IGACtivities NewsLetter

of the International Global Atmospheric Chemistry Project

A Note From the IGAC Chair
Guy Brasseur

Integration and Synthesis

During the last decade the international atmospheric chemistry community has devoted a great deal of effort to defining and implementing large field campaigns. These campaigns have focused on questions related to tropospheric photochemical processes, aerosol characterization, and biosphere/atmosphere interactions, as well as the impact of fires on atmospheric composition. This approach has been justified by the fact that a large number of fundamental processes have to be studied and quantified before societally important issues can be addressed.

With a large amount of data becoming available, time has come to integrate our findings within a coherent framework. What have we really learned in the last 10 years, especially from IGAC-sponsored campaigns? What are the remaining fundamental unknowns, and what is the best approach to address them?

At its last meeting held in Nagoya, Japan, in early November, the IGAC Scientific Steering Committee decided to put more emphasis on integrating and synthesizing the knowledge acquired by the community. The intention is to produce within 2 years or so a comprehensive document that highlights the progress made by the scientific community, and presents answers to well-stated, interdisciplinary questions related to IGBP objectives. This "exercise" will involve a large number of scientific leaders who have played key roles in past scientific research, including those who have contributed to IGAC's sponsored Activities.

The meeting in Nagoya also provided the opportunity for IGAC to refocus its Activities around three scientific themes: (1) Biosphere-Atmosphere Interactions (with emphasis on the exchanges of gases and aerosols between the atmosphere and the continental biosphere, and between the atmosphere and the ocean); (2) Oxidants and Photochemistry (with emphasis on global tropospheric ozone and its precursors); and (3) Atmospheric Aerosols (with emphasis on aerosol properties and their impact on radiative forcing). In addition, several fundamental and cross-cutting Activities will remain on the agenda of IGAC. The effort towards education and capacity building will also remain important.

During the Nagoya meeting, the Steering Committee heard an excellent report on the recent ACE-2 campaign, which took place in the eastern North Atlantic. The purpose of this field study was to assess the role of Saharan dust and anthropogenically produced aerosols in regional climate. A new

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ACE experiment to take place in the North Pacific and focusing on the role of Asian sources in the formation of aerosols in this part of the world is currently being planned under the auspices of IGAC. This issue of the Newsletter includes a brief summary of ACE-2 highlights along with several other articles related to aerosols and climate forcing.

Another important report presented at Nagoya focused on the progress made to plan for the newly created *Global Tropospheric Ozone Project*. This project will coordinate a large number of current and planned initiatives aimed at understanding the chemical and physical processes that control ozone in the global troposphere. A scientific plan is being prepared by a group of specialists and will serve as the basis for this important IGAC initiative.

Finally, I would like to thank and congratulate our Japanese colleagues who organized the *IGAC Symposium on Atmospheric Chemistry and Future Global Environment* which was also held in Nagoya. More than 200 participants attended this very interesting scientific symposium, which demonstrated the vitality of atmospheric scientists in Asia, and specifically in Japan.

Foreword

Atmospheric Aerosols: To the Forefront of Research in Atmospheric Chemistry

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The chemistry of the atmosphere has major impacts on several important environmental factors: climate, the hydrological cycle, acid precipitation, stratospheric and tropospheric ozone, and local and regional air quality. Atmospheric chemistry research has gone through several phases. Until about 1970, research had mainly concentrated on aerosols, carbon dioxide, transport of radioactive materials, and ozone in and between the stratosphere and the troposphere, and air pollution. By the beginning of the 1970´s several fundamental new research areas took over: acid rain, the role of the biosphere in the cycles of trace gases, the importance of photochemical, largely catalytic reactions in the stratosphere and troposphere, the consequences of the release of NO from aircraft and of chlorofluorocarbons for stratospheric ozone, the essential role of HO radicals as the 'detergent' of the atmosphere, and the importance of tropospheric ozone as a precursor of HO radicals and a major driver of atmospheric chemistry. Looking back, it is fair to say that between 1970 and 1990 the major advances in atmospheric chemistry were made in gas phase photochemistry, maybe except for a brief intermezzo of 'nuclear winter' studies.

This picture is now very much changing. The studies by Charlson, Rodhe, Lovelock and their colleagues have again emphasized the substantial role of natural and anthropogenic sulfate aerosol for the radiative properties of the atmosphere and Earth's climate. Studies on the causes of the Antarctic ozone hole have clearly demonstrated the large role of reactions that can take place only on ice and aerosol surfaces. When such reactions are possible in the stratosphere, the more they can influence the troposphere with its abundance of various kinds of aerosol. Considering these factors, and especially because of various breakthroughs in experimental techniques, there is very little doubt that aerosol research will again be prominent in atmospheric chemistry research in the coming decades. This research will involve process studies both in the atmosphere and in laboratories, studies on the sources and sinks of aerosols, chemical analyses of the particulate matter, modeling, and especially regional (campaigns) and global (satellites) observations on the distribution of the atmospheric aerosol. This is all the more important because climate models which now in most cases involve only sulfur chemistry have not been sufficiently tested against observations, despite the potentially great cooling effects of aerosol on Earth surface temperatures which so far may well have substantially counteracted the radiative forcings by the greenhouse gases.

The following articles highlight various aspects of new developments in aerosol research. Hobbs calls attention to the great abundance of the organic component of the aerosol. The origin of the organics, natural and/or anthropogenic, is a matter of great interest, especially in view of the huge production of organics over the disappearing tropical forests. Pszenny and colleagues address new research, pointing to a role of sea salt aerosol in the chemistry of the marine atmosphere. Bergametti and Dulac emphasize the major importance of mineral aerosol for Earth's radiation budget and as a scavenger of gases such as SO_2, H_2SO_4, and HNO_3. Raes and Bates present some preliminary findings which were obtained during the highly international IGAC ACE-2 field investigations over the northeastern Atlantic.

All these papers clearly point to a highly exciting future. They emphasize the importance of high quality measurements. Combined with the appropriate gas phase measurements they will teach us a lot about which processes are responsible for gas-to-particle and particle-to-gas transformations, including how reactions on particles affect the gas phase photochemistry of the atmosphere.

Sulfate Versus Carbonaceous Materials on the East Coast of the United States: Results from TARFOX

Contributed by P. V. Hobbs, University of Washington, USA

Introduction

One of the main goals of the Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX), carried out just off the East Coast of the United States from 10-31 July 1996, was to measure the direct effects of tropospheric aerosols on regional radiation budgets while simultaneously measuring their chemical and physical properties (Russell *et al.*, 1996a). An earlier article in this NewsLetter described the conduct of the TARFOX field experiment (Russell *et al.*, 1996b). In this article, a summary is given of some important (and surprizing) results from TARFOX concerning the relative contributions of various chemical species to the total aerosol mass and the aerosol optical depth (a determinant of aerosol radiative forcing). Full accounts of these results will appear shortly in the *Journal of Geophysical Research* (Novakov *et al.*, 1998; Hegg *et al.*, 1998). All of the measurements described here were obtained aboard the University of Washington's (UW) Convair C-131A research aircraft.
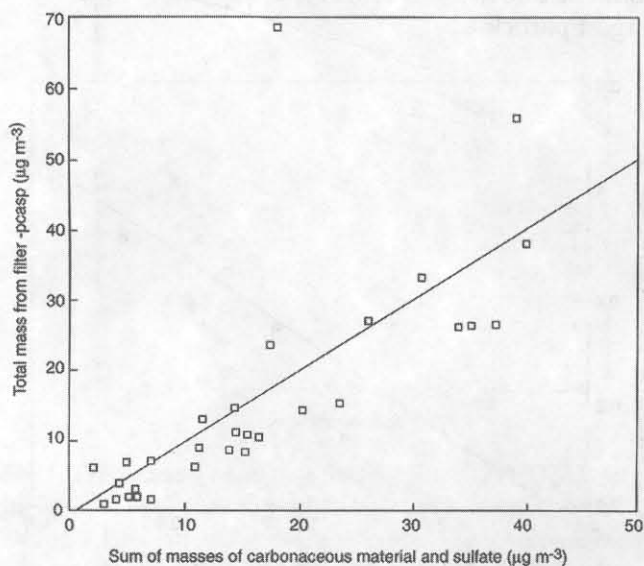

Figure 1. Total dry aerosol mass (from weighing of filters and PMS PCASP measurements) versus the sum of the dry masses of carbonaceous materials and sulfate. (From Hegg et al., 1998.)
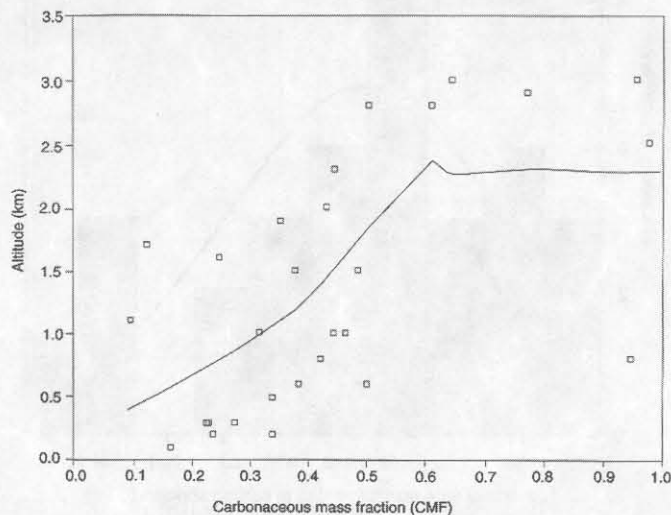

Figure 2. Altitude dependence of the carbonaceous aerosol mass fraction. (From Novakov et al., 1998.)

Most studies of aerosol radiative forcing have dealt with just a single component of the atmospheric aerosol, namely, sulfate. However, chemical species other than sulfate contribute to the sub-micrometer particles in the air that play a role in radiative forcing. Of particular importance in this regard are carbonaceous species, which derive from biogenic sources (organic carbon) and from the incomplete combustion of fossil and biomass fuels (black carbon as well as organic carbon). Both organic and black carbon affect the extinction of solar radiation, and therefore aerosol radiative forcing. Black carbon is the principal light-absorbing aerosol species in the atmosphere, while both organic and black carbon scatter solar radiation.

