cloud processing over the Southern Ocean. In Lagrangian B we were able to conduct three flights while all three balloons remained aloft. Although both the wind direction and speed changed dramatically, the balloons maintained their relative positions (Figure 3). This argues that the target airmass remained coherent for the 30 hours of our observations, although there remained some differential advection due to changes of wind with height.

The evolution of sulfur species concentrations confirmed our picture of remote sulfur chemistry: DMS increased in the nighttime and dropped during the brightest sunlight, while SO_2 increased in the day and dropped at night [Suhre et al., 1998]. Photochemically-derived sulfate and MSA aerosol both increased in the daytime and held relatively constant at night [Huebert et al., 1998]. This is an excellent set of data against which to test marine sulfur models, but for one major problem: while it is likely that much of the NSS was being formed on seasalt [Sievering et al., 1999], our inability to quantitatively collect particles larger than 1 μm from an aircraft made it impossible to close the budget and assess the importance of this reaction pathway. In view of the potential importance of large particle fluxes, it is critical that we be able to sample total aerosols without artifact to close budgets during Lagrangian experiments.

Among the most important conclusions from the ACE-1 Lagrangians is that the evolution of the DMS concentration was consistent with a DMS surface flux as calculated by the Liss and Merlivat (L&M) parameterization [Mari et al., 1998; Russell et al., 1998]. In view of the very different flux predicted by the Wanninkhof relationship, it is significant that under this set of conditions L&M was more effective at predicting the evolution of DMS concentrations.

ACE-2

The strategy was further refined during ACE-2, in which balloons were supplemented with inert chemical tracers during three cloudy Lagrangian experiments of three UK C-130 flights each. Although ACE-2 was too recent to have comparisons with models in print, there is some very interesting behavior to challenge models. The first Lagrangian was in clean air, in which the accumulation mode concentration increased by fourfold as the Aitken mode concentration decreased. Seasalt is being considered as one of the possible sources of the increased accumulation mode number. The other two cloudy Lagrangian experiments were in polluted air. In one case the SO_2 concentration dropped from 800 to 40 pptv, while sulfate fraction increased and cloud processing decreased the Aitken mode concentrations. The other is apparently a case in which the precursor gases were consumed prior
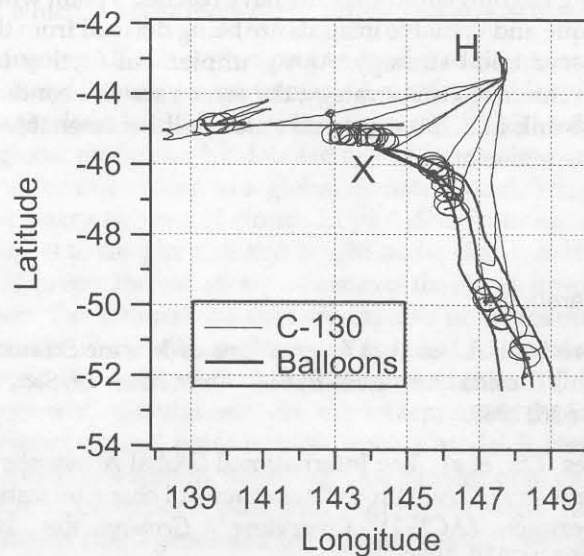

Figure 3. Circular C-130 flight tracks and balloon trajectories during ACE-1 Lagrangian B. X marks the location of the R/V Discoverer, the balloon-launch ship. H marks Hobart, Tasmania, where the aircraft was based.

to a series of cloud encounters, such that no change occurred over the course of one day. A series of cloud-free pseudo-Lagrangian experiments between surface measurements at Sagres, Portugal and a ship offshore present cases in which dry continental air evolves as it moves over the ocean. The ACE-2 Lagrangians will no doubt help modelers refine several process rates in their regional aerosol models.

