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Group Report: Connections between Aerosol Properties and Forcing of Climate

S.E. SCHWARTZ, Rapporteur

F. ARNOLD, J.-P. BLANCHET, P.A. DURKEE, D.J. HOFMANN, W.A. HOPPEL, M.D. KING, A.A. LACIS, T. NAKAJIMA, J.A. OGREN, O.B. TOON, M. WENDISCH

INTRODUCTION

The mechanisms by which atmospheric aerosols influence radiative forcing of climate can be distinguished into three categories:

1. *Direct forcing.* Scattering and absorption of shortwave (solar) radiation by aerosols produces climate forcing by changing the planetary albedo. Absorption of longwave (terrestrial) radiation enhances the atmospheric greenhouse effect and causes surface warming. Absorption of solar or thermal radiation within the atmospheric column produces changes in the atmospheric temperature profile.
2. *Indirect (cloud) forcing.* An enhanced concentration of aerosol particles may result in an enhanced concentration of cloud drops in clouds that form in the presence of this aerosol. Under the assumption that other cloud properties such as liquid water content remain unchanged and provided that the aerosol is not significantly absorbing, the shortwave albedo of the cloud is increased. In the other direction, enhancement of concentrations of black carbon in clouds increases absorption of solar radiation in clouds. The net effect is expected to be enhancement of shortwave albedo of clouds but depends on black carbon concentrations and initial cloud albedo. Quantifying these phenomena requires

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examination of aerosol absorption, and also consideration of possible modification of other cloud properties beyond cloud droplet concentration that may be affected by anthropogenic aerosols and that can affect the resulting forcing: liquid water content, liquid water path, precipitation development, cloud lifetime, and water vapor loading after cloud evaporation.

3. *Indirect effects of aerosols on heterogeneous atmospheric chemistry.* Aerosol particles can serve as the site of atmospheric chemical reactions thereby increasing the rates of these reactions. To the extent that such reactions can influence the concentrations of climate-influencing constituents (such as greenhouse gases) by this process, anthropogenic aerosols can contribute to climate forcing by anthropogenic aerosols.

The working group addressed key issues associated with each of these forcing categories. Our discussions were organized about several key questions that were identified as requiring resolution in order to be able to estimate the forcings of each category. These questions are presented here together with summaries of the corresponding discussions.

DIRECT FORCING

Is the theory understood, or is the problem that needed data are lacking?
What are the processes that must be represented in climate models?
What are the quantities needed to do this?

The basic theory of how aerosols directly affect the radiation budget and hence the forcing of climate is understood. The eruption of Mt. Pinatubo in the Philippines in June, 1991, which provided a natural climate experiment of magnitude unprecedented in recent times, demonstrates this very clearly and also identifies the circumstances that made this climate experiment so successful. First, the perturbation was large, global, and isolated to the stratosphere, away from competing tropospheric aerosol signatures. Second, ground-based and satellite instrumentation was in place to make the measurements needed to characterize the time evolution of the volcanic aerosol, including its spatial distribution, chemical composition, and particle size and shape distribution (see Lacis and Mishchenko, this volume).

The composition of the volcanic aerosol was inferred to be concentrated sulfuric acid, for which accurate refractive index measurements are available over the full range of solar and thermal wavelengths (Palmer and Williams 1975). Since the particles were spherical and of known size, Mie theory could accurately provide radiative parameters—optical depth, δ; single-scattering albedo, ω_0; and phase function, $\beta(\phi)$—for calculating the radiative flux perturbation. Hansen et al. (1992) used preliminary estimates of this information in general circulation model (GCM) simulations of the radiative influence of the aerosol resulting from the Pinatubo eruption to predict a

global cooling by about – 0.5°C by the end of 1992, with a return to normal by about 1995.

Subsequent measurements of the global surface temperature have confirmed both the magnitude and the duration of the cooling resulting from the Pinatubo aerosol. Additional closure is provided by stratospheric heating at the volcanic aerosol level by about 2°C observed posteruption (Labitzke and McCormick 1992) that was due to absorption of upwelling thermal radiation by the Pinatubo aerosol. Atmospheric extinction measurement at thermal wavelengths (Pollack et al. 1991) can provide further verification of the particle size, composition, and optical depth through the spectral dependence of the aerosol extinction in the 8–12 μm region of the spectrum. The Pinatubo climate forcing was also observed in measurements by the Earth Radiation Budget Experiment (ERBE) satellite (Minnis et al. 1993; Kaufman, this volume).

In general, aerosols resulting from volcanic emissions (Pinatubo included) have been poorly documented in the initial period following the eruption. Partly, this is due to the remote location of most major volcanoes, the logistical preparations necessary to mount an observation campaign, and an understandable reluctance to fly aircraft through volcanic plumes. Available satellite measurements (SAGE) lack adequate time and spatial sampling, and become saturated by the large optical depth of injected material. TOMS measurements (e.g., Krueger 1983) provide useful maps of SO_2 column amounts, but systematic measurements of the ash component are lacking. Being comprised of relatively large particles, the ash component falls out during the course of a few weeks, and thus is a small contributor to global climate forcing. Nevertheless the large optical depths of ash particles during the initial period after the eruption can absorb significant amounts of both solar and thermal radiation, and thus produce large localized heating changes which may be important in the local stratospheric dynamics and the eventual dispersal of the volcanic aerosol. Future satellite instruments should include a capability to make more diagnostic measurements during initial phases of volcanic eruptions.

Tropospheric aerosols are far more complex than stratospheric aerosols in terms of their chemical composition, time and spatial distribution, range of particle size and shape, and overall global variability and dependence on human activities. Nevertheless, in principle, the overall radiative forcing due to tropospheric aerosols can also be accurately computed if the optical parameters of the individual chemical species are known and if appropriate mixing rules describing external as well as internal mixing are available. Such mixing rules, especially for the imaginary part of the refractive index, are crucial to describe scattering patterns of mixtures. The present lack of such mixing rules indicates a considerable gap of knowledge.

To evaluate the aerosol optical properties of tropospheric aerosols, it is necessary to identify the principal chemical species of these aerosols (e.g., sulfates, nitrates, silicates, sea salt, soot, organics, etc.) and to characterize the typical size distributions of these components and the variability of these distributions. The task of defining a reliable refractive index data base is made more difficult by the need to distinguish

external and internal mixtures of different chemical species. These tasks are clearly formidable and the necessary instruments may not be available. The high-frequency spatial and temporal variability of the tropospheric aerosol loading presents a further difficulty in modeling the tropospheric aerosol influence.

Unfortunately also, reliable laboratory measurements of refractive indices are lacking for most of the aerosol chemical species, and, since reliable particle size characterization of the aerosol species is also incomplete, it is not currently possible to calculate the radiative forcing by tropospheric aerosols from available data with demonstrable accuracy. Hence, additional measurements such as satellite-observed radiances and ground-based measurements of atmospheric transmission and sky radiance are needed to provide closure, i.e., to verify that optical depths and other radiative parameters retrieved from available measurements, when inserted in radiation models, are able to reproduce the observed radiances. Of particular importance is the determination of the single-scattering albedo of aerosols at visible wavelengths, which is difficult to measure directly and is sensitive to constituents such as soot and organic compounds, which are commonly present in aerosols.

Since changes in column-integrated radiances from which aerosol properties are inferred are small, improvement in measurement capabilities over current systems, such as greater utilization of multispectral polarimetric techniques, is needed to extract the required information. This applies to ground-based as well as satellite instrumentation.

To evaluate the forcing due to industrial aerosols, it is important to establish the preindustrial global background aerosol (relative to which anthropogenically induced changes in aerosol loading can be computed) with the same degree of reliability that the preindustrial greenhouse gas levels are known. Based on aerosol measurements at remote locations (e.g., Cape Grim, Tasmania) as well as the global mean estimates for the naturally occurring sulfate component and for windblown dust, it is thought that the global mean preindustrial aerosol optical depth may have been in the range of $\delta = 0.02$ to 0.05. Andreae (1995) derives an estimate of $\delta = 0.07$ for the global average natural aerosol background.