Measurements at ground level show considerable variability in the relative amounts of sulfate and carbonaceous material (e.g., Malm *et al.*, 1994). However, aerosol radiative forcing is determined by aerosol properties throughout the vertical column, about which much less is known. Therefore, in TARFOX, the UW aircraft was used to obtain information on the composition and physical properties of aerosols in vertical columns in the lower troposphere (up to altitudes of about 3 km, which encompassed the main aerosol layers). At the same time, the total aerosol optical depth above the aircraft was measured continuously with a sunphotometer mounted on the aircraft. The techniques used to obtain the various measurements, and the methods of data analyses, are described in detail by Novakov *et al.* (1998) and Hegg *et al.* (1998).
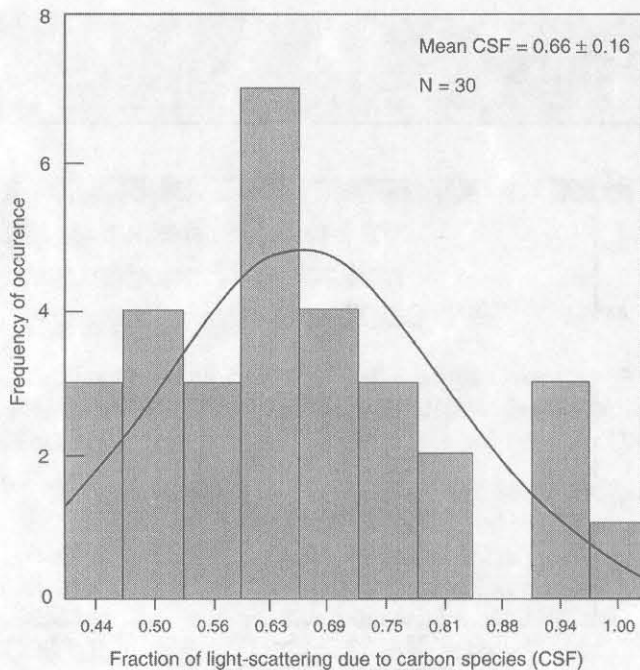
Figure 3. Frequency distribution of the fractional contribution of carbonaceous materials to the dry aerosol light scattering. (From Hegg et al., 1998.)

Components of the Total Aerosol Mass

Figure 1 (see page 3) shows point measurements of total (dry) aerosol mass versus the sum of the masses of sulfate and carbonaceous materials. (Sulfate was the only significant inorganic constituent of the aerosol by mass.) It can be seen from this figure that sulfate and carbonaceous materials generally accounted for virtually all of the aerosol mass. On average, carbonaceous materials contributed about 50% to the total (dry) aerosol mass.

Altitude Dependence of the Carbonaceous Mass Fraction

Figure 2 (see page 3) shows measurements of the ratio of the mass of carbonaceous materials to the total aerosol mass (the carbonaceous mass fraction or CMF) as a function of altitude for the TARFOX data. It can bee seen that the CMF generally increased with increasing altitude. Near the surface, the CMF was 10-40%, in agreement with previous ground-based measurements.

However, aloft the CMF is substantially larger, reaching values as large as 90% at 3 km. One possible cause for this altitude dependence could be that sulfate is removed from the atmosphere by cloud and precipitation processes more efficiently than carbonaceous materials. In this case, carbonaceous materials would have longer lifetimes in the atmosphere than sulfate, and therefore be carried to higher altitudes.

Contribution of Carbonaceous Materials to the Light-Scattering Coefficient of the Dried Aerosol

Figure 3 shows the frequencies with which carbonaceous materials contributed various fractions to the (dried) aerosol light-scattering coefficient. The average fraction of the light-scattering coefficient contributed by carbonaceous materials was 66±16%. Therefore, on average, the majority of the light scattering of the dried aerosol derived from carbonaceous materials.

Comparison of Remote Sensing and In Situ Measurements of Aerosol Optical Depths

The optical depths of the aerosol in the strongly polluted lower layers of the atmosphere, which the aircraft profiled, were derived from the *in situ* measurements of the light-scattering and light-absorption of the dried aerosol, the vertical profile of relative humidity (RH), and the measured humidification factors of the aerosol (i.e., the change in aerosol light scattering with RH). These derived values can be compared with optical depths measured directly with the sunphotometer aboard the aircraft (Livingston and Russell, 1997). Since the sunphotometer measured the optical depth of the aerosol in the total vertical column above the aircraft, values measured at the top and bottom of the aircraft profile were differenced to obtain the optical depth of the layer sampled by the aircraft. The results of these comparisons are shown in Figure 4, where it can be seen that the two independent measurements of layer optical depth are well correlated. The sunphotometer measurements are systematically slightly higher than the *in situ* measurements; this could be due to inefficient *in situ* sampling of the largest particles.
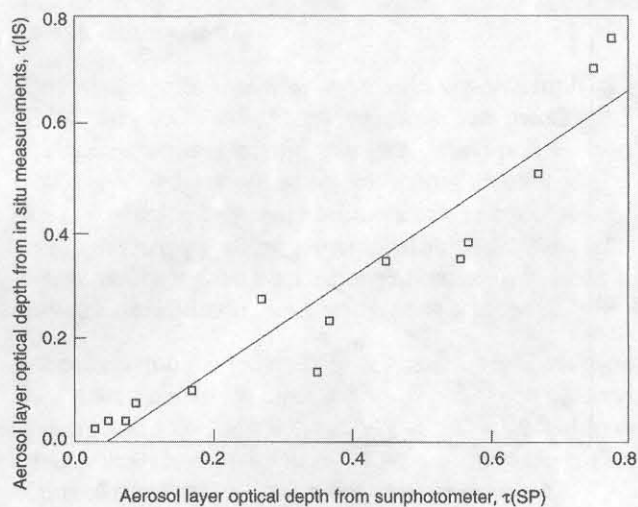

Figure 4. Layer optical depths measured with the airborne sunphotometer versus those derived from in situ airborne measurements of aerosol properties in the same layer. (From Hegg et al., 1998.)
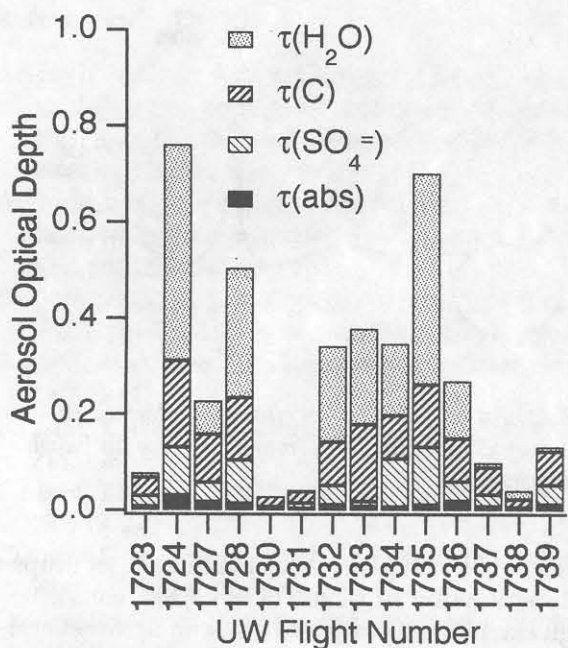
Figure 5. Contributions of the light scattering by water condensed on aerosols, carbonaceous materials, and sulfate in the lower troposphere to the column aerosol optical depth for each of fourteen University of Washington flights off the East Coast of the United States. Also shown is the contribution to the optical depth of light absorption by the aerosols. (From Hegg et al., 1998.)

Contributions of Chemical Species to the Total Aerosol Column Optical Depth

Figure 5 shows the contributions of sulfate, carbonaceous materials, and water condensed on the aerosols, to the column aerosol optical depth derived from *in situ* measurements on each of fourteen flights made in TARFOX. These results show that:

- Condensed water, carbonaceous materials, and sulfate accounted for most of the aerosol optical depth in the column sampled by the aircraft.

- On average, scattering by condensed water accounted for about 35% of the aerosol optical depth. However, for optical depths greater than 0.4, condensed water accounted, on average, for 58% of the optical depth. (These results highlight the importance of accurate measurements of the hygroscopicity of various aerosol types, and the careful incorporation of such information in modeling studies of aerosol radiative forcing.)

- Second to condensed water in contributing to the aerosol optical depth were carbonaceous materials, followed by sulfate.

- Dry sulfate in the lower troposphere contributed, on average, only 16% to the total aerosol optical depth. Even using a generous value of 3 for the hygroscopic growth factor of sulfate, wet sulfate contributed less than 50% to the total aerosol optical depth. Using a more reasonable value of 2 for the humidification factor of sulfate, its contribution to the optical depth was, on average, only 30%.

Conclusions

The widely held view that sulfate dominates aerosol column optical depths, and therefore aerosol radiative forcing, was not verified in TARFOX measurements on the East Coast of the United States, even though this is one of the most likely places on Earth for this paradigm to hold. Instead carbonaceous materials were more important. While one data set collected in one location is by no means definitive, it indicates that much more attention needs to be placed on carbonaceous materials in the atmosphere, and that measurements similar to those obtained in TARFOX should be made in other airsheds around the world.

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Sea Salt Aerosols, Tropospheric Sulfur Cycling, and Climate Forcing

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Introduction

Tropospheric aerosols influence the Earth's radiative balance directly by scattering and absorbing solar radiation and indirectly by altering cloud albedo (e.g., Houghton et al., 1996). Chemical processes involving aerosols also affect the oxidative state of the troposphere and, thus, the atmospheric lifetimes of important trace gases such as methane, ozone, nonmethane hydrocarbons (NMHC), and dimethyl-sulfide (DMS). Because DMS and other sulfur (S) compounds are integrally involved in these processes, understanding the transformations underlying S cycling is prerequisite for a realistic predictive capability for tropospheric oxidizing capacity and climate forcing by aerosols.

Ten years ago, Robert Charlson, James Lovelock, Meinrat Andreae, and Steven Warren postulated a natural negative feedback mechanism involving microscopic oceanic plants which may partly offset positive climate forcing resulting from increased anthropogenic emissions of long-lived "greenhouse" gases (Charlson et al., 1987). Among the testable scientific hypotheses described in that provocative paper is that DMS emissions from some species of marine phytoplankton are the main source of non-seasalt sulfate in the marine atmosphere and that this nss-sulfate forms most of the cloud condensation nuclei in remote, unpolluted marine air. A decade later we are still not certain whether this hypothesis is true or not. In this article we summarize some recent research in this area.

What are "Sea Salt Aerosols"?