The Future

Whether the aerosol optical depth is controlled by chemical growth or the mixing of airmasses with different relative humidities, we need to know what is controlling it in various regions so it can be modeled realistically. If indeed much of the aerosol evolution takes place near source regions, Lagrangian experiments should be conducted as close as practical to the sources, so we can observe the maximum evolution of species concentrations. One essential refinement for Lagrangians in airmasses moving off Asia is to use regional aerosol CTMs to predict where the various species will undergo the most measurable changes under a variety of scenarios. Models can suggest distances from sources and times where the changes are likely to be the most measurable. In that way we can gather data sets that will constrain the models most tightly. We also need to develop fast DMS instruments to improve our knowledge of entrainment rates, and thus to significantly reduce the uncertainty in derived process rates.

There are some needed insights that can only be gained by observing the changes in airmasses over time. By steadily and incrementally improving the technology for doing Lagrangian studies, we have reached a point where unique and valuable insights are being derived from this observational strategy. As we implement further improvements to this strategy, the error bars will continue to shrink and more process rates will be amenable to measurement.

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Aerosol-cloud interactions and indirect forcing

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The indirect climatic effect of aerosols refers to the influence of aerosols on cloud optical depth and albedo, and cloud lifetime, as a result of anthropogenic changes in the number and composition of cloud condensation nuclei (CCN). Tropospheric aerosols currently pose one of the largest uncertainties in prediction of climate forcing from anthropogenically induced changes in the composition of the atmosphere, and the most significant component of that uncertainty is associated with indirect forcing. The large uncertainty attending indirect forcing exists because of the complex sequence of phenomena that connects aerosols with CCN, CCN with cloud droplets, and cloud droplets with cloud albedo and cloud lifetime.

Because of the difficulty in isolating the CCN-cloud relationship in ambient experiments, there are few studies that demonstrate clearly the cause-and-effect relationship between increased CCN and changes in cloud droplet number concentration. Circumstantial evidence can be found in the fact that CCN concentrations are greater in continental air masses (exceeding 1000 cm^{-3}) than in the marine atmosphere (rarely exceeding 100 cm^{-3}) and that continental clouds tend to exhibit greater cloud droplet number concentrations than do marine clouds. Cloud droplet number concentrations have been found to increase with increasing aerosol loading [Pueschel et al., 1986]. Satellite data on mean cloud droplet radii in the Northern versus the Southern Hemisphere appear to reflect the effect of higher anthropogenic emissions of aerosols in the Northern Hemisphere [Han et al., 1994]. The existence of ship tracks, linear features of high cloud reflectivity found in marine stratus clouds, result from aerosols originating from ship stack exhaust emissions [Durkee et al., 1999a,b]. Indeed, ship tracks offer the most promising opportunity to isolate the CCN-cloud droplet number concentration linkage. We'll return to this later.

Because of the challenge of quantifying the relationship between changes in aerosol number and composition and cloud properties, current efforts at representing indirect climatic effects of aerosols in global climate models rely on empirical relations, such as that between cloud droplet number and sulfate mass concentration [Boucher and Lohmann, 1995], or on approximate analytical expressions that calculate the fraction of particles activated based on adiabatic parcel model simulations [Ghan et al., 1993; Abdul-Razzak et al., 1998]. To proceed beyond the purely empirical approach to representing aerosol-cloud interactions, it is necessary to be able to relate changes in atmospheric aerosol properties to changes in cloud properties. Key aerosol properties include particle size, number, and composition. Clouds form when aerosols are subjected to water supersaturations sufficient to cause activation. A coupling exists between the particles themselves and the water vapor field such that simulation of cloud formation depends upon aerosol concentration and CCN properties.