From a more physical point of view, the particle radius—and therefore ω_0 and $\beta(\phi)$—should be dependent on relative humidity for soluble aerosol species, such as sulfate. It will ultimately be necessary for climate models to allow the size distribution to evolve with time (Raes et al., this volume). However, this is currently not feasible in GCM applications; however, the error associated with not varying the size distribution is likely to be small compared to other errors associated with forcing estimates, such as in predicting mass concentrations of aerosol species.

The single-scattering albedo for desert dust is not well established. This has implications on the ability to calculate the forcing due to changes in the loading of desert dust—even regarding sign.

A major issue is that of the pertinent size distribution to employ for various aerosol types. For radiative transfer purposes, the precise form of the size distribution is not important. Rather, what is needed is the optical depth and effective radius for each

aerosol species (see Lacis and Mishchenko, this volume). Typically, in models that calculate aerosol forcing, a finite number of fixed aerosol types and sizes is carried in the computations, with optical depth and vertical distribution as free parameters. Consistency is needed between aerosol parameters as used in radiative models and field measurements, where a continuum of particle sizes and composition mixtures is encountered. To the extent possible, size distributions should be reported separately for individual species and distinction made between dry and moisture-laden aerosol sizes.

What information is missing that is necessary to describe direct radiative forcing of aerosols on climate?

To answer this question it is useful to consider types of studies that may be conducted to evaluate the effect of aerosols in climate models. Three kinds of sensitivity studies may be identified:

1. A passive radiative forcing evaluation, where aerosols are regionally prescribed and the alteration of the radiation balance is calculated.
2. A fixed aerosol concentration distributed regionally as above, but with interactive climate adjustment to evaluate the feedback processes.
3. A prognostic aerosol and climate simulation, where the distribution of aerosols is calculated region by region based on known or estimated source strengths.

The third type of study is explicitly directed toward the climate change issue. It is least constraining and allows the eventual connection with the indirect effect of CCN–cloud albedo relation. It further avoids the definition of a so-called "background aerosol" type. This background is essentially the result of long-range transport of aerosol and precursor gases from source regions to remote locations.

The basic inputs required to describe direct radiative influences of aerosols in climate models are the global, three-dimensional distributions of mass concentration, chemical composition, size distribution, and degree of mixture of the various chemical species within the particles. Associated with chemical composition, models need the complex refractive index and water uptake (or equivalent information). In some climate models, these parameters are prescribed. In others, which are just now being developed for the first time, the model incorporates, on a grid corresponding to that of the GCM, a description of the processes maintaining the aerosol chemical loading and microphysical and optical properties—emissions of aerosols and precursor gases (and perhaps the temporal variation of the emission rate), transformation (gas-to-particle conversion, evolution of size distribution, etc.), vertical and horizontal transport, and wet and dry deposition.

Measuring the global, three-dimensional distributions of the parameters desired for climate models is theoretically possible but prohibitively expensive. Satellites can determine the global distribution of a limited number of aerosol properties integrated

over a vertical column, with some additional information on the vertical distribution; however, they cannot be used to determine the chemical composition of the particles. Surface and aircraft-based platforms can provide much of the very detailed information needed, but with limited coverage in space and time. Even the *in situ* measurements are limited. For example, measurements of aerosol optical properties are obtained at a limited number of wavelengths, whereas the models need to integrate spectrally dependent variables over a wide range of wavelengths, from the ultraviolet to the infrared. Similarly, the models need the angular distribution of light scattering by the particles in all directions, which is rarely measured directly; the closest that current field-proven technology can come is the measurement of the angular scattering function integrated over two different angular ranges or at a very few selected angles.

The conceptual tool that bridges this gap between the requirements of the climate models and the limitations of present and anticipated future measurement techniques is the *closure* experiment (Ogren and Ramaswamy et al., both this volume). Here, an overdetermined set of variables is measured, and the directly observed value of one dependent variable is compared with the value calculated by a model. The extent of agreement between the measured and modeled values is an indicator of the combined uncertainty contributed by the assumptions in the model and the limitations of the measurements. Satisfactory agreement, assessed by propagation of the observed discrepancy through the climate model, can be used to indicate which approximations are acceptable and which modeling or measuring techniques must be further refined. Many different types of closure studies can be defined, ranging from studies on the microscale of the suitability of theoretical mixing rules used to describe the refractive index of a particle containing several chemical species, to the perturbation of reflected radiance fields resulting from anthropogenic sulfur emissions on hemispheric scales. This combined modeling and observational approach has not been systematically used in the past; however, it offers great potential for evaluating the uncertainties produced by limitations in both the models and the measurements.

The representation of aerosol size distributions in global models is an unresolved issue. The common use of log normal or gamma distributions for representation of observations often leads to systematic differences resulting from the shape of the particular function. These differences, however, are minimized if size distribution comparisons are made in terms of their respective effective radii (Hansen and Travis 1974). For this reason, we recommend that several components be used to represent realistically each aerosol mode (nucleation, accumulation, coarse modes). In our view, accurate representation of aerosol forcing is more likely if chemical species are treated separately rather than trying to represent aerosols according to regional types such as maritime, continental, stratospheric, etc. The use of several size modes for a given aerosol species may lead to more accurate representation of the prognostic evolution of size distributions in climate models.

Climate models can suffer from numerical problems resulting in violation of mass conservation or production of unphysical negative concentrations. This problem is most acute in treating a tracer field that contains sharp spatial gradients. Until climate

models can faithfully reproduce observed concentrations of passive tracers in the current climate situation, the prognostic treatment of aerosol size distribution will remain inaccurate.

Direct measurements of aerosol optical properties are necessary for verification and closing the problem of input parameters in climate models. Measurements of optical properties as a function of relative humidity are also necessary for validation of the description of aerosol properties in climate models.

An outstanding issue is the determination of the state of mixture of aerosol particles. This is a key element in the determination of optical properties and also in the determination of hygroscopic properties. It is one important factor in the activation process of CCN since it provides a critical relation between aerosol and cloud properties. Adequate measurement or determination of the aerosol state of the mixture should be considered a necessary part of the sampling procedure.

The above issues pertain to the several different aerosol types of concern. Initially the question arose as to whether aerosols should be classified according to location (e.g., stratosphere, marine, desert) or chemical type (sulfate, sea salt, mineral dust). We generally felt that classification by chemical type will lead to more accurate representation, since a given aerosol will not change its "type" upon being advected to a new location. A possible exception to this generalization is the stratospheric sulfuric acid aerosol, which is unique and strongly decoupled from other locations. Nonetheless, aerosols have been traditionally classified as to location, and it is thus useful to consider the several types of aerosols whose properties must be treated in models. These aerosol types are briefly described below and key questions noted.

Stratospheric Aerosols

Compared to tropospheric aerosols, the stratospheric aerosol morphology is well understood and has been for some time. Owing to sulfur injections from the troposphere (COS, SO_2, volcanic SO_2), sulfuric acid vapor forms in the stratosphere and is converted to sulfuric acid/water droplets, through both nucleation of new particles and growth of existing particles through co-condensation of sulfuric acid and water vapor. The sulfuric acid fraction depends only on the stratospheric water vapor mixing ratio and the temperature and, for normal values of these parameters, is about 75% by mass. Except possibly during polar winter, the aerosol is composed of liquid droplets.

In the absence of major volcanic eruptions, the size distribution can be well represented as unimodal with a number mode radius, r_0, near 0.08 μm. Following major eruptions, a bimodal distribution is observed that results from new particle nucleation ($r_0 \sim 0.05$–0.15 μm) and growth on the preexisting aerosol ($r_0 \sim 0.3$–0.5 μm) (Hofmann 1987). This results in a major perturbation to the stratospheric radiative and chemical equilibrium. The resulting transient radiative perturbation immediately following major eruptions is much larger locally than average tropospheric aerosol forcing. The accumulation mode is transported out of the stratosphere in two to three years, leaving a unimodal distribution that reverts to the background mode in five to

six years. The lifetime depends on the properties of the aerosol (altitude, particle size) and thus can vary with time. The typical e^{-1} time is roughly one year. Long-term observations (past 25 years) of the stratospheric aerosol indicate that the stratosphere has been volcanically disturbed about 90% of the time. Periods of apparent nonvolcanic or "background" conditions (loosely defined as an observation of no decay for periods exceeding one year) are rare (e.g., 1979, 1990) and short lived. On average, the stratosphere has been at about five times this "background" value since 1970 (Hofmann 1990).