Sea salt aerosols are produced at the ocean surface by the bursting of air bubbles resulting from entrainment of air induced by wind stress (e.g., Blanchard, 1983; Gong et al., 1997 and references therein). Production occurs mainly in the "whitecaps" which are familiar to anyone who has seen a stretch of ocean on a windy day. On average, about 1% of the global ocean is covered by whitecaps at any given time. On bursting, the bubbles produce both film drops from disintegration of the bubble film cap and jet drops from collapse of the bubble cavity (see Figure 1). Depending on its
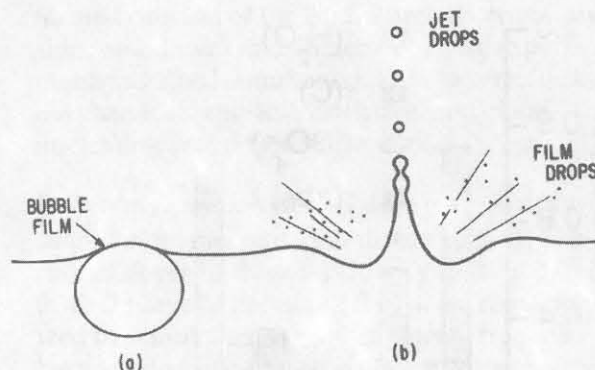

Figure 1. (a) An air bubble at an air-water interface. (b) The production of jet and film drops from a bursting air bubble. After Blanchard (1983).

size, each bubble can generate as many as 10 jet drops with a typical radius of 1-2 μm (1 μm = 10^{-6} meter), although extending to sizes >10 μm, and up to several hundred film drops in the sub-μm size range. Sea salt aerosol number and mass concentrations are strongly dependent on wind speed, typically following exponential relationships. In addition, at high wind speeds (above 7-11 m s^{-1}) the tearing of wave crests introduces ultra-large spume particles into the air. The sea salt aerosol size distribution extends from about 0.05 to 150 μm radius and, whilst number concentration is dominated by sub-μm film drops, surface area is dominated by the super-μm jet drops and volume is dominated by giant spume drops when present (e.g., O'Dowd et al., 1997a). To put the amount of sea salt aerosol in perspective, consider Andreae's (1995) estimates that in terms of mass, the five largest components of the atmospheric aerosol assemblage are: mineral aerosols from soil deflation (global burden = 16.4 x 10^{12} g), sea salt (3.6 x 10^{12} g), natural and anthropogenic sulfates (3.1 x 10^{12} g), biomass burning products (1.8 x 10^{12} g), and organics from NMHC oxidation (1.3 x 10^{12} g). Based on recent estimates of the integrated production flux of sea salt (3.5 x 10^{15} g y^{-1}) and the average sea salt mass-weighted atmospheric residence time (40 h; Erickson et al., 1997), we calculate a larger average global sea salt burden of about 15 x 10^{12} g.

What is "Non-Seasalt Sulfate"?

A given parcel of marine boundary layer (MBL) air will contain all of the above aerosol types (and others) in various proportions. Although sea salt usually dominates on a mass basis, apportioning the amounts of the different types present can be difficult. As their name suggests, fresh sea salt aerosols have an ionic composition similar to that of sea water. They rapidly dehydrate into equilibrium with ambient water vapor and undergo other composition changes involving the

scavenging of reactive trace gases, chemical reactions in the aqueous solutions (or on the surface of sea salt crystals under low humidity conditions), and the volatilization of products. There is a lot of sulfate in sea water—about 0.3% by mass—and this "sea salt sulfate" must be accounted for whether in aerosols, cloud water, or precipitation. Fortunately, the ratios of major ions, including sulfate, appear to be conserved in the sea salt aerosol production process itself. Thus, nss-sulfate resulting from post-production processes can be differentiated from sea salt sulfate by i) measuring a chemical proxy for sea salt (typically sodium or magnesium), ii) calculating the amount of sea salt sulfate present based on its ratio to the proxy in surface sea water, and iii) subtracting this sea salt fraction from the measured total concentration of sulfate to yield the nss amount.

The major sources of nss-sulfate in the marine boundary layer (MBL) are oxidation of DMS produced by marine phytoplankton and oxidation of sulfur dioxide (SO_2) from anthropogenic sources (mainly burning of S-laden fossil fuels) on the continents. Fossil fuel burning now accounts for about two-thirds of S emissions to the atmosphere globally and this proportion is projected to increase as population expands and the demand for energy increases, particularly in Asia. The mechanism of DMS conversion to nss-sulfate is not completely understood. It is a combination of multi-step photochemical reactions likely involving dozens of intermediate species (e.g., Yin et al., 1990) of which only SO_2 and a very few others have been measured in ambient MBL air. SO_2 from DMS oxidation and from anthropogenic sources is converted to aerosol nss-sulfate via two major pathways: i) direct scavenging of SO_2 by deliquesced aerosols and cloud droplets followed by aqueous phase oxidation to produce sulfuric acid (H_2SO_4), and ii) SO_2 oxidation by hydroxyl radical in the gas phase to produce H_2SO_4 vapor which either condenses onto pre-existing aerosols or, when aerosol concentrations are very low, combines with water vapor and ammonia to nucleate new, sulfate-based aerosols.

SO_2 Oxidation in Sea Salt Aerosols

Scavenging of SO_2 and the subsequent oxidation of dissolved S_{IV}[1] by ozone under alkaline conditions has widely been reported to be the principal source of nss-sulfate in the sea salt aerosol (e.g., Chameides and Stelson, 1992). Production via this pathway is limited

by diffusion of SO_2 into the aerosol and available alkalinity. Alkalinity is assumed to derive solely from the parent sea water in amounts conservative with respect to sodium or magnesium, and it is titrated rapidly (<1 hour) by H_2SO_4 produced from S_{IV} oxidation. This titration lowers solution pH and depresses SO_2 solubility, and the S_{IV} + ozone pathway becomes negligible. H_2SO_4 vapor produced by gas phase oxidation of SO_2 and scavenged directly from the gas phase (along with nitric, formic, and other acids) also titrates alkalinity, thereby diminishing the potential for nss-sulfate production via the S_{IV} + ozone pathway (Keene et al., 1997a). Because its solubility in alkaline solution is low, ammonia (NH_3) is not significantly scavenged by fresh sea salt; thus alkalinity produced by NH_3 dissolution does not sustain nss-sulfate production in sea salt aerosols by ozone oxidation of SO_2. Available sea salt alkalinity therefore appears to represent an upper limit for nss-sulfate produced in sea salt aerosols via S_{IV} + ozone. However, ozone

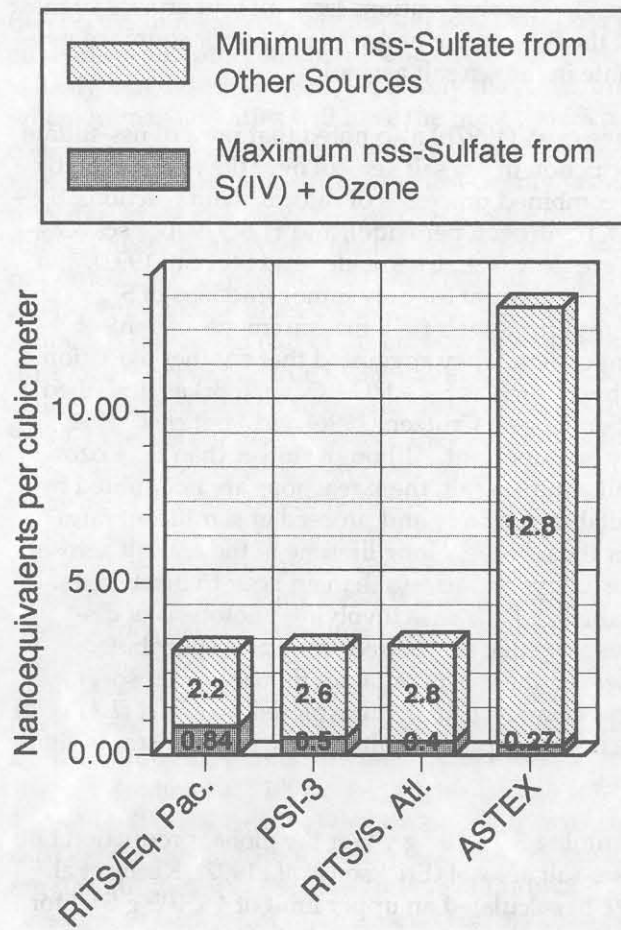Figure 2. Maximum amounts of nss-sulfate from S_{IV} + ozone in super-µm diameter MBL aerosols compared with the minimum amounts produced through other pathways. Values are arithmetic means for cascade impactor samples collected during four cruises in various open-ocean regions. After Keene et al. (1997b).

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[1] The subscript refers to the "oxidation number" of +4 of the S atom in SO_2 and its family of aqueous dissolution products, i.e., bisulfite (HSO_3^-) and sulfite ($SO_3^=$). The S atom in sulfate ($SO_4^=$) has a higher oxidation number of +6 and is a member of the S_{VI} family. Oxidation numbers are discussed in most general chemistry texts.

oxidation of SO_2 in cloud droplets activated upon sea salt nuclei may also be important (see next section).

Keene *et al.* (1997b) used measurements of sea salt aerosol composition obtained during four research cruises in several open Atlantic and Pacific Ocean regions to gauge the importance of $S_{IV} + O_3$ in super-µm sea salt aerosols. Size-segregated marine aerosol was sampled over 0.5- to 2-day periods with high-volume cascade impactors deployed on the bows of ships. The *maximum* nss-sulfate produced from S_{IV} + ozone (S_{ozone}) in each aerosol size fraction was estimated as the lesser of calculated sea salt alkalinity or measured nss-sulfate. The *minimum* nss-sulfate produced by combined contributions from other pathways was estimated from the residual (i.e., S_{other} = measured nss-sulfate – S_{ozone}). S_{ozone} and S_{other} were then summed over the super-µm diameter size range (which contained >95% of sea salt aerosol mass) and averaged across all samples obtained during each cruise. The results are summarized in Figure 2 (see page 7). The observations from all four cruises suggest that the S_{IV} + ozone pathway is a minor source of nss-sulfate in the sea salt aerosol.

Keene et al. (1997b) also noted that rates of nss-sulfate production in sea salt aerosol over the open ocean by the combined processes of radical chain reactions, S_{IV} + H_2O_2 (hydrogen peroxide), and H_2SO_4 vapor scavenging are slow (e.g., Chameides and Stelson, 1992), and thus can account for only minor fractions of S_{other} inferred from their field measurements of aerosol composition. They suggested that another oxidation pathway such as S_{IV} + HOX (X = Cl, Br) as described by Sander and Crutzen (1996) and Vogt *et al.* (1996) may be important. Although slower than S_{IV} + ozone in alkaline sea salt, these reactions are not limited by available alkalinity and proceed at significant rates over the relatively long lifetime of the sea salt aerosol (1 or 2 days versus less than an hour to titrate sea salt alkalinity). Pathways involving photolysis of dissolved organic compounds may also contribute; however, the role of organics in marine aerosol chemistry remains virtually unexplored. Keene *et al.* (1997a) discuss these possible alternative mechanisms in more detail.

