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Group Report: Magnitudes and Geographical Variations and Uncertainties of Properties of Tropospheric and Stratospheric Aerosols and Their Forcing

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INTRODUCTION: GREENHOUSE GASES AND AEROSOLS

Anthropogenic greenhouse forcing is to a large extent caused by increasing infrared radiation (IR)-absorbing gases (e.g., CO_2, CH_4, and N_2O) that are nearly homogeneously distributed around the globe. Recent studies have led to the understanding that, even though some greenhouse gases are distributed uniformly throughout the atmosphere, radiative forcings of climate are not globally homogeneous (Kiehl and Briegleb 1993). Moreover, climate responses to these forcings, possibly enhanced through changing water vapor and cloudiness, are likely to be very heterogeneous. Furthermore, some IR-absorbing gases (e.g., O_3) and aerosol particles are not distributed uniformly; this underscores the need to consider explicitly geographical variations of climate forcings and responses. In particular, the geographical distributions of aerosol particles are controlled by processes that act on local to regional scales, limiting the particle residence times in the atmosphere, and making them patchy (Charlson et al. 1991; Kiehl and Briegleb 1993). Nevertheless, aerosol climate effects may extend over

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hemispheric or even global scales (Taylor and Penner 1994; Roeckner et al., this volume).

Geographical and temporal variations in aerosol concentrations, including anthropogenic changes and resulting climate forcings, are controlled in the first place by source and sink distributions, which to a large extent also determine the aerosol characteristics. The residence time of aerosol particles depends primarily upon their size. The aerosol size fraction that is assumed to be most relevant in climate forcing has a residence time of less than a week in the boundary layer and of up to a few weeks in the free troposphere. One of the primary questions that we posed in our discussions is what part of the atmospheric aerosol is anthropogenic, in particular that of sulfate, windblown mineral dust, organic particles, soot, and possibly even of sea spray. Further, we addressed the role of atmospheric processes in relation to geographical distributions of aerosols, in particular physical and chemical processes that control the residence times of aerosols and their precursors. Microphysical and multiphase chemical processes also regulate aerosol size distributions and aerosol radiative effects, notably scattering and absorption of solar radiation and IR absorption. These effects are strongly affected by particle deliquescence, which emphasizes the need to define the aerosol hygroscopic growth characteristics and how these are influenced by atmospheric chemistry.

The characterization of aerosol properties should be accomplished in such a manner that their incorporation in global chemical and climate models becomes possible. Simultaneously, the question must be raised: what spatial resolution should models have in order to represent realistically aerosols and the processes that control their abundances and radiative effects? Models can be used to study temporal changes of aerosols as a function of changes of emission sources. However, the confidence in models, particularly general circulation models (GCMs) with interactive aerosol representations, should be established through validation programs on the basis of high quality data of aerosol properties and distributions. Observational data on a global scale can be obtained through satellite observations excluding chemical data. However, some pitfalls should be overcome, for example, the determination of the single-scattering albedo through remote-sensing techniques (also via *in situ* measurements). We emphasize the need to understand the uncertainties of the aerosol properties derived from satellite observations. Further, available information from the past should be used, e.g., from time series obtained by monitoring efforts and from ice core analyses, to validate the models so that, ultimately, the effects of anticipated regional shifts or expected changes in anthropogenic emissions can be predicted. This raises a further question: to what extent do aerosol forcings mask greenhouse warming effects and, if so, how might they change in the future?

ANTHROPOGENIC CONTRIBUTIONS TO AEROSOL SOURCES

On a global scale, most of the atmospheric aerosol mass loading is determined by a few major aerosol components, crustal material (mainly mineral desert dust), sea salt,

organic material, soot, and inorganic sulfur and nitrogen species. A main question is to what extent these aerosols are perturbed through anthropogenic emissions. A primary source of sulfur in the atmosphere, particularly in the Northern Hemisphere, is energy production from fossil fuels. About two-thirds of the global source is anthropogenic (IPCC 1992). Also, the global budgets of nitrogen species that contribute to aerosol formation, mainly nitrate and to a lesser extent ammonium, are dominated by anthropogenic emissions. Important sources of sulfur and nitrogen species are fossil-fuel and biomass combustion as well as animal husbandry (IPCC 1992).