Major uncertainties concern the stratospheric aerosol distribution in the absence of volcanic activity. Can a "background" (meaning nonvolcanic) aerosol be usefully defined through natural and anthropogenic sulfur emissions? The latter include both surface emissions and emissions in the lower stratosphere and upper troposphere associated with air traffic. If so, is this "background" changing? These questions are more important for the indirect effect of heterogeneous chemical ozone loss in the lower stratosphere and concomitant changes in greenhouse forcing of climate, and for upper tropospheric contrail/cloud formation.

The ability to describe the radiative influence of future volcanic eruptions is severely hampered by the inability to conduct *in situ* measurements during the early stages following eruptive events. Such measurements are crucial to the characterization of the aerosol when climate forcing is maximum and before a large fraction of the early aerosol falls out.

Upper Tropospheric Aerosols

Because of the relatively low mass concentrations of the upper tropospheric aerosol, it is not considered important radiatively; it, however, is probably very important for heterogeneous chemistry and may interact in an important way with cirrus clouds. The upper tropospheric aerosol is poorly observed and appears to be somewhat more complicated than the stratospheric aerosol, although the composition is probably similar (sulfuric acid/water, possibly with some ammonium sulfate and nonvolatile cores). There is probably a trace amount of soot; however, it is not well characterized. There are large seasonal variations at mid-latitudes associated with stratosphere–troposphere exchange, especially in spring. The lifetime of aerosols in this region appears to be intermediate to that in the stratosphere and surface, i.e., weeks to possibly a month or two. In the summer, the upper troposphere appears to have low concentrations of particles larger than about 0.1 μm radius, and for this reason it may be a source region for new particle production (Raes et al., this volume). An unknown fraction of these particles is related to aircraft traffic. During spring, long-range transport of larger particles, possibly Asian desert dust and/or pollutant sulfate aerosols, results in perturbations to the aerosol distribution in the middle to upper troposphere (2.5–10 km) at northern mid-latitudes. However, long-term measurements (20 years) at one clean continental site (Laramie, Wyoming) indicate that the column loading of submicrometer aerosols in the 2.5–5 km altitude range has been decreasing at a rate of

about 1.5% per year, similar to the reduction of SO_2 emissions in the United States and Europe (Hofmann 1993).

Uncertainties abound in the global distribution of upper tropospheric particle size and composition, and require a coordinated program of satellite and *in situ* observations together with modeling studies to advance our knowledge of this component.

Marine Boundary Layer (MBL) Aerosols

It is important to delineate those MBL air masses that are truly remote from continental influences from those that have recently advected from a continental region and are in transition from continental to marine. Many air masses, particularly in the Northern Hemisphere, never complete this transition before reencountering land.

In the remote MBL, particles larger than 1 μm radius are mainly sea salt whereas those smaller than 0.2 μm radius are predominantly non-sea-salt sulfate. In the range between 0.2 μm and 1 μm the particles are mixed, with relative abundance determined by the two source strengths. Measurement-based algorithms that give either the flux or surface concentrations of sea-salt particles as a function of wind speed are available, but their accuracy at large particle sizes ($r > 10$ μm) is known to be deficient, and their application at diverse locations has not been demonstrated. Sea-salt particle generation extends down into the 0.01–0.1 μm range, but the number concentration of these particles is usually negligible compared to the concentration of sulfate aerosols except in extremely clean (low-sulfate) conditions with higher wind speeds (Hoppel and Frick 1990).

The situation is more complicated in regions where circulation causes transport off of continents. The time required for continental air to transform into a truly marine air mass is ill defined; it depends upon meteorology and is usually ten days or less. The following typical scenario was observed to occur in air masses advecting off the East Coast of the U.S. in clear weather. At the coastline, the number-size distribution was rich in small particles, with a single mode (peak) radius of about 0.01 μm. The peak radius increased to about 0.06 μm and decreased in amplitude during the first twenty-four hours over water. Modeling indicated that the growth was primarily the result of condensation, with coagulation playing a secondary role. After clouds formed at the top of the MBL, the submicrometer distribution split into two modes, with peaks at about 0.03 μm and 0.1 μm radius. The split is caused by the increased sulfate generated in the subset of particles that serve as CCN where the air is processed through the cloud-topped MBL. When precipitation occurs, particles are scavenged. Larger particles are replenished at the sea surface, and the fine particle mode is sustained by gas-to-particle conversion occurring in the MBL or by entrainment of particles from the free troposphere (Hoppel and Frick 1990; see also Clarke 1992).

The greatest optical extinction in the MBL usually occurs near the top, where relative humidity is the highest. Because of the high humidity, MBL particles are usually solution droplets and thus spherical. This situation generally justifies simplifying assumptions regarding the phase function.

Industrial Pollution Aerosols

Industrial emissions are responsible for substantial loadings of atmospheric aerosols (see chapters by Möller and Penner, both this volume). Several distinct aerosol constituents need to be considered: sulfates (including sulfuric acid and its neutralization products, mainly with ammonia: NH_4HSO_4 and $(NH_4)_2SO_4$), nitrates (mainly NH_4NO_3), organic compounds, and graphitic carbon. Because sources of these materials are regionally proximate, these aerosols may be mixed; however, the state of the mixture is not well established and will evolve, for example, by coagulation or by coating existing soot particles with secondary sulfuric acid. These aerosols may be directly emitted into the atmosphere (primary aerosols) or (except for graphitic carbon) may be formed in the atmosphere by gas-to-particle conversion. Sources of these aerosols are at or near the surface, so these aerosols are confined mainly to the lower troposphere. The residence time of these aerosol particles depends on size but is about a week for particles in the range of the principal mass of material (ca. 0.05–1 μm, the so-called accumulation mode; Whitby 1978). Principal removal mechanisms are wet deposition (mainly incorporation into cloud drops followed by delivery to the surface in precipitation) and dry deposition (eddy diffusion to the vicinity of the surface followed by diffusion, impaction, or sedimentation). The removal of these species from the atmosphere differs for the different species, e.g., ammonium nitrate aerosol may decompose to form gas-phase NH_3 and HNO_3, which can be taken up at the surface. The source of the organic aerosol is not well established, e.g., the mix between primary versus secondary (see Penner, this volume). The source inventory of elemental carbon is also not well established but is currently being addressed in research. Nitrate aerosols are becoming increasingly important relative to sulfate in western Europe as emissions of SO_2 are decreasing in response to regulations instituted to decrease acid deposition (see Hidy and Wolf, this volume). On the other hand, emissions of SO_2 in China and southeastern Asia may be expected to increase.

Arctic Haze

Although the Arctic is remote from major source regions of anthropogenic aerosols, the concentrations of these aerosol constituents can occasionally be comparable to those of the rural continental U.S., for example, 1–3 $\mu g\ m^{-3}$ for sulfate and ca. 0.2 $\mu g\ m^{-3}$ to 0.4 g $\mu g\ m^{-3}$ for soot. The particular pathway for injection of these aerosols, together with their slow removal rate in a stable stratified atmosphere, allows for a regional increase of the concentration during winter and spring seasons. The Arctic aerosol is characterized by well-defined stratified layers with a total optical depth of about 0.2 and a single-scattering albedo near 0.87 for dry particles, increasing with relative humidity (Blanchet and List 1983). A large part of this optical depth is due to water uptake by sulfuric acid and is responsible for scattering visible radiation. The larger particles in moist conditions can also alter the longwave radiation budget in the water vapor window region. In the spring, observations and models indicate that the

soot component of Arctic haze is a significant warming agent on a regional scale. According to GCM simulations, the lower atmosphere warming can amount to 1°C or 2°C (Blanchet 1989). Even if the concentration of soot was to increase by an order of magnitude on a regional scale, the predicted warming does not exceed about 2°C in these model simulations, because of a redirection of excess heat from the radiative forcing to snow and ice melt and a resultant change of atmospheric circulation. This result is supported by the observed trend (Kahl, Charlevoix et al. 1993; Kahl, Serreze et al. 1993) of up to 2°C warming in the lower troposphere during spring for the last 40 years in the high-latitude Arctic ocean region. Careful analysis of these results could provide elements for closure of the relation between aerosol characteristics and climate change, a very difficult challenge at the present.

