Aerosol Forcing of Climate

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Group Report: What Factors Influence Atmospheric Aerosols, How Have They Changed in the Past, and How Might They Change in the Future?

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INTRODUCTION

Assessment of aerosol–climate interactions requires an understanding of the factors that determine the abundances and properties of aerosols in the atmosphere. From the standpoint of direct radiative forcing, critical aerosol properties are the surface size distribution, particle shape, and complex refractive index. From the standpoint of indirect radiative forcing (effects on cloud microphysics and atmospheric chemistry), critical aerosol properties are the number size distribution, hygroscopicity, phase, and chemical composition. In this report, we assess our current ability to parameterize the sources, transformations, and removal of aerosols in climate models, and list some major gaps that need to be addressed over the next decade. We then discuss the sensitivity of aerosol concentrations to climatic and socioeconomic changes in the past and in the future.

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AEROSOL SOURCES

Sea Salt

The principal mechanism for emission of sea-salt particles is the bursting of air bubbles at the sea surface. The emission flux is generally parameterized as a function of surface wind speed. Estimates of the flux in the literature vary over a wide range, in part because of differences in the accounting of large particles (> 10 μm), and in part because of differences in the altitude at which the flux is measured (the very large particles tend to settle quickly). So far, attention has focused on measuring the mass flux; however, the number flux (which is dominated by submicrometer particles) is far more important in terms of indirect radiative effects. Organic films and dissolved organic matter may affect the formation and bursting of bubbles, but the magnitude of this effect has not been quantified. Monomolecular organic films break up at wind speeds higher than a few m s^{-1} and are therefore probably not critical for the conditions leading to high emission.

One outstanding question is:

- What is the size-dependent emission flux of submicrometer sea-salt particles, and how does it depend on wind speed and other surface variables?

Mineral Dust

Mineral dust makes a major contribution to the aerosol optical depth. It scatters shortwave and absorbs both shortwave and longwave radiation. The primary sources are the arid and semiarid regions of the world, which account for about 15% of the global land surface (Duce, this volume). Far from source regions, the mass median diameter of mineral particles is generally 1–3 μm, with larger sizes (up to several hundred μm) observed closer to the source. Submicrometer dust particles have been observed in the atmosphere but have so far received little study; they represent a small fraction of the mineral aerosol mass but a significant fraction of the number. The hygroscopicity of mineral dust varies over a wide range, depending on the type of minerals present and the degree of coating with soluble material.

Parameterizations of dust emission are generally expressed as functions of wind speed, soil texture, and soil moisture; the flux is highly variable and a nonlinear function of wind speed. Changes in land type and land use could strongly affect the source of mineral dust; however, this effect has yet to be studied quantitatively.

Three outstanding questions are:

- What is the abundance, source, and hygroscopicity of submicrometer dust particles?
- How has anthropogenic activity affected the emission of mineral dust?
- What are future prospects for changes in land use, and how will they affect the emission of dust?

Carbonaceous Material

Soot, a product of incomplete combustion, is a major aerosol absorber of visible and infrared radiation. Emission fluxes can be estimated on the basis of economic data for fuel combustion and emission factors per unit of fuel used. The emission factors depend highly on fuel use, being in particular 100 times larger for domestic than for industrial coal burning. Future trends in soot emissions will be determined by trends in fuel use and biomass burning, and by efforts to control emissions.

Four outstanding questions are:

- What is the magnitude of soot emission from domestic fuel use in various regions of the world?
- What is the magnitude and the geographical distribution of the biomass burning source?
- What is the hygroscopicity of soot, and how does it affect the atmospheric lifetime of soot particles?
- What is the size distribution of soot emissions?

Organic particles other than soot make a large contribution to aerosol mass in both polluted and remote regions (Penner, this volume). Preliminary observations indicate that the hygroscopicity of organic aerosol particles varies over a wide range and that a significant fraction is highly hygroscopic. Some of the organic aerosol absorbs light in the visible spectrum and is therefore of particular importance for radiative transfer.