The globally predominant source areas of mineral dust are the Sahara, Gobi, and Arabian deserts together with the adjacent semiarid zones prone to land-use change. Measurements on islands in the northern Atlantic, e.g., on Barbados and Bermuda, and from remote locations in the Pacific Ocean have shown that submicrometer dust particles can be transported over thousands of kilometers before being removed from the atmosphere (Prospero et al. 1981). This has also been successfully simulated by global models (e.g., Wefers and Jaenicke 1990). The optical depths of these dust clouds can be significant (Rao et al. 1988). Land-use changes and increases of arid areas enhance the abundance of windblown mineral dust. In particular, desert fringes in semiarid tropical areas are important sources of crustal material to the atmosphere. Changes in these sources could contribute significantly to climate forcings; however, there is no observational information available to quantify this contribution.

Sea-salt particles, released by the sea-spray production mechanism, dominate the aerosol mass in the marine boundary layer (though not the aerosol number concentrations). Anthropogenic surfactant films of organic substances may influence the sea-spray production mechanism. However, these films are only maintained at relatively low wind speeds, while under these conditions sea-spray production is small. Further, oil spills could play a role in this respect, although it is expected that these cause only short-term localized effects as a result of their relatively rapid disintegration. A speculative effect from anthropogenic greenhouse warming is that, under changing wind regimes, sea-spray fluxes to the atmosphere may be altered (Latham and Smith 1990), although the climatic relevance of this may be small.

Solid and condensed organic species can contribute strongly to the aerosol mass in both the polluted and background troposphere (Cronn et al. 1977; Chesselet et al. 1981). Our group distinguished primary aerosols, emitted directly as particles, from secondary organic aerosols; the latter are formed in the atmosphere from gaseous organic compounds. The term "organic" aerosols in this report includes condensed hydrocarbons and living and dead cells or cell fragments. Measurements in Los Angeles indicate that about 90% of the organic aerosol in urban environments may be primary, whereas, on a global scale, only about 80% of the organic aerosol may be primary. Measurements in polluted as well as remote marine locations show that up to 20% of the particles are of biological origin (Jaenicke and Matthias-Maser 1993). The aerosol mass fraction of organics in the atmosphere appears to have strong regional

variations (Penner, this volume). In the western United States, the organic fraction (as C) approximately equals that of sulfate, while in the eastern U.S. the sulfate fraction usually exceeds the organic fraction by about a factor of two.

The organic component from biomass burning dominates the aerosol abundance in the Tropics during the dry season (Penner et al. 1991). The sources of organic aerosol in these regions are controversial. While measurements during the biomass-burning (dry) season in central Africa and Amazonia indicate substantial organic aerosol loadings, almost equal concentrations of organic particles of similar composition have been observed during the wet season in Brazil (Andreae, pers. comm.). This apparent near constancy of condensed organic matter in these regions can, for the time being, only be explained by speculating that vegetation is a direct source of organic aerosols.

It has also been established that smoke particles from biomass and fossil-fuel combustion are effective CCN (Twomey 1974). Since the fraction of organics in such aerosols appears to be substantial, its contribution to CCN number concentrations may be significant. Observational data from a rural location (Pt. Reyes) and from the marine boundary layer (Puerto Rico) are consistent with the assumption that particles containing organic matter are activated as cloud droplets, so that anthropogenic emissions of these species may contribute to the indirect as well as the direct climate forcing effect, the former through cloud albedo changes. Observations by Novakov and Penner (1993) suggest that, even though the sulfate aerosol mass may exceed the organic mass, the number concentration attributable to organics may dominate that of sulfate in the size range normally associated with CCN number concentrations. The state of aerosol mixing (i.e., internal vs. external) was not determined in these experiments.