This well-aged aerosol is remarkably constant in terms of size distribution and composition. For this reason, it has been possible to make reliable aerosol models of the optical properties (Blanchet 1989) in agreement with Mie theory and observations, as has also been done, with some success, for the stratospheric aerosol (see above). In contrast, it is difficult to achieve similar closure for most other tropospheric aerosols because of the high variability in size distributions, poorly known refractive index, and irregular shape of solid particles.

In recent years, with a decreasing trend in sulfur emissions in Europe and in the former Soviet Union, the western Arctic gives evidence of a reduction of the anthropogenic components whereas the eastern Arctic seems to retain a relatively high concentration of pollution during winter and spring. Monitoring of the aerosol concentration and type should be maintained to allow for an assessment of the relationship between source regions and the response of anthropogenic aerosols characteristics in remote areas.

Organic Aerosols

Penner (this volume) presents an overview of the sources and atmospheric concentrations of the organic component of atmospheric aerosol particles. It is important to remember that organic carbon is a collective term that refers to the large number of individual compounds observed in the atmosphere. These compounds can exhibit very different behaviors in the atmosphere, in terms of their interactions with radiation and with other chemical compounds (including water). Many open questions concerning the role of organic compounds in both direct and indirect climate forcing remain:

- *What is the scattering efficiency of organic carbon, and how does it compare to the scattering efficiency for sulfate compounds?* Penner's estimate of the direct climate forcing from organic aerosols assumed that organic carbon and sulfates had similar mass-scattering efficiencies (Penner et al. 1992) However, multiple regression of the relative contributions of organic carbon and sulfate to aerosol light scattering yielded different coefficients for the two categories (White 1990), suggesting significant differences in their size distributions or refractive indices.

- *What is the contribution of organic compounds to the light absorption coefficient of the atmospheric aerosol?* Rosen et al. (1979) reported that a literature search of the optical properties of organic compounds yielded only five compounds (out of 13,000) listed as gray or black in appearance. Nevertheless, questions of the contribution of organic compounds in light absorption are still raised. The situation is further complicated by the separation technique used to distinguish organic and elemental carbon during combustion analysis, since changes in the amount of light absorbed by the particles as temperature increases are used to correct for the pyrolysis of organic carbon.
- *What is the molecular speciation of the organic aerosol?* Current analytical techniques for organic aerosols either determine a total mass of carbon or the masses of the specific species that are identified and quantified in the analysis. To date, a balance between the total mass of organic carbon and the combined mass of the individual molecular species has not been obtained, leaving open questions about the sources and atmospheric consequences of the unidentified or unquantified species.
- *What is the state of mixture of organic compounds with inorganic species? How do organic compounds change the hygroscopic growth characteristics and cloud-nucleating properties of other components of the aerosol?* Water-soluble and surface-active organic compounds have the potential to control the rate and extent to which particles react to changes in relative humidity. As Penner (this volume) points out, surface-active organic compounds can be expected to alter the interaction of water vapor with other species, and the effect can either enhance or diminish the response of the particles to changes in relative humidity. Water-soluble species can decrease the surface tension of an aqueous droplet, thereby enhancing the cloud droplet nucleating ability of a particle. Alternatively, an organic layer on a droplet can slow the rate of growth or evaporation.
- *What are the removal mechanisms for organic carbon?* Unlike sulfur, the fully oxidized form of carbon is a gaseous compound (CO_2). Consequently, it is possible, and perhaps likely, that continued oxidation and particle-to-gas conversions in the atmosphere are major removal mechanisms for some particulate organic species. Furthermore, the effects of organic compounds on the hygroscopic growth of aerosol particles are also likely to affect the efficiency of precipitation as a sink for these compounds.

Biomass Burning Aerosols

Aerosols of biomass burning origin are among the most complex of the aerosol types that affect climate. The chemical composition of these aerosols is sensitively dependent upon the generating process, such as biomass species, fire temperature, and meteorological condition. The aerosol optical characteristics, therefore, vary greatly. For example, the single-scattering albedo of biomass combustion smoke ranges

from 0.5 to 0.9, although there is no systematic measurement of such optical characteristics accompanied by *in situ* chemical sampling. Kaufman and Nakajima (1993) estimated that the imaginary part of the index of refraction of smoke for Amazon forest fires is as small as 0.02 to 0.03 in its absolute value, as retrieved by an AVHRR remote-sensing technique that is an inverse method requiring assumptions. This means that nonab-sorbing carbon constituents other than graphitic carbon (soot) dominate, resulting in a high single-scattering albedo, around 0.80 to 0.85, unless the fire temperature is extremely high. It is noteworthy that these values are close to the imaginary index value of heavily polluted urban atmospheric aerosols such as observed in Asian areas. However, far from the origin of the smoke, the single-scattering albedo of these aerosols can, by condensing water, reach a value as high as 0.98.

The optical thickness of the smoke layer often exceeds unity, so that the longwave radiative forcing cannot be ignored, competing with the shortwave radiative forcing. Therefore the volume concentration of the coarse particle mode should be investigated carefully. Since soot particles ($r_0 < 0.05$ μm) tend to coagulate, forming chain-type complexes, investigation of the structure of mixture of the accumulation and coarse mode aerosols with soot particles is necessary along with measurement of soot concentration.

Closure measurements are required, combining optical and chemical measurements, over biomass burning active areas, such as the Amazon, Africa, and South Asian areas. In these areas, the underlying surface albedo also plays an important role in controlling the climate forcing of biomass burning aerosols. Since the thick tropical rain forest has a low albedo of several percent, the existence of the smoke layer increases the planetary albedo. The Kuwait oil fire event shows that a strongly absorbing soot-rich aerosol sometimes stabilizes the smoke layer, resulting in a distinct change in the transport of smoke. This fact establishes the need to investigate the interaction mechanism among the ground albedo, aerosol characteristics, and transport processes.

Soil-derived Aerosols

Windblown dust extends over a significant portion of the globe. Satellite analysis of optical depths over marine areas show that larger optical depths occur in regions thought to be influenced by such dust (Rao et al. 1989; Durkee et al. 1991). Long-range transport of dust has also been found at a number of sites (Schutz et al. 1990; Arimoto et al. 1992). Dust particles are larger than most other types of aerosols (typically greater than 1 μm) and exhibit more variable values of the imaginary part of the refractive index. The imaginary part of the refractive index of dust can be controlled by the abundance of minor species, such as iron oxides, which vary locally in the soil. Due to the large size and sometimes high absorption, the effect of dust aerosols can be of either sign. These aerosols significantly affect the regional balance of thermal radiation. Finally, mineral particles have absorption bands in the ultraviolet and near infrared. Hence mineral aerosols are not well characterized by "typical" visible

properties, such as mid-visible single-scattering albedos. More work needs to be done, especially to determine the wavelength-dependent optical properties of these aerosols. Human effects on changing dust aerosols, primarily through changing land-use patterns, also need to be better quantified.

Biological Aerosols

Particles of biological origin are derived from living organisms and exhibit identifiable biological features, for example, protein-containing cells, microorganisms, and fragments of living things. This definition includes a wide spectrum of types and sizes of biological aerosol particles. The smallest are viruses (0.005 μm $\lesssim r \lesssim$ 0.25 μm) followed by bacteria ($r \gtrsim$ 0.2 μm), protozoa ($r \gtrsim$ 2 μm), algae and fungi ($r \gtrsim$ 0.5 μm), pollen grains ($r \gtrsim$ 5 μm; Macher 1993), plant debris like leaf litter, parts of insects, and human and animal epithelial cells (supposed $r \gtrsim$ 1 μm).

Biological aerosol particles with a radius greater than 2 μm derive from plants that release pollen and spores actively or passively into the atmosphere. Biological particles less than 2 μm originate to some extent from the "bubble-burst" mechanism (Blanchard and Syzdek 1972), but are mostly produced by industrial (sewage plants, etc.), agricultural (fertilizing, etc.), or municipal activities (Spendlove 1974).