The origin of the organic aerosol is poorly understood. Measurements of the size distribution suggest that a significant fraction is submicrometer and secondary in character (i.e., produced by condensation in the atmosphere). Observed $^{13}C/^{12}C$ ratios over the oceans indicate a dominant continental origin. Biomass burning in the Tropics could provide a major global source of organic carbon aerosol, although measurements over the tropical continents during the wet season indicate also a significant biogenic source. Data collected in the United States show evidence of a major anthropogenic component (Penner, this volume), but measurements in Greenland ice cores do not show a pronounced rise of organic carbon over the past century (as is observed, for example, for sulfate). The Greenland measurements point instead to a large source from boreal forest fires evidenced by correlations with soot, ammonium, and organic acid concentrations.

Three outstanding questions are:

- What is the abundance, hygroscopicity, and speciation of the organic aerosol?
- What are the magnitudes of natural and anthropogenic sources?
- How is human influence affecting organic aerosol concentrations in various regions of the world?

Sulfate

Sulfate aerosol reflects visible radiation and is regarded as a major contributor to aerosol radiative forcing. Over half of the global sulfur source is SO_2 emitted from fuel combustion and industrial activities. Although this anthropogenic source is relatively

well defined, there remains a large uncertainty as to the fraction of SO_2 removed by dry deposition versus that oxidized to sulfate. A 50% yield of sulfate from emitted SO_2 is usually assumed, based on estimates of the relative rates of SO_2 deposition and oxidation, but uncertainties are large. Important sources of uncertainty are the rates of oxidation on dry aerosol surfaces and in aqueous aerosols.

Emission of dimethylsulfide (DMS) from marine phytoplankton is thought to account for 15%–30% of the global sulfur source and may dominate the production of sulfate over much of the ocean surface. The emission flux is generally parameterized as the product of the seawater concentration of DMS and a sea-to-air transfer velocity dependent principally on surface wind speed. There is a need for a model that can predict DMS levels in surface seawater as a function of geographical position and time of year, using data derived from remote-sensing observations; so far it has proven difficult to relate DMS to biological or physical variables. Correlation between DMS and chlorophyll has been observed in some regions but found to be absent in others. Uncertainties in the global and regional estimates of DMS fluxes are a factor of 2–3 and may not be easily improved in the near future. Eddy correlation methods for direct measurement of DMS ftuxes are still in their infancy, since it is difficult to measure concentrations with the precision required.

The oxidation of DMS in the marine atmosphere is not well understood. The standard view is that the OH radical provides the main oxidant; however, the relative diel variation of DMS concentrations in the marine boundary layer is often about twice what can be explained from OH concentrations computed by photochemical models. This implies either unexpectedly large errors in the computed OH concentrations or the presence of additional oxidants such as Cl atoms or BrO.

A number of nonradical sulfur species are produced from DMS oxidation including SO_2, sulfate, methanesulfonic acid (MSA), dimethylsulfoxide (DMSO), and dimethylsulfone ($DMSO_2$). Product yields are highly uncertain, particularly under the low–NO_X conditions observed over most of the world's oceans. This uncertainty is a serious hindrance for models of aerosol dynamics.

Estimates of global SO_2 emission from volcanoes vary over a wide range, up to 20% of the global source. Some of the variability reflects differences in the eruption statistics, while some reflects differences in the SO_2 emission factor. The importance of volcanoes as an episodic source of sulfate to the stratosphere is well established, but the effect on tropospheric sulfate and subsequent radiative forcing is less certain. Recent observations over the north Pacific Ocean suggest that a significant fraction of SO_2 in the free troposphere could be of volcanic origin.

Three outstanding questions are:

- What is the DMS emission flux from the oceans, and how can it be parameterized in global models?
- What is the mechanism for oxidation of DMS to sulfate?
- What are the important mechanisms involved in the oxidation of SO_2 to sulfate in aerosols?

Ammonium

Ammonia is an important component of the secondary aerosol particles, partly or totally neutralizing H_2SO_4. The presence of NH_3 raises the deliquescence point of acidic sulfate aerosol and may also facilitate the production of sulfate by increasing the pH of atmospheric liquid water (cloud droplets, water films on aerosol particles). There is some laboratory evidence that NH_3 facilitates the formation of new sulfate particles.

Sources of NH_3 to the atmosphere include livestock, biomass burning, and biological activity in the soils and in the oceans. None of these sources is sufficiently quantified. Soils and oceans may act as sinks as well as sources. Better understanding of the oceanic source is particularly critical because of its effect on the sulfate aerosol in the marine boundary layer; this aerosol is generally found to have an approximate NH_4HSO_4 stoichiometry.