The classical approach to aerosol activation is Köhler theory, which assumes instantaneous equilibrium between the particle and the local water vapor saturation field; dynamic effects are not accounted for. We now know that time scales can be such that activation can be kinetically limited [Chuang et al., 1997; Ghan et al., 1999]. Classical Köhler theory also assumes a completely soluble solute in the drop, a specified fraction of which may be insoluble. It does not include two potentially important effects that serve to alter the critical particle size for activation: (1) Soluble gases, such as HNO_3, that can dissolve in the growing droplet, and (2) Partially soluble solutes, such as crustal species and organics, that continue to dissolve as more water is added to the drop [Kulmala et al., 1993; Laaksonen et al., 1998]. In addition, the chemical composition of the aerosol, and whether the aerosol is externally or internally mixed, affects the CCN behavior of the air mass [see, for example, Russell and Seinfeld, 1998 and Ghan et al., 1998]. The degree of mixing of the particle population is important because multiple aerosol types compete for existing water vapor, leading to a selective activation of those particles with the more facile activation properties. This effect is important if two populations are competing as CCN, one of which is anthropogenic in origin and one natural.

Coupling all this with a proper model of cloud formation and cloud microphysics is a daunting problem for a single cloud [see, for example, Kogan et al., 1996] let alone a global model; such a detailed model is out of the question for imbedding in a global climate model. Yet, it is necessary to predict cloud droplet distributions with respect to droplet size and height in the cloud, as these will govern the radiative properties of the cloud. It would seem that simulations that are capable of representing realistic in-cloud activation are essential for obtaining sound estimates of cloud radiative properties. While a large-eddy-simulation (LES) model represents the ideal from a physical point of view, such a model is simply too computationally intensive. One-dimensional parcel models, perhaps coupled with Lagrangian parcel trajectories in cloud generated from an LES model [Feingold et al., 1998], may represent an attractive approach to capture realistic cloud microphysics, while not incurring the extreme computational burden of an LES model simulation.

In short, the major issues that involve aerosol-cloud interactions in global models are: (1) Linking aerosol mass

with aerosol number. Currently, GCMs handle aerosol on a mass basis, which is sufficient for direct forcing; (2) Accurately determining the relationship between aerosol number and cloud droplet number. This relationship must consider aerosol composition and the role of soluble gases; (3) Accounting for the degree of mixing in the aerosol population. This would allow multiple aerosol types to compete for existing water vapor; (4) Incorporating cloud processing of aerosols. Chemistry taking place within cloud droplets is influential in aerosol-cloud cycles.

Now, we all know that this is a tall order, and several groups are actively pursuing these issues. When we have such a model for inclusion in a global aerosol model, how do we know that it is "correct"? The type of field program aimed at this question has been termed an aerosol-cloud closure experiment. Such a program was carried out during IGAC's Second Aerosol Characterization Experiment (ACE–2) under the CLOUDYCOLUMN experiment. The goal of an aerosol-cloud closure experiment is to measure below-cloud aerosol size and composition, in-cloud residual aerosol size and composition, and cloud drop number concentration and size distribution more or less simultaneously and then compare what is measured with what would be predicted theoretically based on the best understanding of the microphysics and chemistry involved. Multiple aircraft were involved in these types of experiments in ACE–2, and results are still being evaluated. The difficulties in making simultaneous below-cloud and in-cloud measurements of aerosol and cloud properties with a time resolution appropriate for aircraft speeds pose serious measurement challenges.

A sub-experiment within the aerosol-cloud closure experiment is the CCN closure experiment. In this experiment, aerosol size and composition are measured together with the CCN activation properties of the aerosol using an appropriate CCN instrument. Then one tests whether the CCN measurements agree with what one would predict from Köhler theory based on the measured aerosol size and composition. Again, there was an attempt at such a closure experiment during ACE–2. Measurements of CCN using either single supersaturation or supersaturation spectrum devices have long been a staple of atmospheric aerosol field campaigns. A few of these instruments have been applied on aircraft platforms. Measuring particle activation at realistic ambient cloud supersaturations in a controlled and precise way remains a continuing challenge, as is proper calibration of the devices in which these measurements are carried out. Several types of CCN instruments have been employed; a few of these instruments have been applied on aircraft platforms. A number of groups employ the classical static, thermal diffusion cloud chamber. In this device, a sample is drawn into a chamber between two horizontal, wetted plates that are maintained at different temperatures [Alofs and Carstens, 1976]. Particles in the middle between the