The concentration of primary biological particles in the atmosphere is dependent on seasonal and meteorological conditions as well as on location. The influence of rural regions leads to an increase of particles larger than ca. 0.1 μm, like pollen and spores, whereas the influence of urban regions with industrial and municipal activities as sources for bacterial aerosol leads to a higher concentration of these smaller biological particles (Matthias-Maser and Jaenicke 1994).

Biological particles may play an important role in cloud physics. Decomposition products of terrestrial vegetation have been found to be good ice nuclei (Schnell 1982). The protoplasm of some pollen have hygroscopic properties (Dingle 1966). Under certain circumstances (at temperatures between –2°C and –5°C), bacteria also have ice-nucleating capabilities (Vali et al. 1976; Levin et al. 1980). These cloud-forming processes are dependent on the size of the particles.

What is the reason for the lack of data necessary to evaluate aerosol forcing? Are new instruments required? Are more systematic measurement programs required? Or do the data exist, and all that is required is to compile a climatology of existing data?

Measurements of aerosol chemical composition and optical properties have been conducted for decades, raising the question of why so many open questions remain regarding the description of aerosol influences in climate models. A fundamental answer is that neither the measurements nor the models have treated the chemical and optical aspects of aerosol climate forcing in a *coupled* fashion. Aerosol chemical

measurements are rarely combined with aerosol optical measurements and measurement protocols often neglect the dependence of the measured parameters on particle size and relative humidity; thus many aerosol measurements are useless or irrelevant for climate modeling. Likewise detrimental to estimating aerosol climate forcing is the fact that the description of radiative transfer in global climate models has treated aerosols as either nonexistent or as fixed in space and time. It is only recently that even the mass concentration of any trace aerosol species has been included as a prognostic variable in a global-scale, three-dimensional climate model (Taylor and Penner 1994).

Analogous to line atlases tabulated for greenhouse gases, the spectral dependence of refractive indices for individual chemical species of aerosols provides the means for calculating the radiative parameters—δ, ω_0, $\beta(\phi)$—over the full spectral range of solar and thermal wavelengths for specified particle size distributions. Mie-scattering results are theoretically rigorous for homogeneous spheres, and T-matrix calculations (Mishchenko and Travis 1994) are equally rigorous for randomly oriented spheroids with specified size and aspect ratio distributions. There is, however, only limited evidence on the preferred shape characteristics. An alternative approach, which is capable of describing the scattering patterns of arbitrary-shaped particles, is provided by the discrete dipole approximation (cf. Flatau and Stephens 1990). Unfortunately, this technique suffers from limitations concerning the size of the particles. Hence, semi-empirical methods, such as that proposed by Pollack and Cuzzi (1980), should also be used. These methods have been proven to reproduce the main features of nonsphericity (cf. Wendisch and von Hoyningen-Huene 1992).

Although there have been very significant improvements in aerosol measurement technology over the past two decades, improvements are sorely needed in the methods used to determine key aerosol properties, such as single-scattering albedo, phase function, and the composition of individual particles. A further factor limiting advances in the ability to estimate aerosol climate forcing has primarily been the lack of application of existing methods, generally resulting from an underappreciation of the importance of atmospheric aerosols by agencies responsible for funding basic and applied atmospheric research. Closure studies, where an overdetermined set of variables is measured to allow comparison of calculated versus measured values of a dependent variable, are woefully rare. The few early closure studies, such as those by Ensor et al. (1972) and Russell et al. (1979), have not been repeated systematically. Consequently, an understanding of the ability of the models to predict the key optical properties of atmospheric aerosols and the resultant radiative effects is still lacking. This leads to an inability to quantitate the uncertainty in radiative forcing.

Much effort has been devoted to compiling aerosol climatologies, that is, a three-dimensional representation of particle size distribution and refractive index for different aerosol types. To date, the validation of these climatologies has been inadequate, and serious questions have been raised as to their accuracy (Penner et al. 1994). A fundamentally more serious limitation is that the approach does not link the "climatological" properties with the source fields for the chemical species comprising

the aerosol. As a result, this approach is of limited usefulness for policy decisions that require estimates of the response of aerosol climate forcing to changes in the source strengths and distributions.

INDIRECT FORCING

Do aerosols indirectly affect clouds? How do aerosols interact with cloud microphysical processes, and do the interactions cause changes in radiative properties of the cloud? What is the evidence that aerosols affect cloud microphysical properties and/or albedo? Does the evidence go beyond ship tracks? What information is required to incorporate indirect effects into climate models?

Aerosol particles are the building blocks of all clouds. It is natural, then, to expect that changes in aerosol particle characteristics can have an impact on the properties of clouds. Early work by Twomey (1974, 1977) outlined the processes by which cloud reflectance would be expected to increase because of the effects of increases in concentrations of aerosol particles that act as CCN. Since then, numerous examples of qualitative as well as quantitative evidence of this effect have been presented (see King et al., this volume). These examples include observations of correlation between aerosol concentrations (or sometimes CCN concentrations), cloud droplet number/size, and cloud reflectance. Many of these observations are aided by satellite and aircraft measurements made at near-infrared wavelengths, where cloud reflectance is determined primarily by droplet size rather than optical thickness. Some of the most compelling evidence comes from observations of the effects of ship exhaust on clouds, referred to as ship tracks. The currently available set of observations suggests that the Twomey effect operates in some cloud systems on the local to global scale (Kaufman, this volume).

The currently available observations do not go very far toward providing a quantitative estimate of the climate forcing due to this indirect aerosol effect. Currently, the important processes involved in the indirect cloud effect are not quantitatively understood such that they can be accurately included in climate models. For direct forcing, a credible relationship exists between the source-strength of gaseous precursors to aerosol production and the aerosol optical properties. However, in the indirect cloud effect, the forcing estimate requires a quantitative link between anthropogenic aerosol production and cloud optical properties. The required links include:

- *Anthropogenic aerosol production → CCN*: It is unlikely that a simple relationship will ever be developed to describe this linkage, since the ability of a particle to activate a cloud droplet depends on particle size, solubility, and surface properties; number concentration is not uniquely determined by the mass of gaseous precursors such as SO_2 (Braham 1974; Radke and Hobbs 1976; Hudson 1991).

- *CCN → cloud droplet number or size*: This link depends on the susceptibility of the cloud system to droplet size/number change. Key variables include the slope of the supersaturation spectrum of CCN, the droplet size of the unmodified cloud, updraft velocity, and other processes that control droplet growth such as cloud dynamics and mixing processes that determine cloud parcel lifetimes. MBL cloud models (Baker and Charlson 1990; Ackerman et al. 1993) imply that significant nonlinearity may exist in the relationship between CCN concentration and droplet number, and that the droplet change potential may saturate at high droplet numbers. Observational studies include Leaitch et al. (1992), Pueschel et al. (1986), Gillani et al. (1992), Alkezweeny et al. (1993).
- *Cloud droplet number or size → cloud top albedo*: The radiative properties of clouds, especially their shortwave albedo, are dependent on the size distribution of the cloud drops and are therefore subject to anthropogenic influences resulting from changes in the concentrations of CCNs. This phenomenon is difficult to demonstrate because of the inherent variability in cloud liquid water path, which influences albedo. One approach to quantifying the phenomenon is by means of so-called "ship tracks," which are bright streaks embedded within marine stratus clouds that have been observed to form about the exhaust effluent of ships (Coakley et al. 1987; Scorer 1987; Radke et al. 1989), thereby illustrating the effects of a point source of aerosol on the cloud properties in immediate contrast to proximate unperturbed regions of the cloud. The cloud droplet number concentration increases and radius decreases as the ship effluent interacts with the cloud. The magnitude of the resulting albedo increase is dependent on the magnitude of these microphysical changes. However, the increase in reflectance due to increased droplet concentration can be modified and even reversed if the added aerosol includes significant absorbing material (Kaufman and Nakajima 1993).
- *Cloud droplet number or size → cloud lifetime*: An additional radiative influence of anthropogenic CCN may come from an increase in cloud lifetime. The formation of precipitation within clouds is strongly dependent on the size distribution of cloud droplets. Albrecht (1989) described potential increases in cloud lifetime if precipitation rates were reduced by injection of CCNs. The resulting decrease in droplet size could stabilize the drop distribution, making it resistant to formation of large, precipitation-size drops via coalescence processes. Ackerman et al. (1993) presented model results describing the collapse of stratus cloud formation processes from a deficit of CCN, again indicating that additional CCN could stabilize a cloud aginst this dissipation process.