Two outstanding questions are:

- What are the magnitudes and geographical distributions of NH_3 sources and sinks?
- What is the origin of NH_3 in the marine boundary layer?

Nitrate

Most of the aerosol nitrate in remote regions is present in or on supermicrometer particles, reflecting the deposition of HNO_3 on dust and sea-salt aerosol. This nitrate makes only a small contribution to the mass of the supermicrometer aerosol but may affect its hygroscopicity. Submicrometer aerosol nitrate arising from the reaction of HNO_3 and NH_3 represents a significant fraction of the aerosol in polluted regions where combustion sources of NO_x provide a large source of HNO_3 and where excess NH_3 is available beyond the fraction consumed by the neutralization of H_2SO_4. In such regions in the U.S. and OECD countries, it is expected that the relative importance of nitrate will increase over the next decades as sulfur emissions decrease. Emission factors for NO_x from fossil-fuel combustion are reasonably well defined, at least in the developed countries.

One outstanding question is:

- How important are aerosol reactions of NO_3 and N_2O_5 for oxidizing NO_x to HNO_3 in polluted regions?

FORMATION, MODIFICATION, AND REMOVAL OF AEROSOLS

Formation

One possibly major source of new particles is the direct emission of submicrometer particles from sea salt and soil dust; however, the magnitude of this source is unknown.

The other major source of new particles is the chemical transformation of gaseous precursors into condensable material and subsequent nucleation. It is generally believed that H_2SO_4 is the dominant precursor of new particles, but this has not been demonstrated. The role of NH_3 in enhancing nucleation of sulfate is not understood. Nucleation of organic material could be significant.

The relative importance of nucleation in the free troposphere versus in the boundary layer is a matter of debate (Raes et al., this volume). Recent measurements in the upper troposphere over the Pacific Ocean show evidence for considerable new particle formation, as indicated by ubiquitously high total number concentrations (> 10,000 cm^{-3} for diameters > 3 nm). These concentrations are far higher than typical values in the Pacific marine boundary layer. According to some models, entrainment from aloft could provide the main supply of new particles in the boundary layer; however, observed correlations between DMS fluxes and particle number concentrations in marine surface air support the hypothesis that significant new particle formation also takes place from DMS oxidation within the marine boundary layer. Present models show that only a few percent of emitted DMS would be needed to account for the observed aerosol number concentration in the marine boundary layer, but these models fail to describe the observed particle growth rates.

Four outstanding questions are:

- What is the importance of primary emission of submicrometer particles as a source of new particles?
- What is the importance of multicomponent mixtures (e.g., H_2SO_4–NH_3–H_2O) in facilitating aerosol nucleation?
- Can organic species initiate new particle formation?
- To what extent do sea-salt particles affect the production of new aerosol particles in the marine boundary layer? In particular, how does aqueous-phase oxidation of SO_2 in alkaline sea-salt aerosol suppress new particle formation?

Modification

Aerosol particles undergo considerable physical and chemical transformations during their lifetime (one to three weeks). Ultrafine particles grow into the Aitken mode range (20–100 nm diameter), and Aitken mode particles are transformed into the accumulation mode range (100–1000 nm diameter). The transformation of ultrafine particles into the Aitken mode may be regulated mainly by heterogeneous condensation and aqueous-phase reactions; investigations of the chemical composition and hygroscopicity of Aitken particles would afford a better understanding of this issue. The transition of Aitken particles into the accumulation mode appears to require growth by chemical reactions during cloud processing, e.g., aqueous-phase oxidation of SO_2. Better documentation of this phenomenon is needed. Observations in various regions show that the aerosol is partitioned into distinct hygroscopic and hydrophobic fractions, and this factor should strongly affect aerosol growth.

Aerosol production and growth mechanisms in polluted environments are different from those in clean environments. Little is known about the evolution of aerosols from industry and biomass burning once they have left the source area, i.e., how much growth and chemical change take place during long range transport. Development of parameterizations for nucleation and growth of secondary aerosols in global models is a difficult task because of the overlap in time scales between aerosol dynamics and synoptic meteorological processes (Raes et al., this volume). For the near future, the best approach is to use simplified interactive aerosol dynamics models that resolve number, mass, and other integral properties of the aerosol size distribution, combined with empirical correlations between these integrals and the optical or nucleating properties of the aerosol.