Assuming 3.5×10^{15} g y^{-1} for the global production rate of sea salt aerosol (Erickson *et al.*, 1997), Keene et al. (1997b) calculated an upper limit of 4×10^{12} g S y^{-1} for the production rate of nss-sulfate from S_{IV} + ozone. Their cruise results indicate that actual nss-sulfate concentrations in sea salt aerosols typically exceed this limit by factors of 3 to >10, suggesting an overall global flux of nss-sulfate via sea salt aerosols in the range of 12 to perhaps 40×10^{12} g S yr^{-1}. Assuming that the wet- and dry-deposition fluxes of SO_2 are simu-

lated accurately in the absence of sea salt, this range corresponds to 25% to 80% of the approximately 50×10^{12} g S yr^{-1} produced from SO_2 oxidation in global models. Simple calculations based on percentages of nss-sulfate associated with super-µm sea salt aerosol (10-25%) together with estimated atmospheric lifetimes against deposition for sea salt (1 to 2 days) versus those for total nss-sulfate (4 to 5 days) yield a similar range of relative fluxes over the ocean. The accumulation of nss-sulfate in super-µm sea salt aerosols and its subsequent deposition appears to be a major pathway in the global S budget. This shunt of S away from production of new aerosols via SO_2 oxidation in the vapor phase is a process that should be accounted for in global models for climate forcing.

SO_2 Oxidation in MBL Stratiform Clouds

Increasingly sophisticated modeling studies of MBL stratiform cloud droplet chemistry (e.g., Ayers and Larson, 1990; Hegg *et al.*, 1992; Lowe *et al.*, 1997a) suggest the S_{IV} + ozone pathway dominates over the $S_{IV} + H_2O_2$ pathway in overall nss-sulfate production in this abundant and climatically significant cloud type. In Ayers and Larson's (1990) Lagrangian parcel model of wintertime clouds in southern mid-latitudes (i.e., Cape Grim, Australia), calculated pH values for sea salt-based droplets were initially between 5.5 and 6.0 and remained nearly constant throughout the model run (lifting through a 200 m thick cloud). At the same time, except very near cloud base, most liquid cloud water was associated with droplets growing on the smallest activated sulfate nuclei. Despite being initially very acidic, these nuclei contained so little mass that they soon diluted to pH values well above 5. Predicted bulk cloud water pH values and ammonium ion concentrations agreed well with limited measured values in samples collected from aircraft near Cape Grim (measured pH 5.4 to 5.8; [NH$_4^+$] 6.8 to 8.5 µM). Hegg *et al.* (1992) improved the Ayers and Larson model with a more detailed treatment of chemical activities in the high ionic strength droplets. Their results, also for Cape Grim conditions, suggested that as much as two thirds of the in-cloud sulfate production can take place in droplets nucleated on sea salt even though they constituted <6% of droplet number and <25% of liquid water mass. Lowe et al. (1997a) constructed a model with further-improved treatment of chemical activities and applied it to conditions representative of continentally influenced MBL regions (i.e., initial [SO$_2$] = 0.5 to 1.5 ppbv; initial [NH$_3$] = 1 ppbv). They also concluded that the S_{IV} + ozone pathway was dominant and most nss-sulfate was produced in sea salt based droplets.

We note that none of these modeling efforts appears to have included the possible acidification of cloud
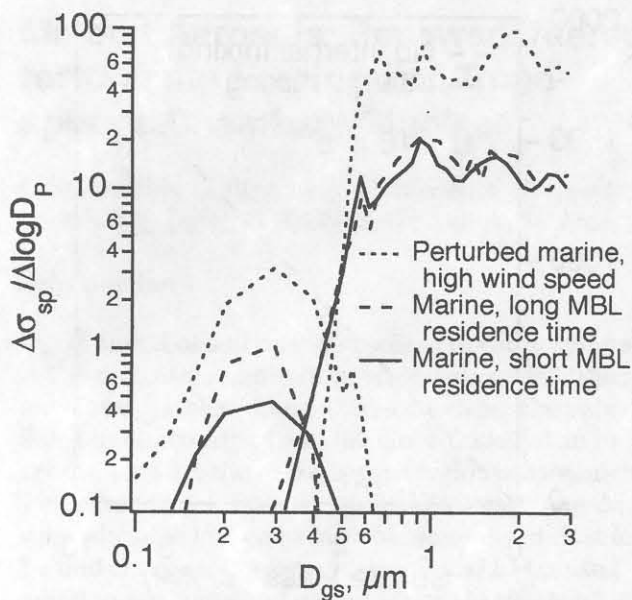
Figure 3. Mie-calculated scattering size distributions as functions of geometric surface mean particle diameter for the accumulation (nss-sulfate) and coarse (sea salt aerosol) modes at instrumental relative humidity (30 to 45%) and 550 nm. Examples are shown for three air mass types sampled during ACE-1. After Quinn et al., 1997.

droplets or the nuclei on which they are formed by nitric acid, methanesulfonic acid (another DMS oxidation product), or formic and acetic acids, all of which are ubiquitous in the MBL. To our knowledge, MBL cloud water has never been analyzed for all of these species or, more precisely, for their respective conjugate bases nitrate, methanesulfonate, and formate and acetate. Thus, their contribution to cloud water acidity can not currently be assessed with much certainty. Although available data are very limited, measurements reported by Munger et al. (1989) show similar concentrations of H^+ in different size fractions of marine stratus cloud droplets despite large size-dependent variability in concentrations of other solutes. The ionic composition of precipitation and, by extension, of cloud water suggests that HNO_3 and carboxylic acids together account for about half of the volume-weighted acidity in remote regions (e.g., Moody et al., 1991) and about a third in polluted regions (Galloway et al., 1989). Because their thermodynamic properties favor partitioning into droplets with higher pH, these other acids should diminish the importance of the S_{IV} + ozone pathway in marine clouds. The influence of other acids on nss-sulfate production in MBL clouds clearly warrants further investigation.

Sea Salt Aerosols and Direct Radiative Forcing: Some Results from ACE-1

The amount of incoming solar radiation that is scat-

tered by aerosols depends strongly on the size distribution of aerosols and on their index of refraction, density, and hygroscopicity. The latter properties are functions of the chemical composition of the aerosol. Especially important is the accumulation mode[2] because aerosols in this size range are most efficient at scattering radiation at visible and near-UV wavelengths. The conventional view is that sulfate aerosols are responsible for most scattering (e.g., Charlson et al., 1991), at least in the Northern Hemisphere where industrial SO_2 emissions now overwhelm natural S sources. However, data from IGAC's *Tropospheric Aerosol Radiative Forcing Observational Experiment* (TARFOX) campaign suggest that carbonaceous aerosols may also be very important in at least some northern regions (see accompanying article by P.V. Hobbs). And other new results from IGAC's *Southern Hemisphere Aerosol Characterization Experiment* (ACE-1) raise the possibility that sea salt aerosols may dominate scattering in oceanic regions that are minimally affected by industrial emissions.

ACE-1 took place in November and December of 1995 in the Southern Ocean region south of Australia. Measurements made aboard a research ship allowed for the determination of the size distribution, composition, and scattering properties of aerosols in the accumulation mode and the larger "coarse" aerosol mode (up to about 10 µm). The relative importance of nss-sulfate and sea salt in contributing to scattering is illustrated in Figure 3 for three types of air masses encountered during ACE-1: Perturbed (by emissions from Australia) marine with high wind speeds, marine with long MBL residence time, and marine with short MBL residence time. Throughout the month-long campaign, the sea salt mass concentration in the accumulation mode was much larger than the mass concentration of nss-sulfate. Hence, these shipboard measurements indicate that sea salt may be the dominant scatterer within the well-mixed, remote MBL not only for the super-µm aerosol but also for the sub-µm aerosol (Quinn et al. 1997). If the only source of nss-sulfate is from the ocean surface, then this MBL picture should be relevant for the entire atmospheric column. If, however, there is a layer of nss-sulfate aerosol above the MBL due to long range transport from a continental region, then nss-sulfate may make a more significant contribution to total column direct radiative forcing.

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[2] Aerosol size distributions frequently show prominent maxima in the 0.2-2 µm size range which are believed due to condensation of material from the gas phase or coagulation of very small particles. Aerosols of this size have long residence times (a week or more) and are therefore referred to as the "accumulation mode" (Wallace and Hobbs, 1977).

The Role of Sea Salt in Cloud Activation and Indirect Radiative Forcing

One of the shortcomings in attempting to quantify indirect radiative forcing, particularly by sulfate aerosols, has been the neglect of other sources of cloud condensation nuclei (CCN), particularly sea salt nuclei over the oceans. Measurements of sub-μm aerosol physico-chemical characteristics have shown that in clean marine air masses and at moderate-to-high wind speeds, sea salt aerosol can contribute a significant fraction of the accumulation mode marine aerosol number concentration and sometimes even dominate this mode with concentrations of the order of 70-100 cm^{-3} (e.g., Murphy et al., 1997; O'Dowd et al., 1997a; Quinn et al., 1997). The accumulation mode is thought to comprise the majority of CCN typically encountered in marine stratiform clouds. Consequently, the presence of sea salt in this size range suggests an important role for sea salt acting as CCN and is corroborated through measurement of cloud droplet residual particle analysis (Noone et al., 1997). Furthermore, since sea salt is more easily activated than nss-sulfate aerosol of the same size, and because sea salt tends to be distributed to slightly larger sizes than nss-sulfate, these nuclei are more likely to be preferentially activated than nss-sulfate nuclei.