two plates are exposed to supersaturation as the steady state temperature and water vapor partial pressure profiles are established. The concentration of particles that activate is measured, either by light scattering from the ensemble of droplets in an illuminated volume, or by observations with a video camera. The limited growth time in such instruments restricts their use to relatively high critical supersaturations, i.e., 0.2%. Measurements of supersaturation spectra require changing the temperature difference, leading to long delays and, in flight experiments, poor spatial resolution.

Continuous-flow thermal diffusion cloud chambers have been constructed to overcome some of the complications that arise due to transient operation. The size and weight of these instruments has limited their use primarily to ground-based measurements, but they do introduce some interesting capabilities. Fukuta and Saxena [1979] developed a version in which a temperature gradient across the width of the flow channel was used to produce a transverse gradient in supersaturation. Measurements at different supersaturations could then be quickly made by shifting the observation point across the flow channel.

Hudson [1989] introduced a temperature profile in the flow direction to establish a streamwise gradient in the supersaturation. Particles that activate at low supersaturations have more time to grow than do those that require higher supersaturations to activate. Hudson infers the critical supersaturation from the activated droplet size measured with an optical particle counter at the outlet of the flow channel. Supersaturation spectra have been reported over the critical supersaturation range $0.01\% < S_c < 1\%$.

A new instrument developed at Caltech [Chuang *et al.*, 1999] employs an approach first proposed by Hoppel *et al.* [1979] to produce supersaturation in a tube flow. The supersaturation is produced by dividing a wet wall tube into a sequence of alternating heated and cooled segments. The hot segment provides water vapor while the cooled segment produces the desired supersaturation. Because of compact size, low weight, and simplicity it was possible to produce an autonomous instrument that could be flown aboard a small airplane without operator intervention. In ACE–2, this alternating gradient CCN counter provided measurements of the number concentration of CCN with critical supersaturations of 0.2% or below.

The relationship between CCN measurements obtained with the various instruments and activation in cloud-forming regions of the atmosphere remains unclear. None of the available instruments simulates the supersaturation-time histories experienced by atmospheric particles as they undergo activation. The thermal diffusion cloud chambers that operate at fixed supersaturation, i.e., the static thermal diffusion cloud chamber and the fixed supersaturation and transverse supersaturation

gradient flow instruments, are generally regarded as absolute instruments, but this can only be true for supersaturations sufficiently high that activation occurs within the limited residence time of the particles in the supersaturation region. The streamwise gradient instrument does not attempt to provide an exact simulation of cloud forming conditions. Because the measurements depend on the kinetics of droplet growth, the instrument must be calibrated. Still, the possibility of simultaneously acquiring the entire supersaturation distribution, including that at low critical supersaturations, has made this technique very attractive and led to its use in a number of campaigns

Even with a well-planned and conducted field measurement program, achieving aerosol-cloud closure is not an easy task. As mentioned above, the ship track phenomenon provides an unusual target of opportunity for studying aerosol-cloud interactions, particularly the effect of perturbations on the background environment caused by anthropogenic aerosols. The Monterey Area Ship Tracks (MAST) experiment, carried out in the summer of 1994 off Monterey, CA under support of the U.S. Office of Naval Research, provided a wealth of data on the phenomenon [special issue of *J. Atmos. Sci.* forthcoming], but the measurements did not address aerosol-cloud and CCN closure in the detail ultimately required. IGAC might consider a future mission or subset of a future mission in which ship tracks can be probed with the specific intention of evaluating aerosol-cloud modules that are to be used in global climate models.

