Annu. Rev. Environ. Resour. 2004. 29:1–30
doi: 10.1146/annurev.energy.29.063003.132549

First published online as a Review in Advance on June 30, 2004

# CURRENT UNCERTAINTIES IN ASSESSING AEROSOL EFFECTS ON CLIMATE

Surabi Menon
*Lawrence Berkeley National Laboratory, Berkeley, California 94720, and NASA Goddard Institute for Space Studies/Columbia University, New York, NY 10025; email: smenon@lbl.gov*

**Key Words** carbonaceous aerosols, aerosol-cloud-climate interactions, climate change, climate predictions

■ **Abstract** The effect of anthropogenic emissions from activities, such as fossil-fuel, biomass, and biofuel burning; transportation; and land-clearing; have a profound impact on the climate system. The impact of these activities is manifested in observed changes in temperature, precipitation, sea-level rise, melting of glaciers, air quality, health, and agriculture yields, to name a few. The obvious question to ask is the role that these different processes play in affecting climate and what action could one impose to curtail or constrain adverse human impacts on climate. Greenhouse gases have long been studied, as they play a major role in changing climate. But over the past 10–20 years, aerosols have emerged as the other big contenders in climate change studies. This review focuses on the current understanding of the effects of aerosols on climate, with an emphasis on the thermodynamical and indirect aerosol effects. We also examine available measurements that could be used to decipher the aerosol influence on climate, with an outlook on how the uncertainties in aerosol effects may impact future climate predictions and policy changes.

CONTENTS

## 1. INTRODUCTION

Human-influenced activity, such as burning of fossil fuel, biofuel, and biomass; land-cover changes; industrial activity; and transportation, can impact climate and may even exceed natural fluctuations within the climate system. However, the exact magnitude of these anthropogenic influences is quite uncertain. Although climate change estimates over the past million years have to be considered to place present-day impacts in context, most reliable estimates of climate change reported are for the past 150 years (the time from the preindustrial revolution to present day). This time period is of relevance because the warmth of the twentieth century appears to be unprecedented during the millennium (1), with several recent years reported to be the warmest on record for the late twentieth century (2). To reconcile temperature trends observed over the past 150 years, which indicate an increase of $\sim 0.6 \pm 0.2$ K, considerable attention was paid to the role of greenhouse gases [which has increased by $\sim$30% since 1850 and continues to increase at $\sim$0.5% to 1% per year over the past few decades (3)] in modifying climate. However, existing discrepancies between model projections of temperature change and surface observations suggest that other factors or processes may also exert significant influence. Solar and volcanic activity no doubt play a major role in influencing climate change over the past millennium, but over the industrial period, $CO_2$ effects, land-cover changes, and deforestation have become increasingly important (4). Over the past 20 years, however, more attention has been focused on the role of aerosols and aerosol-induced cloud changes as important contenders in the modification of the radiation balance of the Earth-atmosphere system and therefore in climate change scenarios.

Greenhouse gases, ozone, and black carbon (BC) aerosols are usually associated with global warming, whereas aerosols, such as sulfates and organic carbon (OC), and their interaction with clouds result in global cooling. Whereas global warming effects from greenhouse gases and ozone can be quantified fairly well, the magnitude of the effects of aerosols on climate still has a large uncertainty associated with it. The evaluation of the change in cloud properties, both microphysical and radiative, due to the interaction between aerosol particles and clouds presents one of the largest uncertainties in the quantification of the aerosol effects on climate. A large component of the uncertainty is due to the aerosols—sources (biomass, biofuel, and fossil fuel derived) and distribution. To predict the response of the climate to the various imposed forcings and to evaluate the changes in the forcings brought about by policy or emission changes, the primary tools relied on are models. However, model results may be subject to large uncertainties, and it is thus essential that climate change projections be presented more credibly by carefully identifying and quantifying aerosol effects on climate.

Comparing recent projections of greenhouse gas-induced changes ($2.5 \pm 0.5\ W\ m^{-2}$) with recent projections of aerosol-induced changes (0.5 to $-4\ W\ m^{-2}$), and the change in temperatures since the preindustrial era to present ($0.6 \pm 0.2$) (1) certainly lends credence to the argument that the uncertainty in quantifying the aerosol effect could be a key issue in predicting future temperature changes. Several approaches have been used to estimate the influence of aerosols on climate observationally and theoretically. The impact of volcanic aerosols on temperature records is one example of the effect of aerosols on climate. Several other examples in the past decade (modification of cloud properties in ship tracks, suppression of rainfall from satellite records in polluted air masses, etc.) suggest mounting evidence exists that aerosols are affecting climate in a significant way. Precipitation changes are easier to notice than are temperature changes, especially in arid regions. If aerosols do indeed alter circulation and precipitation patterns, as shown by some recent studies (5–10), it becomes essential for policy-making initiatives to take into account the role of aerosols on climate. To aid policy decisions, credible results that identify, quantify, and reduce the uncertainties in deciphering aerosol climate effects are needed. This review thus focuses on certain key processes that link aerosol effects to climate (primarily those related to the thermodynamical and hydrological processes), availability of related measurements, current uncertainties, future climate impacts, and policy needs. A more comprehensive review on climate change can be obtained from the Intergovernmental Panel on Climate Change (IPCC) report (1), which provides an assessment of climate change for the past, present, and future and is commonly used as a standard reference to understand climate change and its impacts.