Initial global-scale modeling studies (Jones et al. 1994; Boucher and Lohmann 1995) have made some crude assumptions of the relationships, but there is little empirical evidence that they are realistic. There is even doubt that any unique relationship between SO_2, for example, and CCN exists, since the rate of new particle

production versus particle growth depends on secondary parameters, such as preexisting particle distribution. Furthermore, current global-scale models do not generate realistic, interactive clouds especially for low-layered clouds that are likely to be the most susceptible to the indirect cloud effect. It is therefore unreasonable to expect these models to simulate microphysical characteristics of clouds accurately.

On the cloud scale, process studies are required to determine the aerosol–CCN–cloud droplet relationships. Recent and planned experiments that will make progress toward quantifying these processes include ASTEX (Atlantic Stratocumulus Transition Experiment), SCAR (Smoke Clouds And Radiation), SOCEX (Southern Ocean Cloud Experiment; J. Gras, pers. comm.), and MAST (Monterey Area Ship Track) Experiment (King et al., this volume). Studies of ship tracks provide a particular advantage in that a quantifiable source perturbs a small region of cloud with nearby unperturbed cloud under the same meteorological forcing. Also, during MAST ships will be controlled to vary intentionally the concentration of aerosol. From this, it may be possible to determine the critical relationship between CCN concentration and droplet size.

Additional indirect effects, such as precipitation suppression and modification of cloud amount, are even further from quantitative understanding. Models can simulate precipitation changes but few observations of drizzle modification are available. Some recent observations (Hudson 1991) in a marine environment (ca. 300 km west of Baja California) indicate that some regions exhibit extremely low CCN concentrations (1–5 cm^{-3} at 0.6% supersaturation). Formation of these regions presumably involves the positive feedback of drizzle removal and large droplet formation. The CCN concentrations are so low that cloud formation processes are inhibited. The global distribution of these regions is currently unknown. However, if anthropogenic aerosol particles stabilize cloud formation in these regions and thereby increase cloud cover, the radiative impact has the potential to exceed the forcing from droplet size change alone.

The indirect effect of biomass burning aerosols is complicated because of the competing effects of enhanced droplet number and enhanced absorption. Clouds interacting with smoke exhibit reduced droplet size, as observed by satellites (Kaufman and Nakajima 1993). Yet the sign of the shortwave cloud albedo change is difficult to predict. The enhanced light absorption by particles, due to multiple scattering in the cloud, sometimes causes a significant reduction of the cloud albedo as expected by Twomey (1977). The sign of the cloud albedo change depends on the CCN concentration, cloud type, and surface albedo.

Is the mechanism of aerosol influence on cloud properties the same for all cloud types? Stratus and cirrus are two types of clouds that are potentially susceptible to aerosol influence on albedo.

Although aerosols may affect cloud properties, other phenomena such as boundary layer depth, wind shear, time of day, or temperature will also control such parameters

as liquid water content or mean particle size. For example, numerical models of ship tracks show that whether the liquid water content of the cloud varies with aerosol abundance may depend on the time of day when the aerosol injection occurs. Likewise, such models show a diurnal cycle of cloud droplet size due to changing boundary layer structure, not diurnal variations of CCN concentrations. Such diurnal variations of particle size have been noted in satellite studies. One-dimensional models of ship tracks suggest that variations in aerosol concentration, boundary layer depth, time of day, etc. exert significant influence on the magnitude of the albedo change occurring in a ship track. However, the dissipation of ship tracks and their frequency of formation may be related to the degree of vertical shear of the horizontal wind in the boundary layer. Modest shear may disperse and dilute the aerosol to such an extent that no ship tracks can form. It is also possible for ship tracks to create self-sustaining circulations due to their radiative differences with the surrounding boundary layer. Ship exhaust outside of ship tracks would be less likely to induce such circulations, so extended exhaust trails may not occur. At present, the processes controlling ship track lifetime are not known.

Limited theoretical studies suggest that cirrus clouds may respond quite differently than stratus clouds to variations in aerosol concentrations. Cirrus clouds contain very few ice crystals and are therefore highly selective in the aerosols they nucleate. One hypothesis is that selection occurs apparently because once ice crystals form, they quickly reduce the ambient supersaturation. Hence, the cloud particle number densities are self-limiting to some extent and may not significantly change if aerosol concentrations change. Contrails are sometimes cited as a frozen analog to ship tracks, since they seem to indicate that the addition of aerosols to the atmosphere can induce cirrus cloudiness. It has been observed that the size of ice crystals in contrails is smaller and the ice crystal density larger than in nearby cirrus, which is evocative of the "Twomey effect." However, contrails differ significantly from ship tracks because the cloud formation occurs in the warm, moist exhaust from the engines, not in the preexisiting cloud as it does for ship tracks. Contrails may form because the supersaturation in the exhaust exceeds that in the ambient air, although both ambient air and exhaust may be supersaturated with respect to ice. In the contrail case, the high ice-particle concentrations may or may not result from the high supersaturation in the exhaust rather than from the enhanced aerosol concentration in the plume. Although contrails doubtless represent information that is relevant for understanding how ice clouds respond to aerosols, that information seems to be less clear than in the case of ship tracks because of the confounding changes in supersaturation.

We concluded that many factors control cloud radiative properties in addition to the concentration of ambient aerosols. Current understanding of these controlling factors is limited. Hence, both observationally and theoretically, the signal of aerosol indirect effects on climate is difficult to separate from variability resulting from other cloud physical processes.

What properties of aerosol particles influence their ability to serve as nuclei for cloud drop formation? What are the relationships between particle number concentrations, CCN concentrations, and cloud droplet concentrations?

These questions have been the subject of investigation for more than one generation of cloud physicists, but combined studies of the chemical *and* microphysical properties of both aerosol particles and cloud droplets are a relatively recent phenomenon. The known, key properties influencing the ability of a particle to serve as a CCN are the mass of soluble material and number of ions formed per unit mass, responsible for vapor pressure lowering as a function of radius, and the surface tension responsible for vapor pressure increase. Köhler's (1923) relationship describing the size of a solution droplet in equilibrium with its surroundings has served as the foundation of understanding droplet nucleation, with droplet growth subsequently controlled by molecular diffusion and thermal conduction. Recent studies of the chemical, microphysical, and hygroscopic growth characteristics of both the scavenged (CCN) and unscavenged (interstitial) particles suggest that factors other than size and soluble mass can also be important in determining which particles become the condensation nuclei in a cloud (Svenningsson et al. 1992; Hallberg 1994). Among the likely factors are the presence of dissolved or surface-active organic species, which can either enhance or diminish the uptake of water by the growing droplet as relative humidity increases. Furthermore, dynamical limitations might be important, as the ambient supersaturation must remain above the critical level long enough for a particle to grow to its critical size if it is to become activated.

Other recent work has found that the fractional activation of accumulation mode particles (number concentration, N_{amp}) is nearly complete at low particle concentrations ($N_{amp} \leq 400\ cm^{-3}$) but that this fractional activation falls off with increasing N_{amp}. At higher N_{amp}, the fractional activation can be related to other environmental variables such as lapse rate, which may be a surrogate for updraft velocity and in turn maximum supersaturation (Gillani et al. 1992). These findings are consistent with modeling studies (e.g., Jensen and Charlson 1984). Again, however, the number of studies actually conducted are quite few, and we believe more work is necessary before the uptake of aerosol particles into cloud drops can be confidently parameterized in climate models.

Another phenomenon that deserves attention is the influence of successive cloud encounters on the nucleating ability of particles. Most clouds evaporate rather than precipitate, and thus most particles experience numerous cloud events; the average number is thought to be about ten (Pruppacher and Klett 1980). It is thus possible that a particle activated in an earlier, more vigorous cloud, such as a clear-air cumulus, would be modified in the process, e.g., by the occurrence of aqueous-phase SO_2 oxidation that adds solute mass. This would have the effect of decreasing the maximum supersaturation required for activation in a subsequent cloud cycle, allowing that particle to serve as a droplet nucleus in a less vigorous cloud, such as a marine stratus. This is yet another mechanism whereby anthropogenic emissions can change cloud drop concentrations and thereby exert an indirect radiative forcing.