Where are the gaps? Certainly there is much to do in the realm of translating detailed knowledge of cloud microphysics and chemistry into practical modules for global climate models. But perhaps an even greater challenge exists in the realm of executing the types of airborne measurements that can unambiguously establish the extent to which the atmosphere is behaving the way we think it is.

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Nuts and bolts of radiative forcing by mineral dust

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Interactions of dust with Earth's radiation field are more complex than those of most other atmospheric aerosols because mineral particles are able to scatter and absorb UV, visible, and infrared radiation, leading to either heating or cooling of the climate system. The direct radiative forcing is used to quantify a change in the planetary radiation balance. Negative forcing corresponds to cooling of the underlying surface-atmosphere system, while positive forcing corresponds to warming. At present, due to our limited knowledge of the properties of mineral aerosol, the magnitude and even the sign of its net (solar plus infrared) direct radiative forcing remains unclear. It is almost certainly positive in some locations and negative in others. The presence of dust alters the surface radiation budget, which affects surface temperature and thus various surface-air exchange processes. Furthermore, the atmospheric temperature profile and hence atmospheric dynamics may be affected by additional radiative heating or cooling occurring in the dust layer itself. Overall, the radiative impact of mineral aerosol is important relative to that of other types of aerosols—such as sulfates and smoke particles—due to the widespread distribution and large optical depth of mineral dust.

Part of the complexity in estimating dust radiative impact comes from the fact that dust sources and sinks are not uniformly distributed, and that the lifetime of mineral aerosol in the atmosphere is relatively short, at most a few weeks. Consequently the dust burden has a complex spatial and temporal pattern. Moreover, the physical and chemical characteristics of dust may evolve during the transport. Some of the major processes governing the evolution of the particle size spectrum have been identified and included in climate models (such as dust emission, sedimentation, and dry and wet removal), while others have been only hypothetically formulated (such as heterogeneous chemistry on dust particle surfaces, cloud processing, and interactions with other atmospheric aerosols).

A further complication arises because mineral aerosol is a collective term referring to widely varying mixtures that may include many constituents, including quartz, various clays (mainly kaolinite, illite, and montmorillonite), calcite, gypsum, hematite and others. These minerals each have very different physical and chemical properties (e.g., size spectrum, particle shape, density, solubility, chemical reactivity). The abundance of the various constituents depends on the place of dust origin, how it was mobilized, and physical and chemical transformation processes occurring during transport. Furthermore, because spectral optical constants (or refractive indices) vary widely from mineral to mineral, the optical properties of dust are determined by the relative abundance of each mineral and the details of how the minerals are mixed together. The differences in the refractive indices of dust samples collected at diverse locations cause large variations in the major dust optical characteristics and, consequently, in dust radiative impact [Sokolik et al., 1998]. No climate model has yet included this aspect of the radiative forcing problem.

In addition, the interaction of dust particles with other atmospheric aerosols and clouds can affect their properties. For instance, studies by Parungo et al. [1996] demonstrate that dust particles originating from China's desert are coated by sulfates or soot after passing through polluted industrial regions downwind of the desert. In contrast, Saharan dust transported over the Atlantic Ocean is often coated by sea-salt. Dust particles internally mixed with soot, sulfates, nitrates or aqueous solutions can have drastically different properties from those which are evident at the dust source. The ability of dust particles to scatter and absorb light can be altered in different ways depending on which species aggregate with dust particles. To illustrate, Figure 1 shows how the single scattering albedo of internally-mixed multicomponent particles changes as a function of the ratio of dust volume to total particle volume. The single scattering albedo is a key optical characteristic for calculating the heating or cooling effects of aerosols. As the fraction of dust increases from 0 to 1, the single scattering albedo of aggregates of dust-water (solid curve) and dust-sulfates (dotted curve) decreases, resulting in more heating. In turn, aggregation of dust with soot particles causes a decrease in the single scattering albedo (dashed curve), enhancing heating.