From the effects of the recent eruption of Mt. Pinatubo, it is known that changes in the aerosol loading of the lower stratosphere can exert a significant radiative forcing upon climate (Minnis et al. 1993). The nonvolcanic sulfate aerosol in the stratosphere (Junge layer) is maintained through photochemical breakdown of carbonyl sulfide (COS) and other sulfur species upwelling from the troposphere. Balloon-borne measurements in the central U.S. (Wyoming) over the past three decades show that, as a result of regularly occurring volcanic eruptions, the stratospheric aerosol is present at its assumed background level only during relatively short periods (~10% of the time). The measurements during these relatively rare occasions over the past decades yield indications of an increase of the sulfate load (Hofmann 1990). However, it is questionable whether a true stratospheric "background" aerosol can thus be defined so that a secular trend is difficult to establish. The aerosol loading of the stratosphere is largely determined by sulfates of volcanic origin. The optical depth of the background aerosol in the stratosphere is very small, so that the direct climate forcing by upwelling anthropogenic sulfur species from the troposphere is not expected to be significant.

ATMOSPHERIC PROCESSES CONTROLLING SULFATE AEROSOL PROPERTIES

When estimating the magnitude of the direct forcing of anthropogenic sulfate aerosols, the two most uncertain factors are (Penner, Charlson et al. 1994):

- the fraction of SO_2 oxidized to sulfate,
- the average residence time of the sulfate particles in the atmosphere.

Neither can be measured directly but must be inferred from model simulations and validation of such simulations against measurements. The first factor, which is of the order of 0.5, depends upon the relative magnitudes of deposition processes (mainly dry deposition) and oxidation of SO_2. The uncertainty range is estimated to be a factor of ca. 1.5. About the same uncertainty applies to the second factor, which is determined mainly by the efficiency and spatial distribution of precipitation scavenging. The average sulfate residence time is estimated to be about three to seven days (e.g., Langner and Rodhe 1991).

To reduce the above uncertainties, more measurements are needed of the concentration of SO_2 and aerosol sulfate in areas both outside and within the most polluted regions in Europe and North America (of which some data already exist) and especially in the free troposphere (of which almost no data exist). Refinements of the transport models are also required, including better descriptions of the oxidation of SO_2 as well as the vertical mixing and scavenging associated with convective and frontal cloud systems. Several of the transport models used to simulate the large-scale distribution of aerosol sulfate seem to underpredict the concentration in high latitudes during winter. It is possible that additional SO_2 oxidation processes are active even under such conditions. In particular, multiphase processes within clouds are expected to be important, in the face of the reality that the representation of cloud transformation processes in global models is still in its infancy. The total source strength of anthropogenic SO_2 and its geographical distribution, of course, have a direct influence on the magnitude of the forcing. It is possible that the uncertainty in source strength is larger than estimated previously. In particular, emission estimates from non-OECD countries are relatively uncertain. Most of the uncertainty is due to a lack of knowledge of the fuel sulfur content. Also, the emission height is not always well known; the altitude at which pollutants are transported strongly influences the extent to which these species are subject to removal in the vicinity of the sources.

The efficiencies by which aerosol particles are formed and by which they grow through deposition of gases and coagulation determine, to a large extent, the aerosol optical properties and their capability to form cloud droplets. The complex atmospheric processes that generate ultrafine particles, control coagulation, and determine how cloud "processing" influences the aerosols, are likely to be different for sulfates and biomass-burning aerosols. However, the details of the processes, in particular the

generation of new particles and how rapidly they grow into the accumulation mode size range, are not yet fully understood. Several observations have shown that, whenever conditions are favorable, "bursts" of new particles are encountered (e.g., Clarke 1993). The most important factor in the formation of new particles is that relatively little preexisting particle surface area should be present, so that newly produced sulfuric acid molecules or other particle precursors are not scavenged by these particles.

Bi-molecular particle nucleation ($H_2SO_4 + H_2O$) is relatively well understood and can be simulated by models. It is known, however, that the addition of NH_3 to a mixture of H_2SO_4 and H_2O enhances particle formation. The theory of the tri-molecular mechanism is poorly understood, although it is certain that the process is favored energetically over bi-molecular nucleation. Laboratory studies have indicated that the nucleation process may be independent of light. It is likely that new particles are formed continuously, but that relatively small molecular clusters cannot survive the coagulation process in the presence of significant aerosol surface areas. Favorable conditions for particle nucleation often occur in the upper troposphere as a result of the relative scarcity of preexisting aerosol surface (Hofmann 1993). After new particles are formed in the free troposphere, they may be mixed into the marine boundary layer by entrainment, which could be a significant supply of new particles (Raes 1995). This hypothesis, however, needs support through additional measurements in the remote marine boundary layer and free troposphere.