Measurements and modeling studies of the impact of supplementing the nss-sulfate CCN population with

Figure 5. Comparison of predicted sulfate aerosol number concentration as a function of sulfate mass for internal mixing and no internal mixing scenarios.
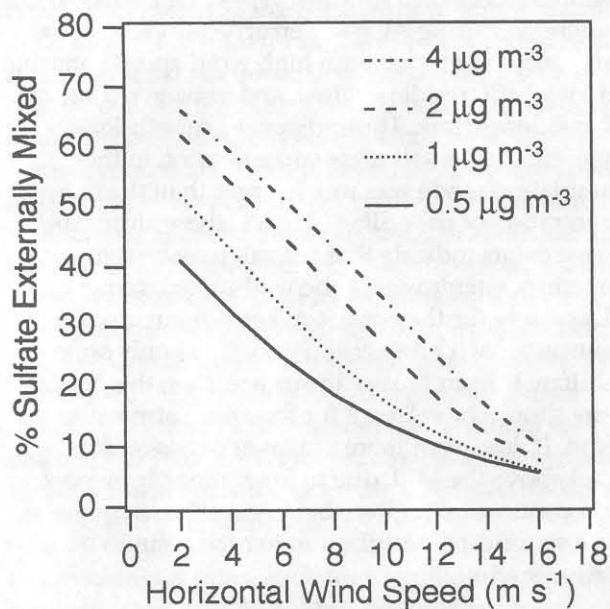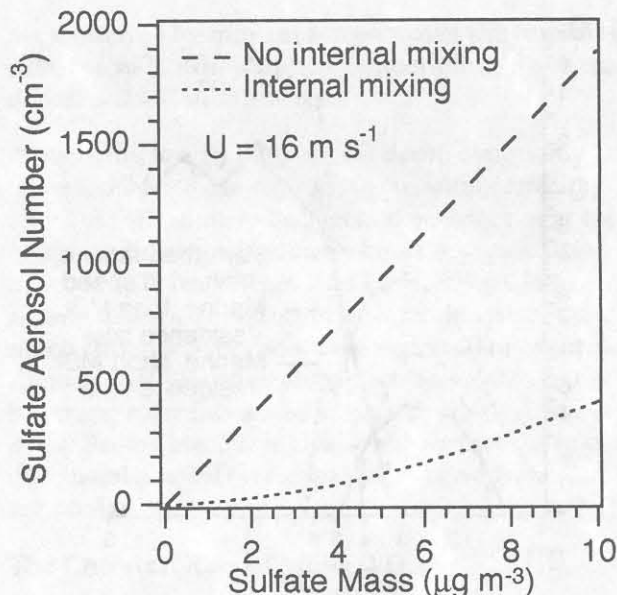

Figure 4. Percentage of nss-sulfate mass which is externally mixed with sea salt as a function of wind speed (and, consequently, sea salt concentration). This percentage of externally mixed nss-sulfate mass is that which is available for sulfate CCN production. Curves are shown for model runs with nss-sulfate concentrations of 0.5, 1, 2, and 4 $\mu g \, m^{-3}$.

sea salt CCN (O'Dowd et al., 1997a, b) illustrate that the effect on cloud droplet concentration is not only to reduce the relative contribution of sulfate aerosols to activated CCN, but also to reduce the droplet concentration, even though the concentration of so-called "available" CCN is enhanced. This unexpected effect arises from the fact that the sea salt nuclei are preferentially activated first into cloud droplets and thus reduce the peak supersaturation achieved in cloud. This leads to a reduction in the contribution of nss-sulfate to cloud droplet formation since the minimum size of the actual CCN is increased as supersaturation is reduced. The net microphysical effect of the presence of sea salt is to reduce the importance or contribution of nss-sulfate aerosol to the marine CCN population and, more importantly, reduce the actual cloud droplet concentration, and consequently, albedo.

Along with this microphysical interaction between sea salt, sulfate, and cloud droplets, the chemical coupling of the marine sulfur and sea salt cycles must also be accounted for in climate and radiative forcing models. As discussed previously, a large fraction of nss-sulfate is internally mixed with super-μm sea salt aerosol. Furthermore, measurements and modeling studies estimate that the fraction of nss-sulfate internally mixed with sea salt is as high as 70-90% when the sub-μm aerosol partitioning is also included (O'Dowd et al., 1997c), and, thus, not present as new sulfate CCN.

Most estimates of climate forcing by anthropogenic sulfate and, indeed, natural sulfate have assumed that all CCN are nss-sulfate and that the majority of nss-sulfate aerosols are available for the production of new CCN. In view of the processes highlighted above, this

implies that the role of sulfate in indirect climate forcing has almost certainly been overestimated. Recent inclusion of a multi-component aerosol droplet parameterization encapsulating the interaction of nss-sulfate and sea salt with cloud nucleation and internal/external sulfate partitioning with sea salt aerosol suggests that the role of sea salt is considerably more important in the climate system than previously thought (Lowe et al., 1997b).

By way of example, Figure 4 illustrates the effect of wind speed, a surrogate for sea salt concentration and readily available in GCMs, on the concentration of externally mixed nss-sulfate aerosol. The effect of increasing the wind speed and, consequently, the sea salt concentration, is to significantly reduce the fraction of sulfate mass available for new CCN production.

Illustrated in Figure 5 is a plot of nss-sulfate aerosol number concentration as a function of nss-sulfate mass for cases with no internal mixing with sea salt aerosol and with sea salt aerosol at 16 m s⁻¹ wind speed. The relationship is linear for the case with no internal mixing, but it is nonlinear when internal mixing is considered. The effect of internal mixing is to greatly reduce the number concentration of nss-sulfate aerosol available as CCN. Also shown (Figure 6) is the cloud droplet concentration for cases with and without sea salt. For the case with sea salt, the droplet concentration is higher for low nss-sulfate aerosol concentrations due to the presence of sea salt nuclei, but, for high nss-sulfate concentrations, the
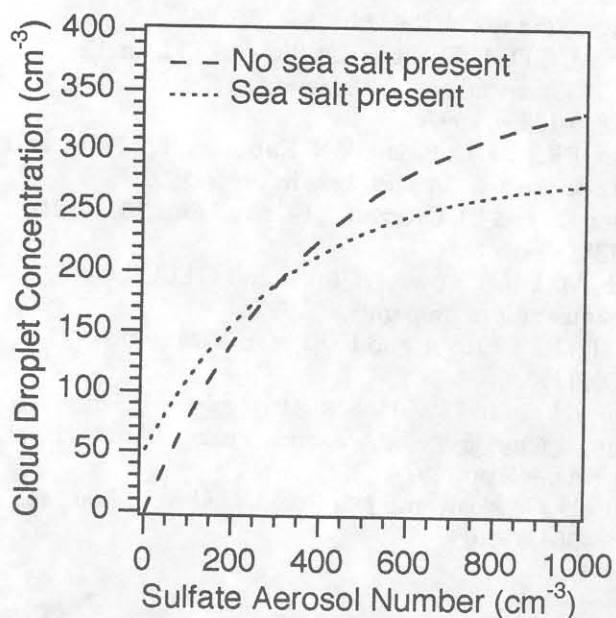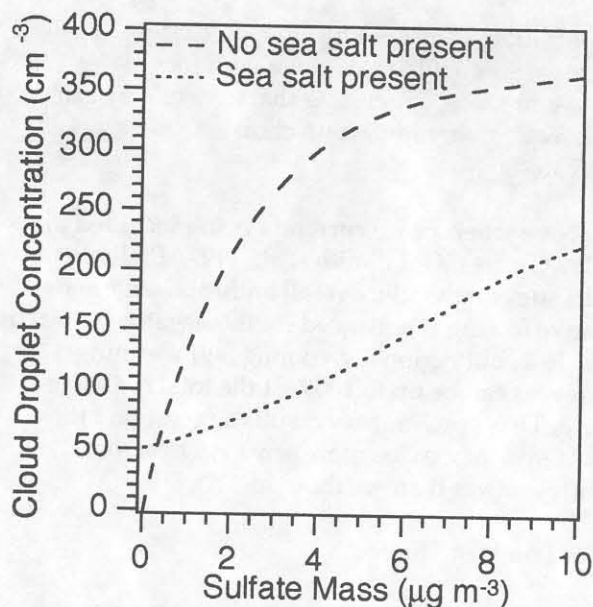

Figure 7. Predicted cloud droplet number concentration as a function of sulfate mass concentration for scenarios with and without sea salt.

droplet number is reduced compared to the nss-sulfate only case. Finally, Figure 7 illustrates the effect of including internal mixing and sea salt CCN on cloud droplet concentration as a function of nss-sulfate mass. It is clearly evident that when internal mixing with sea salt aerosol is included in this parameterization, the absolute number of cloud droplets predicted for a given total sulfate mass is greatly reduced.

In terms of climate forcing, we can imagine the increase in nss-sulfate mass in Figure 7 as resulting from increased anthropogenic S emissions with time. Say, for example, the pre-industrial era had a natural background nss-sulfate mass concentration of 4 μg m⁻³ and the post industrial revolution (current day) mass loading is 8 μg m⁻³. The scenario whereby all nss-sulfate contributes to new CCN and sea salt does not make a contribution to the CCN population, similar to the scheme used by Jones et al. (1994) to estimate the anthropogenic sulfate indirect radiative forcing, is compared to the scenario which includes both sea salt CCN and internal mixing in sea salt aerosol. The scheme which includes sea salt indicates that the current anthropogenic sulfate forcing would be greater than that estimated using a scheme similar to Jones et al. since the difference between the cloud droplet concentration for the 4 μg m⁻³ and 8 μg m⁻³ cases is greater. Also, the inclusion of sea salt tells us that the absolute nss-sulfate radiative forcing is less than currently thought (as per Jones et al.). An even more important point is that the scheme including sea salt suggests that further anthropogenic S emissions will have a greater effect on cloud droplet concentration as the rate of change in droplet concentration as a function of sulfate mass is greater for more polluted sulfate

Figure 6. Cloud droplet number concentration as a function of sulfate aerosol number concentration for scenarios with and without sea salt CCN

concentrations. The scheme which includes sea salt is thus more susceptible to change with increasing sulfate emissions, suggesting that further emissions will have a greater impact on climate than originally anticipated.

The above schemes are currently being included in the Hadley Centre GCM (Smith *et al.*, 1997). Preliminary results suggest that the overall anthropogenic aerosol radiative forcing is enhanced (in the negative sense) by about 10%, but regionally, cooling and warming differences can be up to 100% of the total radiative forcing. These preliminary results suggest that the climate system may be more sensitive to aerosol radiative effects than we thought.

A Word on Wind Speed

We close by reemphasizing that sea salt mass is an exponential function of wind speed. As such, climatic change in global wind fields would have a disproportionately large influence on sea salt concentrations and fluxes in the MBL with associated impacts on S cycling and cloud microphysics. Based on what we have summarized above, it would seem prudent to incorporate effects associated with sea salt into global climate models.

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Mineral Aerosols: Renewed Interest for Climate Forcing and Tropospheric Chemistry Studies

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Introduction

Mineral or soil dust aerosols in the atmosphere originate mainly from wind erosion in tropical and subtropical arid and semi-arid areas. The Sahara-Sahel in North Africa and the Gobi-Taklamakan in central Asia are the major source regions (Bergametti, 1992; Prospero, 1996). The spectacular and sometimes unpredictable lofting of massive amounts of dust (e.g., behind sharp cold fronts crossing Asia) has caused aviation accidents and other local-scale effects which have been the subject of case studies (e.g. Pitty, 1968; Prospero *et al.*, 1970; Carlson and Prospero, 1972; Prodi and Fea, 1979). Dust is now recognized as a major component of the tropospheric aerosol assemblage. The global annual input of mineral aerosol to the atmosphere is estimated to be of the order of 1 to 2 billion metric tons per year, representing about half of the total production of tropospheric aerosols by both natural and anthropogenic sources (IPCC, 1994; Duce, 1995).