## 2. AEROSOL SOURCES AND DISTRIBUTIONS

Aerosols of interest include sulfates, carbonaceous aerosols (OC and BC), nitrates, sea salt, and dust. Table 1 lists the primary aerosol sources and the range in atmospheric burdens compiled from different models (1, 11) from emission sources. Although carbonaceous aerosols from biofuel burning are prevalent in countries such as India, biofuel emissions are usually not separated from biomass or fossil fuel emissions. This issue will have to be addressed in future emission inventories. The wide range in atmospheric burdens of these aerosols can result in a wide range of forcing estimates that are not related linearly. To narrow the range in forcing estimates would require tighter constraints be placed on the source and distribution of aerosols. This can be quite formidable due to the complexities in quantifying emission sources and in determining the transformation of emissions to particles.

Carbonaceous (OC and BC) aerosols come from similar sources (incomplete combustion from biomass, biofuel, fossil fuel, and dung burning; emissions from diesel engines; etc.) but have different optical properties. OC aerosols are mostly hydrophilic and reflective except for some absorption in the ultra violet (UV) and visible region (12). BC aerosols are hydrophobic but can be hydrophilic as BC particles get coated with other particles as they age. However, unlike OC, BC is

**TABLE 1** Range in estimated source strength (Tg aerosol year$^{-1}$) for present-day aerosols (1, 11)

| Type | Source strength |
|---|---|
| Sulfate | |
| Industrial | 65–92.4 |
| Ocean | 10.7–23.7 |
| Aircraft | 0.04 |
| Biomass burning | 2.0–3.0 |
| Organic carbon | |
| Fossil fuel | 10–20 |
| Biomass | 30–45 |
| Black carbon | |
| Fossil fuel | 5.8–6.6 |
| Biomass | 6.0–17.2 |
| Nitrates | |
| Fossil fuel | 0.3 |
| Biomass | 5.7 |
| Other (human, soils, animal, agriculture) | 74.5 |
| Sea salt | |
| $<2$ $\mu$m | 82 |
| $>2$ $\mu$m | 2583 |
| Dust | |
| $<2$ $\mu$m | 243 |
| $>2$ $\mu$m | 4859 |

mostly absorbing in the visible and UV region. It is this absorbing property of BC and its effectiveness in warming the atmosphere that could potentially offset, by a small amount, the cooling caused by the other aerosols. Although BC particles are estimated to have a small total global forcing effect of 0.4 to 1 W m$^{-2}$ (13–16), their efficacy as a forcing agent can be high mainly due to their effects on snow albedo (17). Because BC aerosols can affect human health, agricultural yields, precipitation, temperature, and large-scale circulation through thermodynamical, radiative, and microphysical processes, their influence on regional and global climate can be quite significant, especially in areas where BC concentrations are high.

Several techniques and methods are used to detect trends in aerosol burdens. Deposition of aerosols on the surface serves as a useful proxy that can be used to extrapolate information for local aerosol loadings. Ice-core sampling, lake bed sediments, etc. present some evidence of the amount of aerosols present several centuries ago, though these are mainly useful to infer local trends. Estimating $SO_2$ emissions is relatively straightforward because these depend mostly on the mass of fuel burned and its sulfur content. However, obtaining estimates of BC emissions

can be much more complicated. Being a product of incomplete combustion, the amount of BC emitted depends on how efficiently the fuel is burned in addition to the fuel amount. Thus, the mass of BC emitted per unit weight of fuel (emission factor) depends on fuel type (i.e., coal, diesel, and biomass) and other factors that influence the combustion efficiency. This makes BC emissions technology dependent. As an example, coal burned in inefficient stoves and furnaces produces orders of magnitude more BC than the same fuel burned in modern electric power plants. Similarly, coal use for space heating by individual household stoves results in much greater BC emissions than central heating. In industrialized countries during the past half century, moving coal consumption from inefficient to more efficient combustion was the main cause of decreasing BC emissions. Another example of technology effect on emissions is improvement in the diesel engine technology. Emissions from this source have been significantly reduced in many western countries as a result of improved engine design.

Thus, within the past 50 years, such changes in technology in the developed world have led to large reductions in aerosols emitted as compared to emissions in the developing world (18). Transfer of technology from the developed world to the developing world could lead to changes in future emissions that may be more difficult to quantify, thus possibly leading to some discrepancies when estimating changes in aerosol sources for future projections.