Once an aerosol is formed, its scattering cross section is strongly affected by hygroscopic growth and deliquescence. Thus aerosol hygroscopic growth characteristics must be defined and the mechanisms resulting in the observed hygroscopic properties understood. Three areas of interest have been identified:

1. Most aerosol chemical and physical properties are determined at very low relative humidities (in the instruments). To relate these results to ambient conditions in an attempt to quantify the direct aerosol forcing, transformations as functions of relative humidity must be understood.
2. How do hygroscopic particle properties affect the way they are processed by clouds and thus are related to the indirect aerosol forcing?
3. What do the observed hygroscopic particle properties reveal about aerosol sources and the aging processes that have a bearing on aerosol lifetimes?

The concept of an internally mixed aerosol and experimental data based on large volume samples can no longer be accepted as the universal view on aerosol chemical composition since *in situ* single-particle growth data have given systematic results (at least partly) of external mixtures. Yet a comprehensive theory for this feature as well as sensitivity studies which would evaluate the range of aerosol optical parameters and resulting direct forcing as a consequence of these findings, are still lacking.

Much can be learned by making use of data sets that have been established in the past, for example, how aerosol chemical compositions have changed in time, so that

we may be able to assess what consequences these changes have had for aerosol radiative forcing. To estimate the radiative forcing by aerosols (preindustrial to present), the mass loadings of various types of aerosols (e.g., sulfate, organic carbon, soot, etc.) are needed. In addition, information on the particle number and mass changes in time due to changes in emissions are required. Changes in emissions also lead to changes in chemical composition of aerosols. These lead to changes in the refractive index, hygroscopic properties, and size distribution of the aerosol. These changes in aerosol physical properties lead, in turn, to changes in aerosol radiative properties, and hence radiative forcing. The question is: to what extent are these changes important in estimating the anthropogenic radiative forcing? An example is the reduction in SO_2 emissions over the past decade in industrial regions in Europe and the eastern U.S., while emissions of other aerosol precursors, i.e., nitrogen oxides and ammonia, may have increased. The tropospheric aerosol abundance over one clean continental site (Laramie, Wyoming) has decreased at the same rate as that of SO_2 (Hofmann 1993). We recommend returning to sites where key optical parameters were measured 20 to 25 years ago, and repeating these measurements to see if the aerosol optical properties have changed. Changes in scattering efficiency would be of great interest.

AEROSOL REPRESENTATION IN MODELS

An important issue is how to include particle formation and "aging" of the aerosol size distributions into global models. Two options were identified:

1. Prescribing "maps" of aerosol types of which optical properties are only a function of the model calculated particle mass.
2. Explicitly including controlling processes.

It can be stated that, although the second option is the more desirable one, knowledge is incomplete and computer resources are still a limiting factor. Explicit representation of particle size changes as function of distance from the source regions in models may not yet be possible. An additional option that may be mentioned for future applications of satellite observations is to try to relate particle properties derived from remote-sensing measurements to *in situ* measured quantities.

A new data compilation has been presented in which the global radiative forcing fields through natural and anthropogenic aerosols have been included, the Global Aerosol Data Set (GADS); radiative forcings have been calculated with a delta-Eddington radiation model (Schult and Grassl, pers. comm.). Data are now internally consistent with respect to aerosol number concentrations, aerosol mass per volume, and optical properties. In GADS, 11 main aerosol components are defined through their particle size distribution and their wavelength-dependent index of refraction. Typical components are water-soluble and -insoluble species, soot, sea salt, and

mineral dust; the latter in three different size classes. For these components, the normalized optical parameters are determined separately. Additionally, the chemical composition fields of the aerosols are defined, including their vertical profiles for January and July. Preliminary conclusions made from including the GADS into the Hamburg climate model indicate that the radiative forcing is very sensitive to the aerosol chemical composition.