Three outstanding questions are:

- What is the role of cloud processing in modifying the size distribution of the aerosol?
- What are the time scales for aerosol growth? To what extent is the aerosol distribution determined within the source region, and how much does it change during long-range transport?
- What determines the partitioning of the aerosol into hygroscopic and hydrophobic fractions, and how does this partitioning affect aerosol growth?

Removal

Aerosol removal is achieved through coagulation (reducing number while conserving mass) and deposition (reducing number and mass). Coagulation appears to be especially important in decreasing the number of ultrafine particles that grow to the Aitken mode. Deposition of submicrometer particles is mainly through precipitation and is generally parameterized in global models using first-order loss rate constants normalized to precipitation intensity. Proper parameterization of this loss is difficult because the efficiency of aerosol scavenging in cloud (rainout) is sensitive to the abundance and the hygroscopicity of the aerosol, while the efficiency of aerosol scavenging below cloud (washout) is sensitive to the aerosol size distribution. In middle and high latitudes, much of the precipitation and hence aerosol scavenging is due to ice crystals; only a tiny fraction of the aerosol population is involved in ice crystal formation, yet this fraction determines strongly the efficiency of aerosol scavenging. The sources, concentrations, and temperature-activity dependences of ice nuclei and their relationship to the formation of ice crystals in the cloud are poorly known.

Deposition of supermicrometer particles is driven not only by wet but also by dry processes (impaction, sedimentation). The dry deposition flux is strongly sensitive to the upper end of the size distribution and is therefore not easily quantified.

Two outstanding questions are:

- What processes control the formation of ice crystals in cold clouds?
- How can aerosol scavenging from cold clouds be parameterized?

PAST RECORDS OF AEROSOL CONCENTRATIONS

Ice cores in Greenland and Antarctica provide surrogate archives of aerosol concentrations at high latitudes (Legrand, this volume). The sulfate record for Greenland indicates a factor of 4 increase in deposition from preindustrial times to the 1970s, and a factor of 2 decrease since then. The decrease since the 1970s is greater than one would expect from the overall decline of anthropogenic emissions at northern mid-latitudes, and may reflect a disproportionate decline of emissions in regions that directly affect the Greenland ice cap (e.g., smelters in Siberia). In contrast to Greenland, no significant rise of sulfate over the past 100 years is observable for Antarctica. The Antarctic data indicate an increase of MSA deposition, a trend that is not seen in Greenland. The relationship between MSA deposition and climate remains an open question; deposition of MSA is a function of many factors which are poorly understood (transport from source regions, wind speed at the sea surface, sea ice modification, biological activity).

Low ammonium concentrations in Antarctic ice suggest that a large fraction of sulfate deposited at high southern latitudes is only partly neutralized by ammonia. Ammonium concentrations in Greenland ice cores do not show an increase over the past century that could be attributed to anthropogenic emissions. If anything, concentrations appear to have decreased slightly over that period. Peaks in the historical record appear to be associated with boreal forest fires, as indicated by their correlation with soot.

Dust concentrations in ice cores indicate considerably higher values in glacial times than today (30–50 times higher in both Greenland and Antarctica). The cause is not well understood; contributing factors could include changes in wind speed over continental areas, aridity, glacial outwash, and exposure of continental shelves in response to sea surface lowering. Records of sea salt in both Greenland and Antarctica indicate concentrations five times higher in glacial times than today in spite of extensive sea-ice cover; this suggests changes in surface wind speed over the ocean or changes in the efficiency of meridional transport. General circulation model simulations of paleoclimates have, in general, been unsuccessful at reproducing the magnitudes of the dust and sea-salt enhancements in glacial times.

Preliminary data for Greenland ice cores show no clear evidence of a rise in carbonaceous aerosol concentrations over the past century (Legrand, this volume). The rise, if present, is less than that observed for sulfate. Peaks in the record appear to be due to boreal forest fires. More specific investigations of organic components (polycyclic aromatic hydrocarbons, dissolved organic species) could be of great use for assessing human influence.

Improved interpretation of the ice core records requires further investigation of the link between the concentrations found in the snow and the concentrations in the atmospheric column overhead, and the link between the atmospheric column overhead and the large-scale circulation. Interpretation of records for sulfate and MSA requires particular care in distinguishing between polar and mid-latitude sources.