Coagulation of dust with other aerosol or cloud particles as well as uptake of atmospheric gases on dust particle surfaces followed by heterogeneous chemical reactions may both result in the formation of multicomponent aerosols (MCA). However, the relative importance of these processes remains unknown. Unfortunately, there is no reliable theory for modeling optical constants of the MCA aggregates [Sokolik and Toon, 1999].

While heterogeneous chemical reactions may be important in the formation of an internal mixture of dust with other aerosol compounds, in turn, the heterogeneous reactions on dust surfaces can be crucial in understanding tropospheric chemistry. There is growing evidence that heterogeneous chemical reactions on airborne mineral aerosol surfaces can play an important role in the tropospheric chemistry of SO_X, NO_Y and O_3 [Dentener et al., 1996]. To model the related chemical reactions, the size-resolved mineralogical composition of dust is required. For instance, the removal of SO_2 and HNO_3 from the gas phase depends on the alkalinity of dust aerosols, so the calcium content needs to be known. Hydrogen

radicals like HO_2 and OH react on atmospheric particles through pathways involving redox reactions with iron, so assessments of this process depend on measurements of Fe^{3+} abundance in dust samples. Furthermore, sulfates and nitrates can be formed via heterogeneous chemistry on dust particle surfaces, resulting in drastically modified radiative properties discussed above.

Therefore, incorporation of regionally and temporally varying size-resolved dust mineralogical composition into global and regional climate models is a desirable and promising approach to decrease the currently large uncertainties in the assessment of radiative forcing by the natural and anthropogenic components of the airborne mineral aerosol. Accomplishing this will require a great deal of effort to 1) make the relevant observations (surveys) of dust layers at various altitudes and locations, 2) achieve a better understanding of related physical and chemical process rates.

Although a large body of data on ground-level dust properties already exists, it is of limited usefulness because in most cases only a few chemical or physical properties were measured, in isolation from the others. In particular, a large gap exists between measurements of dust particle size distribution, optics, radiation, and chemistry. These need to be measured simultaneously (as in closure experiments) to test the hypothetical relationships between them. Another limitation is the deficiency found in different measurement techniques. Improvements are
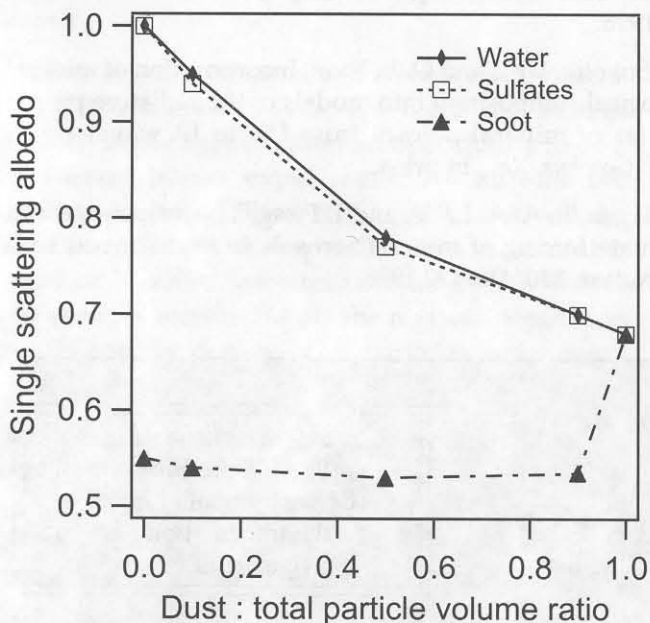

Figure 1. Single scattering albedo calculated at wavelength of 500 nm as a function of the ratio of dust volume to total particle volume. Solid curve is for dust particles aggregated with water (refractive index, n = 1.335); the dotted curve is for dust with sulfates (n = 1.55), and the dashed curve is for dust aggregated with soot (n = 1.96 - 0.66i).

sorely needed in the methods employed to determine the composition of individual particles, their shapes, spectral refractive indices, aerosol absorption of light, and aerosol scattering phase function.