Only a few global-scale modeling studies have been carried out that were based on treatment of sources, transformations, and transport and sink processes, although the model representation of many of these processes was highly parameterized and aerosol size distributions were not treated explicitly (e.g., Wefers and Jaenicke 1990; Langner and Rodhe 1991; Pham et al. 1994; Penner, Atherton, and Graedel 1994). A tracer model for mineral dust was recently developed that treated particles sizes explicitly (Tegen and Fung 1994). Currently, improved trace gas source distributions are becoming available (e.g., Global Emissions Inventories Activity, GEIA, a component of the IGAC project), that is intended to be used for regional and global modeling. The spatial resolution of these emissions is 1 × 1 degrees, which sets the preferred scale for near future global modeling studies (Graedel et al. 1993). Computational costs can be limited by applying relatively high model resolutions on regional scales and using these computations as input for coarse-grid global models, e.g., through model "nesting." Ideally, the processes that control aerosol size distributions and their chemical compositions should be parameterized on the basis of first principles, i.e., explicit description of aerosol formation and coagulation in models rather than prescribing empirical relationships. Also, cloud–aerosol interactions need to be included to account for in-cloud oxidation of SO_2, which strongly affects the scattering properties of the aerosol that is released by cloud evaporation (Lelieveld and Heintzenberg 1992). Furthermore, it is extremely important that these model simulations are validated against observational data on regional and global scales.

AEROSOL OPTICAL PROPERTIES AND REMOTE SENSING

An important parameter that defines the aerosol particle scattering relative to its absorption properties is the single-scattering albedo, ω_0. This parameter, however, is not known to a sufficient degree of accuracy for mineral dust, aerosols composed of organic material, and for industrial aerosols. Also, ω_0 has not yet been studied for ultraviolet (UV) wavelengths. The latter may be particularly significant for windblown desert dust, which appears to have a relatively high absorption in the UV; this may be significant for photochemistry in the lower atmosphere. Furthermore, the absorption and emission of IR by coarse-mode (diameter >1 μm) particles is usually neglected in radiative transfer calculations, although it is known that these particles absorb significantly at these wavelengths.

Satellite measurements in the solar spectrum over bright surfaces can be used to derive ω_0 (Kaufman 1987). Determination of ω_0 from space is very sensitive to aerosol

size distributions and particle nonsphericity (Nakajima, pers. comm.). A method should be developed that combines satellite measurements with optical measurements of the aerosol column from the surface. Simultaneous *in situ* measurements should be performed for validation and to define the aerosol chemical components and particle size distributions, so that a relation can be inferred between the aerosols and their properties and sources.

Considering the current and future availability of remote-sensing data from satellites, we wish to emphasize the need to evaluate the uncertainties of measured aerosol properties and how these can be reduced. While stratospheric aerosol optical depths are measured relatively accurately, this is not the case for tropospheric aerosols, largely due to the lack of adequate instruments and continuity of observations. Much has been learned about the spatial and temporal distributions of tropospheric aerosols through analysis of AVHRR observations (Kaufman, this issue). However, the development of multichannel instruments in the U.S., Japan, and Europe will strongly enhance our capability to infer aerosol optical depths and ω_0 in the troposphere. In particular, the measurement of ω_0 is important to assess the radiative forcing of climate caused by changing aerosol concentrations. Mineral dust and aerosols that contain very large particles show a strong wavelength dependence of absorption. Thus, there is a need for multiwavelength satellite sensors that also cover the thermal IR parts of the spectrum. The accuracy of global-scale aerosol data sets should be improved by conducting experiments aimed at closure between aerosol column measurements from space and from the surface in combination with *in situ* measurements of aerosol optical parameters and chemical compositions. These global data sets, in turn, will support validation of GCMs in which aerosols are simulated explicitly. The study of source characteristics in relation to aerosol optical properties will also benefit from LIDAR measurements in association with *in situ* measurements by aircraft.