Although mineral dust is composed of relatively large particles with a mass-median diameter of 1.5-3 µm, it can be transported over very long distances. For instance, dust raised over the central Asian deserts has been observed over the North Pacific Ocean as far as 5000 km from the source region, and dust from the Sahara is regularly observed on the western side of the tropical Atlantic (e.g., Uematsu *et al.*, 1983; Prospero, 1996). This is because, unlike other tropospheric aerosol species, dust is raised rapidly by high wind events to altitudes of several kilometers above source regions and is mainly transported above the atmospheric boundary layer (e.g., Prospero and Carlson, 1972; Iwasaka *et al.*, 1988; Swap *et al.*, 1992). Spectacular lidar observations from the NASA space shuttle LITE experiment in September 1994 confirm this pattern.

Mineral dust aerosols are drawn earthward by gravity along their transport paths and are deposited to lands and oceans where they provide iron and other micronutrients to terrestrial and marine ecosystems. Sedimentation of windblown dust over millions of years is indeed responsible for the continental loess formations, one of the most fertile soils on Earth (Pye, 1987), and Saharan dust is the most important parent for quaternary soils on Caribbean and western Atlantic islands (Muhs *et al.*, 1990). It also greatly contributes to the deep-sea sediments of the western tropical Atlantic (Delany *et al.*, 1967; Rea, 1994). Saharan dust deposition provides essential nutrients to soils in the Amazon basin (Swap et al., 1992), and influences the trace metal chemistry of the remote ocean (Buat-Ménard and Chesselet, 1979). In some high nutrient-low chlorophyll remote oceanic areas, mineral aerosol appears to be the major and limiting source of iron and other elements which could control the growth of phytoplankton (Martin *et al.*, 1994; Jickells, 1995). Because the mineral dust cycle is strongly correlated with climate variations (Jouzel *et al.*, 1993) and with the hydrological cycle (Yung *et al.*, 1996; Moulin *et al.*, 1997a), records of deposited mineral dust in deep-sea sediments and in polar ice are also a useful tool for reconstructing past meteorological and climatological patterns (De Angelis *et al.*, 1987; Rea, 1994).

Mineral dust also has noticeable impacts during its atmospheric transport before being deposited. For instance, images from early meteorological satellites (e.g., Prospero *et al.*, 1970; Shenk and Curran, 1974) demonstrated the impressive direct radiative effect of dust in the solar spectrum and thermal infrared. Neutralization of rain acidity by dust is also well known (Löye-Pilot *et al.*, 1986). Recently, new considerations have emerged in the frame of global change because of the effect of dust on the radiative budget of the Earth (e.g., Tegen and Lacis, 1996) and on the way by which it could affect the photochemical oxidant cycle (Dentener *et al.*, 1996). Moreover, anthropogenic perturbations are thought to have caused a significant increase in dust emissions (Tegen *et al.*, 1996).

The Radiative Role of Mineral Dust

It is now believed that mineral dust affects the radiative budget of the Earth mainly by absorbing and scattering solar and terrestrial radiation (direct effect). First estimates of its role were in terms of sources of cloud condensation nuclei, which suggested no (or limited) indirect radiative effect on a global scale.

Sign and Magnitude

It is generally stated that the net radiative effect of dust corresponds to a cooling effect at the surface and a warming at the altitude of the dust layers (Carlson and Benjamin, 1980). But the sign of the effect of the mineral aerosol on the radiative budget at the surface and at top-of-atmosphere depends on highly variable dust parameters, primarily the concentration, mineralogical composition (through the refractive index), size distribution and vertical distribution of the particles, and on external parameters such as the surface albedo and temperature (Tegen and Lacis, 1996; Sokolik and Toon, 1996). The dust is a variable mixture of clay minerals, quartz, carbonates, oxides, etc., having different scattering and absorption characteristics. Its absorption capacities at both solar and terrestrial

wavelengths make it peculiar regarding its interaction with the Earth's radiative budget. Moreover, the size distribution of the transported dust particles is gener-. ally quite broad (geometric standard deviation usually of the order of 1.5 to 3), but closer to the source-regions the size distribution can be quite different due to an enrichment in larger particles (up to 50 μm). Thus, the size-range covered by the mineral aerosol is suffi-ciently large to allow it to scatter both the incoming visible radiation and the outgoing terrestrial radiation in the thermal infrared spectral region.

In terms of magnitude, Andreae (1995) estimated the relative importance on climate of the various aerosol types. This estimation was based on a mean atmo-spheric burden (deduced from production rates and mean residence times) and relevant mass extinction coefficients. Although mineral aerosol is much less efficient than sulfates in scattering solar radiation, its atmospheric concentrations are generally significantly larger over and near source regions The result is that mineral aerosol is a main natural contributor to the aerosol optical depth with a global mean value of 0.023. The corresponding direct radiative effect would be of order -0.75 W m^{-2}, close to that of anthropogenic sulfates (-1.1 W m^{-2}).

Regional Climate Forcing by Mineral Aerosols

Such an approach provides only order-of-magnitude estimates of the relative importance of various aerosol types on climate forcing because of required assump-tions about the behavior of the different types. Aero-sols have short residence times in the troposphere (hours to about a month at most) which limits mixing of the various aerosol types. Spatial variability of aerosol composition is reinforced by the geographical differences of respective source-regions (arid and semi-arid regions for mineral dust, mid-latitudes of the Northern Hemisphere for anthropogenic sulfates, oceans for sea salt, etc.). Finally, emissions of most natural aerosols are very sporadic, adding further temporal variability to the spatial heterogeneity. Thus, to improve the precision of estimation of aerosol radiative forcing, higher spatial (i.e., regional scale) and temporal (i.e., with respect to the residence times of the aerosol) resolutions are needed in terms of both measurements and modeling.

For mineral aerosols, such high spatial and temporal resolution can be achieved with satellites. As an example, Husar et al. (1997), using NOAA/AVHRR satellite images, showed that the marine regions where larger optical depths are observed are located down-wind of the main deserts of the Earth. This suggests that mineral dust exerts a strong direct radiative effect in the tropical and subtropical belt. Ground based measurements at Barbados by Li et al. (1996) con-firmed these satellite observations, showing that the

net scattering by mineral aerosols over the tropical and subtropical North Atlantic was about four times that of non-sea salt sulfate aerosols.

Accounting for the high optical depth commonly observed over these regions, the resulting effect in terms of the radiative budget at the surface or at top-of-atmosphere can frequently be a decrease of the order of 0.1 W m^{-2} (Tegen and Lacis, 1996; Claquin et al., submitted). The effect of dust storms is obviously much stronger over the source regions. For example, Zhou et al. (1994) observed a decrease of 40% of the incoming solar flux at the surface during a dust event in the Beijing area. This clearly shows that the impact of mineral aerosol on climate must be addressed at the regional scale.

The Chemical Role of Mineral Dust

Mineral dust aerosols have until recently been re-garded mainly as non-reactive, hydrophobic chemical substrates. Only their alkalinity was considered for its neutralizing effect on rainwater acidity (Löye-Pilot et al., 1986). This alkalinity is mainly controlled by the calcium carbonate content of soils of the arid and semi-arid regions and operates via the reaction:

$$CaCO_3 + 2 H^+ \rightarrow Ca^{2+} + CO_2 + H_2O$$
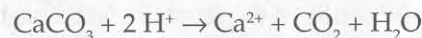
In marine regions which are dominant pathways of dust transport this leads to a shift in rainwater pH from 4-5 to 6-8 (Losno et al., 1991; Avila et al., 1997). In the western United States, it has been shown that soil-derived particles are frequently abundant enough to neutralize the acidic aerosol completely (Gillette et al., 1992), demonstrating the important role of mineral dust in the acidity budget of atmospheric deposition.

Recently, Dentener et al. (1996) used a global, three-dimensional model which coupled mineral dust processes with photochemistry and the nitrogen and sulfur cycles in order to evaluate the role of the mineral aerosol particles on tropospheric chemistry. Although their results suffer from large uncertainties in heterogeneous reaction rates, they suggest that interactions of N_2O_5, O_3, and HO_2 radicals with dust cause ozone to decrease by up to 10% in and near dust source areas. Their simulations also suggest that more than 40% of HNO_3 could be neutralized on dust over large parts of the globe, with obvious consequences on the acidity of atmospheric dry and wet deposition to ecosystems.

An exciting result of Dentener et al. (1996) concerns the interactions between dust and the sulfur cycle. Build-ing on a classical description of the atmospheric sulfur cycle, they used their model to evaluate the impor-tance of the uptake of SO_2 on mineral aerosols (which is considered dependent on the alkalinity of dust and

on the atmospheric water content). Despite uncertainties on the rate of the uptake of SO_2 on the dust particles, the simulations agreed well with existing observations, suggesting that the order of magnitude of the phenomenon is correctly estimated. The results suggest that over Asia, the Western United States, Australia, and North Africa, an annual average of 40% of the total sulfate is associated with mineral dust. Moreover, the regions for which more than 10% of the sulfate is associated with mineral aerosol extend over major parts of the world ocean. This has important implications especially for the role of increasing sulfate in climate forcing. Indeed, the size range of mineral particles is 2 or 3 μm while sulfates produced from gas phase reactions are sub-μm in size. Thus, having a larger fraction of sulfates associated with the larger particles may diminish the local cooling effect of the sulfate aerosol. Moreover, the sulfate present on the mineral dust particles will change the radiative properties of dust and also will enhance the ability of dust to act as cloud condensation nuclei (CCN). Indeed, it has recently been shown that mineral dust particles over the eastern Mediterranean are often coated with sulfates following cloud processing of aerosols. This converts them into effective giant CCN, and seems to affect precipitation development and ice crystal concentrations in convective clouds (Levin *et al.*, 1996). Thus, as mentioned by Dentener *et al.*, (1996), the present estimates of negative radiative forcing which ignore mineral aerosols may be too large.

Dust: A Human-Induced or Natural Climate Forcing?

In terms of climate forcing, mineral aerosols are generally regarded as a natural constituent and thus are not included among the anthropogenic forcing factors that may affect climate in the future. Considering dust emission processes, however, such a statement may be misleading. The strength of mineral dust emissions is highly sensitive to small changes in climatic parameters. For example, dust emission strongly depends on surface wind speed and only occurs when the wind speed exceeds a threshold value (Gillette *et al.*, 1980). This threshold (or friction) velocity is quite variable and depends on surface characteristics (roughness, size of soil particle aggregates, etc.; Marticorena *et al.*, 1997a). A limited change in the wind speed distribution could drastically modify dust emission strength and location. Dust emission strength is a function of the third power of the wind friction velocity indicating that small changes in wind speed will be strongly amplified in terms of dust emissions.