Two questions that should be emphasized in future studies are:

1. What factors determine which particles actually become activated in real clouds?
2. What is the role of entrainment and mixing in altering the relationship between the chemical composition and size distribution of the particles existing at cloud base and the number concentration of cloud droplets aloft?

What are the mechanisms for production of new particles and CCN in the vicinity of clouds and in the MBL?

A source of particles is required to sustain the concentration of CCN in the MBL against depletion by precipitation scavenging. This source was originally believed to be sea-salt particles injected directly at the sea surface. It is now well established that only a small fraction of the CCN are sea-salt particles. An exception may occur infrequently in remote regions and is most likely during higher seas, when sea-salt particle generation is enhanced. A number of hypothetical processes to provide the non-sea-salt particles have been suggested. These include:

1. transport from the continents in the free troposphere with subsequent entrainment into the MBL,
2. nucleation of new particles in the MBL,
3. formation of new particles in or just above the tops of marine stratus, with subsequent entrainment into the MBL,
4. nucleation in the free troposphere with subsequent entrainment into the MBL.

It is probable that each of these mechanisms contributes to the supply of CCN; however, their relative importance under different environmental conditions has yet to be sorted out. Nucleation of new particles, both in the MBL and free troposphere, has been observed; the former may be an infrequent occurrence whereas the frequency of the latter has not yet been established. Once small particles have been nucleated (see Pts. 2–4 above), growth to radii sufficiently large to be active as CCN must occur. The processes controlling nucleation and subsequent growth are not well understood, and certain aspects of the proposed mechanisms are problematic.

Removal of particles (CCN) in the MBL is more rapid than in the free troposphere because of the higher rates of precipitation scavenging that occur in the MBL. Particles in the free troposphere (either from continental sources or formed *in situ*) are readily transported to more remote regions and entrained into the MBL (Raes et al., this volume), where they are eventually removed by precipitation scavenging.

The concentration of CCN is important in determining the number of cloud droplets formed in marine stratus and thus impacts cloud albedo. Cloud susceptibility (change in albedo divided by change in droplet concentration) is inversely proportional to the cloud droplet concentration. This is related to, but not the same as, sensitivity to

changes in the concentration of CCN. For a given cooling rate, the number of cloud droplets depends not only on the CCN concentration but also on the slope of the CCN supersaturation spectrum, which can vary significantly in different situations. Attempts to obtain the change in cloud droplet concentration from total sulfate load or even total CCN concentration measured at a single supersaturation are not physically based and must be validated by other means.

What observations can be made and process studies conducted that will improve understanding of the influence of aerosols on clouds? Do we need new instruments, or are concentrated campaigns sufficient?

Since it was shown theoretically (Twomey 1959), and later confirmed experimentally (Twomey and Warner 1967), it has been widely accepted that droplet concentration in a cloud is established within the first few hundred meters of cloud base and is governed primarily by the CCN spectrum below cloud base with a secondary dependence on updraft velocity. To strengthen confidence in the aerosol influence on clouds, it is therefore important to make measurements beneath clouds of aerosol chemical composition, size distribution, and CCN spectra; these quantities can subsequently be compared to *in situ* microphysics and radiation measurements. In the latter context, an extremely valuable measurement to make within clouds is the angular distribution of scattered radiation deep within the cloud over the wavelength range approximately 0.5–2.3 μm. This is perhaps most clearly demonstrated in clouds modified by emissions from ships, as described by King et al. (1993 and this volume).

In the area of microphysics, the particulate volume monitor (PVM; Gerber et al. 1994) is an important new instrument for measuring liquid water content and integrated particle surface area, from which the effective radius can be derived. Both the liquid water content and effective radius are important microphysical properties that significantly affect cloud radiative properties. Both parameters are known to change under some conditions of cloud modification by aerosols (e.g., ship tracks, which are summarized by King in Figures 13.3 and 13.4, this volume). Distinct advantages of the PVM probe over conventional measurements of effective radius are the greatly increased sampling volume (factor of 5×10^4) and a much shorter response time. However, as the PVM probe is unable to measure the full drop size distribution, it is still important to measure the drop size distribution using conventional instruments such as the optical array probe and the forward scattering spectrometer probe (Knollenberg 1981), which provide the size distribution of drizzle-sized droplets ($r \cong 100$ μm). A promising new instrument for measuring the size distribution of droplets is under development (Lawson and Cormack 1995). The measurement principle is similar to that of the PVM, but the forward scattered light is measured with high angular resolution using charge-coupled detector arrays. The size distribution of the droplets is obtained from the measured forward scattering function by numerical inversion techniques. Another recent technique that appears promising is examination of spatial distributions and interdroplet distances of drops in clouds (Borrmann et al. 1993).

Radiation measurements that are important, though sometimes difficult to obtain, are nearly simultaneous reflection and transmission measurements, both narrow-band radiance and broad-band flux measurements. Such measurements were obtained in ASTEX by coordinating aircraft flights with satellite overpasses (NOAA AVHRR or Landsat TM) with ER–2 aircraft (above), University of Washington C–131A (within) and UK C–130 (below), since all of these aircraft, as well as the satellite, carry sophisticated spectral scanning radiometers. Such a "simple" flight pattern shows (a) whether cloud reflectance, transmittance, and internal scattered radiation are consistent with microphysical measurements (optical thickness, effective radius, single-scattering albedo) or whether some unexplained "anomalous absorption" is acting on this cloud layer, and (b) whether changes in microphysical characteristics produced by changing aerosol injections indirectly affect the cloud's radiative properties. To aid in assessing the impact of aerosol on clouds and radiation, it is also advantageous to use microwave radiometers to derive the integrated liquid water content of the cloud layer. This is especially useful in studying ship tracks since, as noted above, observations suggest changes in cloud liquid water content sometimes result from aerosol injections from ships.

Additional *in situ* measurements that have proved beneficial in the past are counterflow virtual impactor (CVI) measurements. This instrument affords the opportunity to sample selectively cloud droplets of diameter greater than a specified value, to evaporate subsequently the liquid water, and to measure the optical, chemical, and microphysical properties of the aerosol incorporated within the droplets. In the past such measurements have been used to show that the soot content within marine stratocumulus clouds off the West Coast of the U.S. was as great as 160 ng soot/g cloud water (Twohy et al. 1989).

Finally, the relatively new concept of cloud susceptibility, proposed by Twomey (1991) and Platnick and Twomey (1994), is an important parameter to be inferred in the future. This parameter, discussed in detail by Toon and King et al. (both this volume), suggests that not all clouds are equally susceptible to changing their albedo when modified by enhanced aerosol injections. In particular, marine clouds with unperturbed albedo (≈ 0.5), low droplet concentrations, and large droplets are more susceptible on a per particle basis to the indirect effect of aerosol on clouds than are continental clouds having high droplet concentrations or cirrus clouds with low reflectance. Hence the above discussion has focused on marine clouds in the lower boundary layer (e.g., marine stratocumulus and stratus clouds). Thus far the concept of cloud susceptibility has not included the effect of decreased albedo due to black carbon. Confidence in understanding of these processes will be greatly increased by modeling the interactions of aerosols and clouds and comparison with measurements.

The above discussion implicitly assumes well-calibrated and well-characterized sensors. In view of the difficulty of accurately measuring liquid water content and effective radius, we strongly recommend that PVM and FSSP be flown simultaneously and that their accuracy and calibration be checked independently in wind tunnel tests.

Similarly, CVI measurements require well-characterized measurements of droplet sizes being examined as a function of inlet geometry, counterflow speed, and aircraft velocity. Finally, all remote sensing or *in situ* applications of spectral radiation measurements depend directly on the accuracy of the radiometer calibration. Consequently, future designs of satellite sensors will require a greater emphasis on calibration accuracy and signal-to-noise characteristics.