The strongest solar and IR radiation-absorbing aerosol in the atmosphere is made up of impure elemental carbon, soot. It is produced by combustion processes, in part from natural sources (e.g., forest fires ignited by lightning) and by anthropogenic biomass burning and fossil-fuel use. Soot is usually not measured by size resolving techniques and the measurements often include other organic particles, so that many current data sets are unreliable. Deposited soot can influence the albedo of the Earth's surface, especially if the undisturbed albedo is very high, for example, of snow. Such an albedo reduction has been observed in the Cascade Mountains, Washington State, and in the Arctic sea ice (Grennfel et al. 1981). This albedo effect may be amplified in melting snow if the soot remains on top of the surface (Warren and Wiscombe 1985). Albedo effects of deposited mineral dust are roughly fifty times less than those of soot, which are thus insignificant, except close to the dust source. Before any definite conclusions about the climate forcing effects by anthropogenic aerosols are drawn, the influences of soot must also be evaluated. Realistic ω_0 values for mixtures of soot and sulfate must be determined to be included in climate forcing calculations. Even if the net forcing of such mixtures at the top of the atmosphere would be zero as a result of cancellation by scattering and absorption, significant effects from a

vertical redistribution of heating fluxes within the atmosphere can still be expected, although these effects are not expected to be as large as the direct effect by sulfate aerosols.

The potential effect of aerosols within clouds to enhance absorption of solar radiation has been recognized for over twenty years. Comparison of measured and calculated cloud reflectances has led to the hypothesis of additional cloud absorption. It is not clear whether these differences between measurements and calculations are due to problems with measurement techniques used in the past or with erroneous assumptions in the model calculations (e.g., plane parallel geometry). A few measurements have been performed of soot in clouds (Heintzenberg 1988; Twohy et al. 1989), which do not indicate a significant contribution to in-cloud shortwave absorption. Stephens and Tsay (1990) have reviewed the various hypotheses to explain this *anomalous* cloud absorption. At present, more observations are required to improve our understanding of the importance of aerosols in increasing cloud absorption.

REGIONAL SOURCE CHANGES AND AEROSOL MASKING OF GREENHOUSE FORCINGS

Past and future changes in human population, land-use changes, and implementation of air pollution abatement measures (per capita emission changes are most significant) affect the regional distributions and extent of aerosol-related emissions to the atmosphere. The economic changes within the former Soviet Union may have caused an almost stepwise decrease in aerosol precursor emissions in this region. The stratospheric aerosols generated as a result of the eruption of Mt. Pinatubo have caused a temporary cooling forcing; this has been confirmed by satellite observations and modeled succesfully (Boville et al. 1991; Young et al. 1994). These events should be used to validate GCMs in which these changes can be prescribed. Future emission increases of aerosol-precursors may be expected in the developing world. China, in particular, has rapid economic growth and large reserves of sulfur-containing coal. The characteristic regional forcing by increasing aerosol particle concentrations in the atmosphere should be pointed out very clearly. Although the IPCC (1992) report mentions sulfur emission scenarios up to the year 2100, it does not make any regional distinction, which underscores the need to emphasize this aspect.

Estimation of current climate forcings by aerosol particles requires the explicit evaluation of aerosol precursor source and sink characteristics as well as spectrally resolved radiative transfer calculations in GCMs. As reviewed by Kiehl and Rodhe (this volume), a significant negative forcing by sulfate aerosol particles is likely to occur, masking greenhouse warming and locally "contaminating" surface temperature records. Another effect of increasing both CO_2 and sulfate in the model is an enhancement of the latitudinal climate forcing gradient between the Tropics and the mid-latitudes in the Northern Hemisphere, which could have effects on the dynamics of the atmosphere. To calculate realistically long-term anthropogenic climate changes

in GCMs, representations of all relevant greenhouse gases (including ozone, which also acts on regional scales) and of aerosols (including varying aerosol optical properties) need to be included simultaneously. Only the inclusion of all forcings can accurately account for counteracting effects or mutual enhancements of climate forcings that act in very different ways.

SUMMARY AND CONCLUSIONS

As a result of sulfur emissions reaching the stratosphere from relatively strong volcanic influences, it is not certain whether a true stratospheric "background" aerosol can be defined. Nevertheless, there are some indications from balloon-borne measurements that the background sulfate load of the stratosphere is increasing due to upwelling of sulfur species from the troposphere. Since the optical depth of this aerosol is relatively small, however, this increase is not expected to exert a significant climate forcing.

Organic species can contribute strongly to the aerosol mass in the polluted and the background troposphere. The organic mass fraction is roughly half of the sulfate fraction. This includes condensed hydrocarbons and living and dead cells or cell fragments. A fraction of 80% to 90% of this aerosol may be primary, i.e., emitted as particles; up to 20% of these aerosols are biogenic. The organic mass fraction of tropospheric aerosols appears to vary strongly regionally. Furthermore, the contribution of organic aerosols to CCN number concentrations may be substantial.