Precipitation is also a major parameter controlling the dust cycle. Most mineral dust is emitted in arid and semi-regions mainly due to sparse vegetation cover that is insufficient to protect the soil surface against wind erosion. Thus, changing rainfall in the subtropical belt has important consequences on the vegetation cover of the desert margins and significantly modulates the extent of the potential source regions of dust. For example, N'Tchayi *et al.* (1994) showed that the number of dusty days in Gao (Mali) increased from about 20 to more than 250 during the Sahelian drought. During the same period, Prospero and Nees (1977) recorded an increase by a factor 5 of the mineral dust load over the equatorial North Atlantic Ocean, which underscores the large-scale impact of such changes in the source regions.

The dependence of dust emissions on climatic parameters such as wind speed and rainfall strongly suggests that the atmospheric dust load should be significantly affected by any climatic change that may result from human activities. In fact, human activities have probably affected dust emissions already, especially by changing land use in some sensitive ecosystems. For example, in the United States, the highest dust production occurs in regions where the soil was disrupted by cultivation and other intensive land uses. However, it is likely that desert margins, especially the Sahel, are where human activities will have the most significant consequences in the future. Population has increased rapidly in these regions during the last twenty years, inducing or intensifying practices such as deforestation, cultivation, and overgrazing, which temporarily or permanently expose soils to wind erosion. Tegen *et al.* (1996), by means of an inverse modeling exercise, estimated that about half of the current atmospheric dust loading could be due to the disruption of soils by changing use of land in semi-arid regions such as the Sahel. In terms of radiative effect, they concluded that these "anthropogenic" soil-derived particles "cause a decrease of the net surface radiative forcing of about 1 W m^{-2}, accompanied by increased atmospheric heating that may be a significant forcing of atmospheric dynamics". An increasing trend in dust export from North Africa has been documented (Prospero, 1996; Avila *et al.*, 1997). However, it has also been found to correlate with a similar trend in the North Atlantic Oscillation, which controls the transport of humidity from the tropical Atlantic towards Europe and North Africa (Moulin et al., 1997a; see Figure 1, page 16). Therefore it is not clear to what extent the increase in dust export from Africa directly results from increasing desertification in the Sahel due to anthropic pressure, or because of more general climate change constraints (see Figure 2, page 16). Anyway, the fraction of the mineral dust particles that results from both man-made climatic change and intensive land-use should be considered as an anthropogenic climate forcing factor.
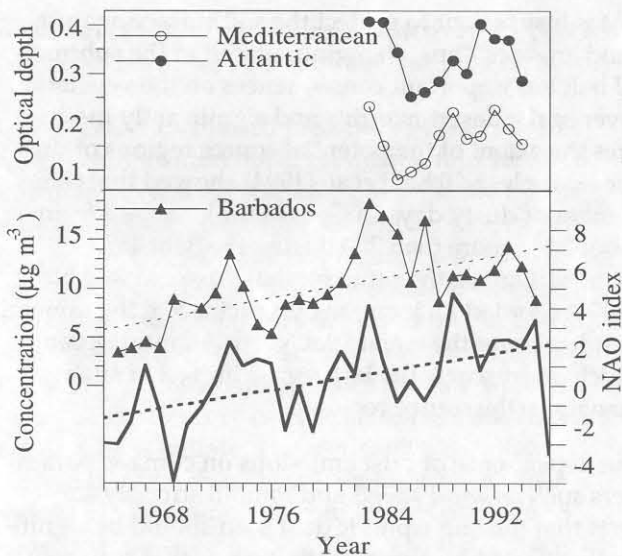
Figure 1: Comparison of the North Atlantic Oscillation (NAO) index (bottom, bold continuous line, 1964-1996 period; data from J.W. Hurrel) with the annual mean mineral dust surface concentrations at Barbados (1965-1995 period; data from J. M. Prospero) and Meteosat-derived summer mean dust optical depth over the eastern Atlantic and the Mediterranean (top, 1983-1994 period; data from C. Moulin). Dotted lines show trends from linear regressions. Correlations between NAO and all dust indices are statistically significant (Moulin et al., 1997a).

Concluding Remarks

As for other aerosol species, long-term monitoring of atmospheric dust concentrations has been very limited. Only at one ground-based station (Barbados) have more than 30 years of continuous dust measurements been performed (Prospero, 1996). Only one or two decades of dust aerosol observations from satellites are available from different meteorological satellite data sets (Husar *et al.*, 1997; Herman *et al.*, 1997; Moulin *et al.*, 1997b). New satellite sensors such as SEAWIFS, POLDER, MODIS, and future spaceborne lidars will increase our capabilities for dust monitoring. Extensive use of such space observations already allows powerful tests of dust emission and transport models (Marticorena *et al.*, 1997a; Schulz *et al.*, in press). A full and precise assessment of the effects of mineral dust particles on climate will require global models that can account for the interactive processes between dust emissions and climate (Andreae, 1996). This will be only possible with models that are able to link dust emissions explicitly with relevant meteorological and surface parameters at pertinent temporal and spatial scales. Such models have recently been designed (Marticorena and Bergametti, 1995; Shao *et al.*, 1996) and applied with success to desert regions for which the surface characteristics had been mapped (Marticorena *et al.*, 1997b). An operational regional model for weather and dust transport forecasting in the Mediterranean region is now available (Söderman and Dulac, 1997; http://pentium.ccsem.infn.it).

These new tools will be operational for global scale modeling in the near future allowing new advances in our understanding of the role of dust on climate.

Recent advances require that a new view be taken of atmospheric mineral dust and its interactions with Earth's climate and atmospheric chemistry. Dust is the largest component of the tropospheric aerosol on a mass basis. The strength of dust emission may be subject to drastic changes in the future due to anticipated climatic changes and land use intensification, making these particles a key element of the human-induced climate forcing. In terms of chemistry, the mineral aerosol provides an efficient substrate for acid-base reactions leading to neutralization of atmospheric deposition, generating new sinks for key species in the nitrogen and sulfur cycles and modifying the hygroscopic character and optical properties of both sulfates and the dust itself.

Mineral dust can not be neglected in global change studies. Understanding of the role of mineral dust on environmental and climate forcing remains incomplete and will require intensive studies taking into account the interactions between the different aerosol species, atmospheric water and oxidants, soil features, radiation and climate.
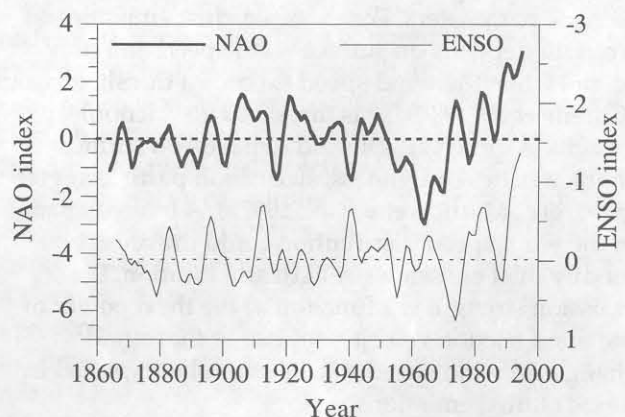
Figure 2. Trends in the NAO (top; Hurrell, 1995) and ENSO (bottom; Jones et al., 1996) indices (5-yr weighted averages) since the mid-18th century, suggesting a possible climate change since the last decades, compared to relative stability during the preceding one hundred years.

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ACE-2 Field Phase Completed

Contributed by *F. Raes*, *CEC Joint Research Centre, Italy*
and *T.S. Bates*, *NOAA Pacific Marine Environmental
Laboratory, USA*

The IGAC aerosol research community success-
fully completed the main field phase of its *North
Atlantic Regional Aerosol Characterization Experiment*
(ACE-2) on 25 July 1997. The intensive campaign ran
for 6 weeks to study the radiative effects and control-
ling processes of anthropogenic aerosols from Europe
and desert dust from Africa as they are transported
over the North Atlantic Ocean. The experiment
involved over 250 scientists from Europe and the
United States. It included 60 coordinated aircraft
missions with 6 aircraft (for a total of 450 flight
hours), one ship, and ground stations on Tenerife,
Madeira, and mainland Portugal. Operations were
directed from Tenerife.

At the present stage of data analysis, ACE-2 can only
be seen as 6 different activities that were operating at
the same time, in the same place and making use of
the same operational logistics. This is already an
achievement. It is, however, the integration of results
from the various platforms and activities, and the
comparison of ACE-2 with ACE-1 data, that will yield
the added value that these large scale experiments are
designed to provide. The added value will come from
testing the consistency of our thinking about and
discovering new links between the multitude of
aspects that are related to the problem of aerosols and
climate.

Preliminary highlights from each of the ACE-2
activities are given below. More information about
them can be found on the ACE-2 homepage: *(http://
rea.ei.jrc.it/~vandinge/ace2/ace2main.html)*.

- A wide range of aerosol types was encountered
  throughout the ACE-2 area, including background
  Atlantic marine, European pollution-derived, and
  (although less frequently than expected) African
  mineral dust. Local closure studies were per-

formed upwind of the ACE-2 area (Portugal and
ship) and downwind (Tenerife). These experi-
ments involved simultaneous characterization of
the chemical, physical, radiative, and cloud
nucleating properties of the aerosol.

- Pollution from Europe and dust from Africa were
  found to be transported in distinct layers. The
  optical depth of these layers ranged from 0.05 to
  0.36. On several occasions they were character-
  ized by simultaneous measurements from air-
  borne and surface-based platforms, using *in situ*
  and remote sensing techniques, in conjunction
  with satellite overpasses.

- It was found that aerosols originating over
  Europe and Africa and transported up to 3000 km
  from the source still have an impact on the
  chemistry and radiative properties of the atmo-
  sphere. Particularly noticeable effects were
  observed on the stratocumulus cloud deck over
  the NE Atlantic. Measurements from a mountain
  ridge on Tenerife showed that during clean
  periods, the cloud water chemistry was domi-
  nated by marine ionic species, the pH was typi-
  cally 5.5 and aerosol particles as small as 40 nm
  diameter were cloud-processed. During polluted
  periods many more droplets were activated and
  the cloud water was highly acidic, pH typically
  3.5, with a substantial contribution of nitric acid.
  The aircraft measurements showed that in pol-
  luted conditions only a fraction of the available
  (accumulation mode) aerosols activated and
  formed cloud droplets.