Additional information on soot and dust concentrations in the past can be obtained from lake and deep-sea sediments. Temporal resolution, however, is less than for ice cores (thousands of years for deep-sea sediments).

FUTURE TRENDS

Future emission scenarios can be developed on the basis of emission inventories for present-day conditions by using forecasts of trends for socioeconomic factors, land use, and climate (Hidy, this volume). Socioeconomic projections for the next fifty years suggest that worldwide emissions of aerosols and aerosol precursors will continue to increase unless strong countermeasures are taken. Sulfate will remain a major constituent of the aerosol. Anthropogenic emissions are likely to be dominated by energy use (particularly coal combustion in developing nations), biomass burning, and production of cement; these emissions will disproportionately affect the submicrometer fraction of the size distribution. The geographical distribution of emissions will shift to the developing countries due to increasing emission controls for SO_2, NO_x, and particulate matter in the developed world. Increased haziness can be expected over Asia, India, and the western Pacific Ocean. Regional perturbations to radiation fluxes resulting from haze occurrence in combination with greenhouse-gas accumulation may create systematic thermal gradients influencing atmospheric processes.

General circulation model (GCM) forecasts of climate change in the 21st century have focused so far on mean temperature and precipitation. While all GCMs forecast increases in temperature and absolute humidity, and decreases in sea ice cover, the magnitudes of the changes vary among the models and will be modified by inclusion of aerosol effects (Roeckner et al., this volume). Predictions of mean precipitation changes are uncertain, with increases in a few models and decreases in others. Forecasts of aerosol loading depend on additional environmental variables that are available or derivable from GCM calculations but are not routinely scrutinized. These include the variance of temperature, the intensity and frequency of precipitation, the relative humidity, the surface wind speed, gustiness, the frequency of lightning, and changes in biomass.

To predict future trends in the atmospheric loading of aerosol as a result of global change, we must first be able to quantify the sensitivity of the aerosol loading for individual species (y) to changes in the processes or factors (x_i) that produce, modify, and remove the aerosol from the atmosphere. We can express the future loading $y(t_o + \Delta t)$ in terms of the present-day loading $y(t_o)$:

$$y(t_o + \Delta t) \simeq y(t_o) + \left.\frac{\mathrm{dy}}{\mathrm{dt}}\right|_{(t_o)} \Delta t \,. \tag{10.1}$$

The total derivative $\mathrm{d}y/\mathrm{d}t$ can be expressed as a sum of partial derivatives:

$$\frac{dy}{dt} = \sum_i \frac{\partial y}{\partial x_i} \frac{\partial x_i}{\partial t} . \tag{10.2}$$

In Table 10.1, we estimate the sign of $\partial y/\partial x_i$, the direction of change in aerosol loading as a result of an increase in factor x_i, as well as the direction of the anticipated change of x_i in the 21st century. Table 10.1 is only a first attempt at assessment; in reality, the effects of each factor cannot be viewed in isolation as the system is inherently nonlinear. We see from Table 10.1 that individual factors of global change have widely different effects on the burdens of specific aerosol components. Some important features are discussed below.

Mineral Dust

Increased temperature leads to enhanced drying of soils and hence higher dust emission. Higher winds and gustiness also increase dust emission. Increases in relative humidity raise the soil moisture and favor the growth of vegetation cover which stabilizes soils. Increased precipitation decreases soil dust both because of increase in soil moisture and because of more efficient aerosol scavenging. Agriculture and deforestation tend to increase wind erosion. Industrial activity increases dust emissions but is a relatively minor source.

Conclusion: The rate and extent of dust generation appear to be quite sensitive to global change and are especially dependent on land-use factors and soil conservation practices.

Sea Salt

Sea-salt emission increases with wind speed and gustiness. Removal of sea-salt aerosol should increase with relative humidity (because of increased aerosol size) but the effect in the marine boundary layer is expected to be small.

Conclusion: Sea-salt aerosol is relatively insensitive to climate change variables.