To gain a better understanding of diverse dust impact, a promising strategy is to perform comprehensive coordinated measurements of the physical and chemical properties of dust in targeted regions, on various space and time scales. Saharan, Central Asian, and Arabian Peninsula regions, which are the most important dust sources, are of primary importance. In this context, the ACE-Asia international experiment, focusing on Central Asia, will provide a unique opportunity to obtain coordinated measurements of dust chemical and physical properties at the dust source, in industrial and urban areas, and in the marine environment. Such measurements are urgently needed to obtain a quantitative understanding of the spatial and temporal variations of the Asian mineral aerosol and to determine the extent to which these variations are important for radiative and climate model simulations.

The above discussion reveals two major issues which must be resolved to improve modeling and prediction of dust impact: 1) quantification of dust production from both natural and anthropogenic sources, and 2) identification of the main processes controlling the evolution of the physical and chemical properties of dust during its life cycle.

Dust production

Information is needed on production rates and size, space and time scales from both natural and anthropogenic sources. There is a large body of data on the mineralogical and chemical nature of the Earth's soils, which demonstrates the complex spatial variability of soil composition. Existing global data sets of soil properties currently include soil texture and types but they do not provide information on size-resolved mineralogical composition of the parent soil. There is a clear need for a new data set to provide this missing information.

Along with the composition of bed surfaces, dust mobilization processes partly determine the initial particle size distribution of airborne minerals as well as their composition and the degree of particle aggregation. Therefore the airborne dust may exist as an external mixture of individual minerals and/or as a mixture of aggregated particles. As was demonstrated by Sokolik and Toon [1999] for a given composition and under similar atmospheric conditions, a mixture of aggregates can cause positive radiative forcing while a mixture of individual minerals gives negative forcing.

Significant progress has been made recently in incorporating surface mineralogy and surface roughness into dust emission schemes to simulate the size-resolved dust vertical flux from Saharan sources [e.g., Marticorena and

Bergametti, 1995]. Such schemes can be extended to simulate size- and composition-resolved fluxes of dust from other major production regions (Africa, Asia, Middle East, India, Australia and USA). The overall goal is to quantify dust emission and relate the physical and chemical properties of dust to the source.

Evolution of dust properties during the dust life cycle

During dust transport, its initial characteristics, determined by a given dust source and production mechanism, may be altered by the following processes: deposition, sedimentation, wet and dry removal, heterogeneous nucleation, coagulation with other aerosols, heterogeneous chemistry, and cloud processing. It is important to establish the relative roles and characteristic scales of all these processes. This is a difficult problem due to the nonlinearity of the processes, possible feedbacks, and the overlap in time scales between aerosol processes and atmospheric dynamics.

Recent global and regional dust models consider only a few processes (dust production, dry and wet removal, and deposition) responsible for the evolution of particle size distribution, ignoring composition of mineral aerosol, coagulation, and heterogeneous processing [e.g., Tegen *et al.*, 1996]. Dust composition is so fundamental that it must be included for models to be realistic. In turn, observations of dust composition will help to develop better physically-based treatments of the processes needed for climate models and for remote sensing applications. For instance, wet removal of dust is currently parameterized in models in an oversimplified fashion. If dust composition (and hence its hygroscopicity) were known, however, the treatment of this process could be greatly improved.

It would be an impossible task to incorporate every naturally occurring mineral into climate models. A practical alternative is to take into account a handful of major mineral components to correctly represent dust spectral optical properties and its heterogeneous chemical process rates. A multidisciplinary coordinated program of laboratory measurements, *in situ* and satellite observations together with modeling studies will be required to advance our knowledge in this area.

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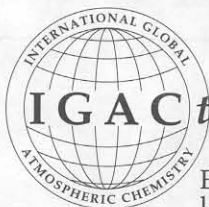
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