A new instrument, the droplet aerosol analyzer (DAA), has recently been used in ground-based cloud and fog studies to determine the size distributions of cloud (fog) droplets as well of unactivated haze (interstitial) particles. Cloud droplets are unipolarly charged and then dried in a diffusion dryer. The residue particles of the cloud droplets and of the interstitial haze particles are then analyzed in a differential mobility analyzer (DMA); the number of charges is an indicator of the original droplet size. The size distributions of various size cuts can then be determined with subsequent DMAs to ascertain the size distributions of the residue particles. This new device, developed by Martinsson (B. Martinsson, University of Lund, Sweden, pers. comm., 1994) has not yet been adapted for aircraft use.

Few reliable measurements have been made of the supersaturation within clouds, and those only from stationary platforms (Gerber 1991). For measurements to contribute to understanding aerosol–cloud interactions, they will have to be made around the critical supersaturation, frequently 0.15% or less. The value of these measurements would be enhanced by simultaneously measuring the cloud droplet size distribution and comparing these measurements with CCN spectra obtained below cloud base. Clearly for such studies, deployment in aircraft is necessary.

In addition to coastal and intercontinental shipping lanes, an important area where aerosol injections to clouds are expected to modify the radiative properties of clouds is in large regions of biomass burning (see Kaufman, this volume). An experiment in the Amazon region of Brazil, planned for September 1995, should contribute further to quantitative understanding of the indirect effect of this important aerosol type on climate.

INDIRECT CLIMATE EFFECTS OF AEROSOLS VIA HETEROGENEOUS CHEMISTRY

How do aerosols affect the climate through heterogeneous chemistry in the stratosphere? How well are the properties of polar stratospheric clouds understood, especially as they influence heterogeneous chemistry? What is the role of aerosols of volcanic origin?

Aerosols in the lower stratosphere, together with increased concentrations of chlorofluorocarbons, are believed to be associated with the reduction of stratospheric ozone (~3% per decade) at mid-latitudes that has occurred during the past 15 to 20 years. Ozone in the lower stratosphere is a radiatively important gas; for example, it has been

estimated that the increase in greenhouse warming associated with the increase in chlorofluorocarbon emissions has been offset by cooling associated with the concomitant ozone loss in the lower stratosphere.

Heterogeneous chemical loss of ozone in the lower stratosphere is believed to be related to reactions occurring on sulfuric acid/water particle surfaces in nonpolar regions and on PSC element surfaces in regions where temperatures fall below the condensation point of nitric acid vapor. Important reactions are thought to include the surface hydrolysis of N_2O_5 and $ClONO_2$ to form HNO_3. In the absence of intense sunlight (e.g., polar spring, mid-latitude winter/spring), which would photolyze HNO_3 and return nitrogen to the active phase, the loss of reactive nitrogen (NO_x) results in destruction of ozone owing to HO_x and ClO_x chemistry below ca. 25 km. Above this altitude, NO_x reduction would result in the net production of ozone owing to reduction of the natural NO_x ozone sink. Heterogeneous N_2O_5 hydrolysis, which occurs efficiently at normal stratospheric temperatures, is believed to be saturated in the lower stratosphere (below 20 km), even for background aerosol levels. The heterogeneous conversion of $ClONO_2$ to nitric acid and active chlorine increases exponentially with aerosol water content and thus with decreasing temperature. This reaction becomes important below temperatures of about 205 K, and thus well above the formation temperatures of nitric-acid PSCs (195 K) (Toon, this volume).

The long-term downward trend of stratospheric ozone at mid-latitudes may be a local effect, related to local concentrations of H_2SO_4/H_2O aerosol, and/or a remote effect, involving PSC-related ozone depletion in the polar regions with subsequent transport of ozone-poor air to mid-latitudes.

The Pinatubo eruption provided a useful test of models of H_2SO_4/H_2O aerosol-related heterogeneous effects on active nitrogen and ozone. Both were observed to decrease beyond natural fluctuations at mid-latitudes in accord with theory. The fact that ozone reduction was greatest the second winter following the eruption is related to the change in altitude of the aerosol layer and the associated change in chemistry (Hofmann, Oltmans, Harris et al. 1994). Unusually low ozone in the Antarctic spring of 1993 is believed to be related to ozone loss augmentation associated with the Pinatubo aerosol (Hofmann, Oltmans, Lathrop et al. 1994). Ozone loss may have occurred over Antarctica on Pinatubo aerosols at temperatures above those at which PSCs are formed. Pinatubo-derived aerosols may also have affected ozone concentrations indirectly, through changes of the PSC size distribution caused by the enhanced volcanic aerosol serving as PSC nuclei. The complete recovery of the ozone layer at northern mid-latitudes in 1993–1994 (the third winter) was accompanied by a return to very low aerosol surface-area densities at this time (Hofmann, Oltmans, Harris et al. 1994). Recent models of these processes are able to reproduce the observations, at least qualitatively (Rodriguez et al. 1994; Tie et al. 1994).

There are feedbacks involving stratospheric temperatures and water vapor concentrations. Both stratospheric cooling, related to ozone reductions, and increases in stratospheric water vapor, which are now observed to be increasing at northern mid-latitudes at a rate of about 1% per decade owing partially to methane increases

(Oltmans and Hofmann 1995), will result in larger, more aqueous aerosol droplets and concomitant increases in heterogeneous chemical effects on ozone.

Although the nature of the volcanic aerosol is relatively well understood, the details of PSC formation are not. Uncertainties lie in knowledge of the particle phase, both of the condensation nucleus (H_2SO_4/H_2O liquid droplet or H_2SO_4 solid hydrate) and the resulting PSC particle (liquid $H_2SO_4/HNO_3/H_2O$ system or solid HNO_3 hydrate). Cooling rates are probably important and different phases may result under different conditions. Processes similar to those in PSCs may also take place in cirrus and Antarctic stratus clouds.

Are there heterogeneous reactions of climatic importance that are influenced by tropospheric aerosols? What observations and process studies are required to elucidate this chemistry?

In addition to reactions involving the oxidation of SO_2 in cloudwater, which are of obvious importance in governing the amount and form of sulfate aerosol in the atmosphere, there are additional heterogeneous reactions in the troposphere that have a potential climate influence, specifically as they can modify the concentration of ozone, a key greenhouse gas. One such reaction that has been identified is the uptake of N_2O_5 and/or NO_3 on aqueous aerosol particles and in clouds. These reactions have the effect of depleting NO_x, thereby altering the cycle of reactions that produce ozone. Furthermore, the aqueous-phase reaction of two HO_2 radicals to form hydrogen peroxide in clouds and the alternative pathway that destroys O_3 are important (Lelieveld and Crutzen 1990). In addition to the direct influence on climate through the influence on ozone concentrations, there is further indirect influence through other chemical reactions initiated by ozone photolysis, involving e.g., $O(^1D)$ and OH. The inclusion of the N_2O_5 hydrolysis reaction in a global-scale chemical model leads to a reduction in O_3 concentrations of up to 25% in some locations and seasons (Dentener and Crutzen 1993). However, the actual extent of this reaction is sensitive to aerosol loading and physical state (the particles must have an aqueous phase) and necessitates further investigation to allow more confident modeling. Likewise the rate and extent of in-cloud HO_2 reactions are sensitive to cloudwater pH, leading to yet another indirect influence of anthropogenic sulfur emissions on climate. Ultimately, models will be required to address all of these phenomena simultaneously. Other reactions of potential importance are possible generation or destruction of free radicals on suspended soil dust particles. Laboratory studies have indicated the possible significance of there reactions; however, little is known about their occurrence in the atmosphere.

The oxidation of SO_2 in sea-salt droplets by O_3 has received recent attention (Sievering et al. 1991; Chameides and Stelson 1992). This reaction can proceed at a substantial rate because of the high pH of these droplets. The reaction removes SO_2 that might otherwise lead to direct radiative forcing. The importance of this reaction is not yet fully established. Consideration of the gaps in instrumentation to examine these reactions point to the need for measurements of the composition of single

particles, to reveal the state of mixing in aerosols. A promising approach appears to be chemical ionization of volatilized particles. This technique may be especially useful if applied to cloud drop nuclei sampled with a conterflow virtual impactor.

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