Carbonaceous Aerosol

Higher temperatures may be expected to increase the source of natural soot emissions because of greater susceptibility to fire, and should also stimulate the emission of organics from both natural vegetation and crops. However, the source of anthropogenic soot might decrease because of reduced need for heating fuels. Increases in wind speed and gustiness increase drying of vegetation and enhance the spread of fire; they also lead to the emission of more primary organic particles through the shaking of plants and the release of plant debris. Higher winds facilitate the removal of anthropogenic soot by dry deposition, but the effect should be small since most of the removal is by wet deposition.

Table 10.1 Speculated sensitivity of aerosol loading to climate change variables.

Aerosol Type	Loading Sensitivity $\left(\frac{\partial y}{\partial x_i}\right)$															Net Effect in 21st Century
Mineral Dust	+	+	+	+	–	–	+	0	(–)	?	+	–	+	+	(+)	↑*
Sea Salt	0	0	+	+	(–)	–	+	0	(+)	–	0	0	0	0	0	→
Soot:																
Natural	+	+	+	+	–	–	+	+	0	0	0	+	–	+	0	↑*
Anthro.	–	+	(–)	?	(–)	–	+	0	0	0	0	0	+	+	+	↑*
Organic:																
Natural	+	?	+	+	?	?	?	+	(–)	(–)	0	+	–	+	0	↑
Anthro.	+	?	(–)	?	?	?	?	0	0	0	0	0	+	+	+	↑
Sulfate																
Natural	?	?	+	+	(–)	–	+	0	(+)	?	+	0	0	(+)	0	→
Anthro.	–	+	(–)	?	?	–	+	0	0	0	0	0	(+)	(+)	+	↑*
Nitrate	?	?	?	?	?	?	?	+	0	0	0	+	+	+	+	↑
Ammonium	+	?	?	?	(+)	?	?	+	(–)	(+)	0	+	+	+	(+)	↑
Climate Variable	temp.	temp. variability	wind speed	wind speed variability	relative humidity	mean precip.	precip. variability	lightning	sea level	sea-ice cover	volcanic eruptions	natural vegetation	agricul-ture	biomass burning	industrial activity	
Expected Change in 21st Century	+	?	?	?	?	?	?	+	+	–	0	?	+	+	+	

( ) = small change
* = particulary bears watching

Increasing humidity reduces the frequency of natural fires and hence soot emission; it also causes soot particles to grow to larger sizes, facilitating removal. Increased precipitation reduces the fire hazard for both natural and agricultural biomass but increases biomass loading which could result in larger and more frequent fires.

Conclusion: The most likely effect of climate change and global development is an increase in the concentrations of carbonaceous aerosols. There are, however, many uncertainties regarding changes in the natural emission of organics, particularly as it depends on the composition of the biosphere.

Sulfate

The effect of temperature on biogenic sulfur emission is unclear. Higher temperatures should reduce the demand for heating fuels and hence anthropogenic SO_2 emissions. Increased fuel use in presently underdeveloped countries will override this decrease. A rise in humidity would facilitate aqueous-phase oxidation of SO_2 in aerosols, but would also increase the size of sulfate particles and thus facilitate deposition. Increase in average precipitation would increase the scavenging of sulfate aerosols but increased variance in precipitation should result in higher sulfate levels because of prolonged dry periods. Biomass burning emits sulfur gases but this is not a major source since the sulfur content of vegetation is relatively low.

Conclusion: Global change over the next century is expected to increase anthropogenic sources of sulfate while having little effect on natural sources.

Ammonium

Rises in temperature and humidity should increase NH_3 emission from soils because of increased microbial activity. Higher winds enhance ventilation of soils and hence emissions, but also increase the rate of deposition to surfaces. Increased precipitation increases soil production of NH_3 but also reduces the lifetime of NH_3 in the atmosphere. Advanced agricultural techniques use high-nitrogen fertilizers which result in the direct release of NH_3 and the indirect increase of NH_3 emissions from soils. One of the major sources of NH_3 is urine from livestock; population growth and a shift to increased meat in diets will result in increased emissions.

Conclusion: Emission of NH_3 will probably increase greatly over the next century.

Nitrate

The sensitivity of nitrate to changes in temperature, wind speed, humidity, and precipitation is uncertain but likely small. Industrial development will likely be the major modifier of nitrate concentrations.

Overall Conclusion

Climate change and population development in the 21st century are expected to cause general increases of atmospheric aerosol concentrations. Trends in mineral dust, soot, and sulfate particularly bear watching.

RECOMMENDED READING

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