- The optical properties of clouds forming in clean
  and polluted air masses were studied by high-
  and low-flying aircraft, in conjunction with
  satellite overpasses. These highly coordinated
  experiments (up to 4 aircraft) allowed detection of
  the indirect aerosol effect. This means that varia-
  tions in cloud optical depth could be ascribed to
  variations in cloud droplet concentrations and
  deconvoluted from the variations in the geometri-
  cal depth of the cloud. Furthermore, the data will
  allow the number of cloud droplets to be related
  to the physical and chemical properties of the
  marine boundary layer aerosol on which they
  form.

- The Lagrangian experiments carried out during
  ACE-2 were particularly successful. In all three
  experiments balloons and PFC tracers were
  released from the ship and they confirmed very
  well the (hence excellent) predictions by the
  meteorological forecasting team.

- Scientifically, the three Lagrangians showed a

wide variety of aerosol evolution. The first Lagrangian was carried out in a clean, maritime air mass with only small changes in the aerosol spectra. The second and third Lagrangians took place in much dirtier air masses and these showed a surprizingly different evolution of the aerosol. In the second Lagrangian, relatively extensive and deep stratocumulus clouds were encountered and it appeared that significant cloud processing of the aerosol occurred. In the third Lagrangian the air mass was just as dirty and there were a few clouds (cumulus and stratocumulus) around, but there was absolutely no change in the boundary layer aerosol spectrum from the first flight to the last.

- Nucleation of new particles was frequently observed at the coastal measuring site in Portugal. New particles were also observed during upslope conditions at the free-tropospheric site on Tenerife, where they appeared to be organic. However nucleation was largely absent in the boundary layer over the open ocean, and the airborne measurements showed that it was also absent in the free troposphere up to 10 km above Tenerife. The airborne measurements did reveal layers of sulfuric acid (gas plus particulate) around 5 km. They also indicated that mineral
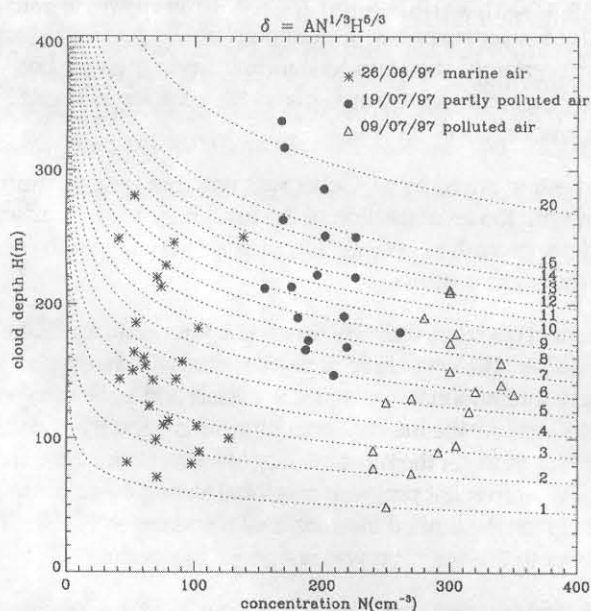

Fig 2. *Smart balloon track and flight pattern of the first three MRF C-130 flights during the first ACE-2 Lagrangian on 3-5 July 1997. A fourth flight was performed just upwind of the Canary Islands by the CIRPAS Pelican (not shown). (Data: Meteorological Research Flight, UK.)*

Fig 1. *Preliminary analysis of three CLOUDYCOLUMN flights, showing the range of cloud droplet concentrations and cloud optical and geometrical thicknesses encountered. Plotted are values of optical thickness $d = AN^{1/3}H^{5/3}$, where A is a constant, N is cloud droplet concentration, and H is cloud thickness. The geometrical thickness largely determines the optical thickness, but more detailed analysis shows that an effect of cloud droplet number can also be observed (Data: Meteo France.)*
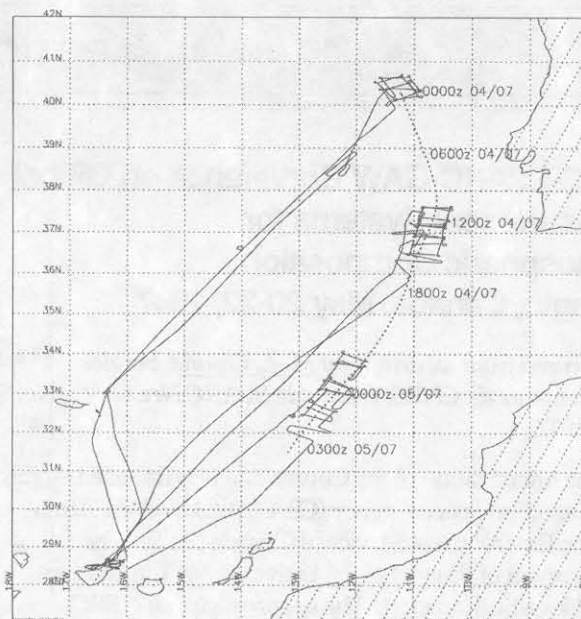
dust particles might have an effect on gas phase chemistry, most likely due to the scavenging of reactive species (radicals).

- At 5 sites across the ACE-2 area, aerosol chemical measurements have been running continuously since January 1996 and throughout the ACE-2 intensive period. These measurements will continue until the end of 1998, and will put the ACE-2 campaign in a larger temporal and geographical context.

- During the experiment, the meteorological forecasts, satellite data and *in situ* data were accessible on-line via the ACE-2 web page. This turned out to be a must for the coordination and daily decision making. About 30,000 hits of the web page were recorded during the 6 week period, of which 4,000 came from outside the ACE-2 Operations Center domain.

All ACE-2 data will be collected in the ACE-2 data archive at the Joint Research Centre (JRC), Ispra, Italy. To start the scientific integration process, a first ACE-2 data workshop is scheduled from 30 March to 3 April 1998 at the JRC.

Meeting Report

IGAC-SPARC-GAW Conference on Global Measurement Systems for Atmospheric Composition
Toronto, Canada, May 20-22, 1997

Condensed from an article by M.-L. Chanin, Service d'Aéronomie du CNRS, France, in SPARC Newsletter No. 9, July 1997

The organization of the Conference was financed by the Canadian Space Agency (CSA/ASC) and the Atmospheric Environment Service of Canada (AES). The Department of Physics of the University of Toronto provided logistical support. The sponsorships of NERC, COSPAR, ESA, and GAW are also to be acknowledged. The Committee Chair, Professor J.R. Drummond, did an excellent job in organizing the Conference and is warmly thanked here.

The arguments to organize this Conference were the following: The chemical composition of the atmosphere is changing on a global scale. This has far reaching implications for the health of the environment and the future of human society. This recognition leads many nations to participate in assessing the current state and trend of the chemical state of the atmosphere. This requires in turn the assembly of global measurement systems to describe the atmospheric composition.

Therefore the aim of the Conference was to bring together scientists, managers, and policy makers to discuss current knowledge of atmospheric composition and predictive capabilities for measuring it, to define the near-term requirements for global measurement systems and to begin developing a framework for more comprehensive systems in the future. This Conference was intended to stimulate interaction between those who plan and conduct large-scale measurement programs both as planners and experimenters, and modelers. The Conference format was well designed to facilitate discussion and exploration of synergistic opportunities.

The major outputs of the conference were summed up by M. Geller in the last paper entitled: "Where are we now, and where are we going?". He presented the main conclusions and raised some unanswered questions with are summarized here:

After the presentation of the current space instrumentation (TOMS, GOME UARS, LITE, ADEOS...) and of the efforts to assure that those multi-nation, multi-investigator space measurements provide valid well-calibrated results, one may wonder whether these efforts are sufficient and question to what extent crucial efforts of one nation (or agency) are dependent upon another one.

Valuable measurements utilizing different platforms were presented (ground-based, research aircraft, commercial aircraft, measurements on ships, even measurements on trains). Those are placed under the responsibility of many nations, industrial and developing, and one has to ensure that these will continue and be expanded in the future. The experience in meteorology shows that sometimes valuable proven technologies are deserted without a proven viable alternative.

In parallel with the acquisition of these measurements, new models are also being developed and new model/data procedures (i.e., data assimilation) are to be developed; such efforts should be encouraged.

Even though plans for the future look quite bright: (EOS/CHEM, ENVISAT, ODIN, space station, new aircraft, ground-based network...), gaps were identified, in particular related to troposheric O_3 network, trends in UT/LS water vapor, aerosols and their role in atmospheric chemistry. Programs should be developed on those topics.

Another question is to know if the community is prepared and ready for surprises: One should not be portraying too much certainty in our understanding, and we should be ready for setting up campaigns on the spot, for example if a volcanic eruption occurs.

A question raised by M. Geller was whether the community is putting too great fraction of the funding into big hardware and not enough in people. The answer may not be the same for different countries.

The final question was whether it is appropriate for IGAC, SPARC and GAW to hold at regular intervals meetings where international managers, scientists and policy makers would review the international programs, identify gaps, etc., in an effort to get the most intelligent international research and measurement program on global atmospheric composition, given the limited international resources available. The answer to this question was not given in real time.

It should be mentioned that the objective of this Conference fits within the mandate of GCOS (Global Climate Observing System) and a special meeting on GCOS in relationship with atmospheric chemistry was held the day following the end of this conference.

The participants have been invited to submit their presentation for a special issue of the *Journal of Atmospheric Science*. It is hoped that a large number of contributions given during the Conference will appear in this issue.

Announcement

IGAC Directory of Atmospheric-Biospheric Chemistry Data

A considerable wealth of data on the chemical composition of the atmosphere has been obtained in recent years. These data are scattered in many places and are often not readily accessible. For these reasons, IGAC has begun to compile a "Directory of Atmospheric-Biospheric Chemistry and Related Data". The focus is on chemical data relevant to global change issues, but data related to regional problems are also included.

This Directory is intended to complement the World Data Centers and other archives operated under WMO, NASA, and other institutions by providing a ready source of locator information for datasets. The Directory resides at the National Center for Atmospheric Research in the U.S. and is now accessible on the Internet via:

http://web.mit.edu/afs/athena.mit.edu/org/i/igac/www/datadirec.html

Through this web site it is possible to *contribute* to the Directory as well as use it.

For further information or assistance with the Directory contact: Dr. Zaichun Hu, Atmospheric Chemistry Division, National Center for Atmospheric Research, P.O. Box 3000, Boulder, CO 80307-3000, USA; Tel: (+1-303) 497-1874; Fax: (+1-303) 497-1400; Email: zchu@acd.ucar.edu.

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