Deserts are the predominant sources of airborne mineral dust; adjacent semiarid areas subject to land-use change can also yield anthropogenic mineral aerosols, although observational evidence is lacking. Measurements have shown that submicrometer dust particles can be transported over thousands of kilometers; moreover, the optical depths of these dust clouds can be considerable. Further, absorption of shortwave solar radiation by windblown dust is expected to be important. Although usually neglected in radiative transfer calculations, additional absorption and emission of infrared radiation by coarse-mode particles may be significant.

Quantities that contribute most strongly to the uncertainties in the estimates of the direct climate forcing of anthropogenic sulfate aerosols are the fraction of SO_2 oxidized to sulfate (~50%) and the mean sulfate residence time in the atmosphere (3–7 days); estimated uncertainty ranges for both quantities are about a factor of 1.5. The role of multiphase chemistry is poorly quantified. Also, emission estimates from non-OECD countries are relatively uncertain (e.g., fuel sulfur content). These uncertainties need to be reduced through measurements, inside and outside the most polluted regions in Europe and North America. Since the oxidation and lifetime of sulfur species can only be quantified through the use of transport/chemistry models, the models need to be improved and validated carefully against obervational data on regional and global scales.

The aerosol optical properties and their capability to form cloud droplets are, to a large extent, determined by how the particles are formed and how they grow by deposition of gases and by coagulation. These processes are complex and not well

quantified; they are likely to be different for different aerosol types, e.g., sulfate and biomass-burning aerosols. An uncertain but important factor is cloud "processing" of aerosol particles. A controlling factor in the nucleation of new particles is the absence of surface area of preexisting particles, so that aerosol precursors that are newly produced by chemical processes are not directly scavenged. Bi-molecular particle formation is relatively well understood, but tri-molecular particle formation is not. Favorable conditions for nucleation often occur in the free troposphere.

The particle-scattering cross section is strongly influenced by deliquescence, so that the aerosol hygroscopic growth characteristics need to be understood and quantified. The hygroscopicity of particles affects their processing by clouds. Thus, this is related also to the indirect aerosol forcing. The aerosol physical and chemical properties are strongly dependent upon whether they are internally or externally mixed. Measurements, although scarce, indicate that most aerosols are at least partly externally mixed. A comprehensive theory for this is still lacking.

Evaluation of aerosol data sets that have been established in the past can be of great use in understanding how aerosol chemical compositions have changed in time and what the consequences are for aerosol radiative forcing. We recommend reevaluating existing data sets and returning to sites where key aerosol optical parameters (e.g., scattering efficiencies) were measured several decades ago, to see if the aerosol optical properties have changed over time.

The modeling of aerosol sources and sinks as well as transport processes on regional and global scales should preferably include explicit calculation of particle size changes as a function of the distance to source regions. Knowledge, however, is still incomplete and computer resources are a limiting factor. Hence, for the time being, models must be relied upon that prescibe aerosol types so that particle optical properties are only functions of the model-calculated particle mass concentration. For the modeling of gas-to-particle formation in global models, we recommend the general use of the trace gas source distributions that will become available in the near future, e.g., from the GEIA acivity.

Because remote-sensing data on tropospheric aerosols will become increasingly available, there is a need to evaluate and reduce the uncertainties of measured aerosol properties. The current development of new multiwavelength satellite instruments will strongly improve the near future capability to derive the aerosol single-scattering albedo and optical depth. The single-scattering albedo is particularly important for assessing aerosol radiative forcing.

The calculation of radiative forcings and climate responses with GCMs will ultimately have to include explicit treatment of aerosol source and sink processes and spectrally resolved radiative transfer calculations. Recent model calculations suggest a significant climate cooling forcing by anthropogenic sulfate particles, which can regionally mask greenhouse forcing by CO_2 and other infrared radiatively active gases. Including sulfate aerosols in GCM calculations indicates a significant increase of the latitudinal gradient of climate forcing in the Northern Hemisphere, which may affect dynamical processes in the atmosphere.

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