Once aerosol emissions are known, changes in particle radii, single scattering albedo as a result of aging, type of fuel burned, and combustion phase need to be quantified carefully (19) to estimate climate forcing. A multimeasurement approach from several platforms (satellites, laboratory, field observations) may serve to constrain the uncertainty associated with aerosol properties—physical and optical. Satellite-based retrievals that distinguish between fine and coarse particles can separate dust from pollution or smoke aerosols (20). Laboratory-based analysis of isotopic composition can also be used to gather information regarding the natural or anthropogenic origin of particles. To estimate past BC fossil fuel emissions, Novakov et al. (18) used annual consumption data for principal BC-producing fossil fuels and BC emission factors separated by source sectors: industrial, residential/commercial, power generation for coal, and transport for diesel fuel. These data sets, obtained for several countries and the rest of the world, were used to provide a time series of temporal trends in BC amounts from 1875 to 2000. The data show the increase from 1875 to 1950 for industrialized countries and their gradual decline in the past few decades in contrast to the data for developing countries (China and India), which indicate a general increase since l950. Calculated single scattering albedos from Novakov et al. (18) agreed reasonably well with recent observations, and these values can be used to obtain an approximate measure of the degree of cooling or warming caused by aerosols.

## 3. ESTIMATES OF AEROSOL EFFECTS ON CLIMATE

Both aerosols and greenhouse gases modulate the energy and hydrological cycle. In general, greenhouse gases lead to warmer and wetter climates, whereas sulfates and

OC tend to produce cooler and drier climates (21). Being absorbing, BC aerosols cause warmer temperatures (16), and their vertical distribution can affect the magnitude and sign of the surface temperature change and rainfall patterns through thermodynamical changes. Here, we examine the processes—direct, semi-direct, thermodynamical, and indirect—through which aerosols affect climate, paying more emphasis to the latter three, which are more uncertain. We also show some new results from the Goddard Institute for Space Studies (GISS) climate model that relate to aerosol vertical distributions and its impact on the thermodynamical process.

## 3.1. Direct Effect

The direct effect of aerosols was studied extensively in the late 1980s and early 1990s. This effect refers to the direct scattering or absorption of radiation by aerosol particles without the modification of cloud properties. From the optical characteristics (asymmetry parameter, optical depth, single scatter albedo, etc.) of the aerosols, their size distribution, and changes due to relative humidity effects, the radiative fluxes can be computed. This is relatively uncomplicated, although assumptions such as relative humidity effects on size, sphericity of the particles, transformation of the optical properties for aerosols due to it being internally mixed with other aerosols, vertical location of the aerosols, and the presence of clouds above or below the aerosol layer can affect forcing estimates. Among these, perhaps one of the greatest uncertainties is the increased absorption efficiency of BC when located inside a scattering particle (15, 22), which can change the single scattering albedo of the particle and thereby the sign of the aerosol forcing (18, 23).

Uncertainties in quantifying aerosol emissions and tranformation of emissions to atmospheric burdens, assumptions used in characterizing the physical properties of the aerosols, and the resulting optical properties can each affect estimates of direct forcing calculated by different models. The range of uncertainties associated with all these processes is not well known and hence cannot be characterized accurately. Nakajima et al. (24) found that the direct forcing estimates could vary as much as 40% depending on the optical depth and single scattering albedo used in the radiative transfer code. Their results were based on model simulations, surface radiation measurements, and satellite remote sensing. Variance in the aerosol size distribution was also thought to contribute to a 65% uncertainty in forcing estimates (1). These uncertainties produce wide ranges of forcings estimated by different models depending on the assumptions chosen.

Rather than compare the direct forcing estimates computed by different models, we compare the forcing efficiencies for aerosols, expressed as the ratio of forcing (W $m^{-2}$) to anthropogenic burden (g $m^{-2}$), which are given in Haywood & Boucher (11) for several models. These values can be used to estimate the efficacy [effectiveness of a forcing in producing a climate response (16)] of aerosols and thereby the changes that can be expected from controls on emissions of certain aerosol particles.

Forcing efficiency values for sulfate aerosols range from $-125$ to $-460$ W $g^{-1}$.

(The direct forcing values range from $-0.29$ to $-0.82$ W m$^{-2}$ for sulfate column burdens of 1.14 to 3.3 mg m$^{-2}$, with lower burdens producing lower forcing values in general.) One can expect that the values for OC are similar to that for sulfates because they are both mainly reflective in the visible spectrum. However, for fossil fuel BC aerosols, forcing efficiencies vary from 1123 to 3000 W g$^{-1}$. (The direct forcing for fossil fuel BC ranges from 0.17 to 0.42 W m$^{-2}$ for column burdens of 0.13 to 0.16 mg m$^{-2}$.) Separate model estimates are not available for BC alone from biomass burning because these also include OC. The forcing efficiency for both BC and OC from biomass burning ranges from $-80$ to $-210$ W g$^{-1}$. (The direct forcing ranges from $-0.14$ to -0.74 W m$^{-2}$ for column burdens of 1.76 to 3.7 mg m$^{-2}$.) Clearly, the forcing efficiency for BC surpasses that of sulfate and OC by several orders of magnitude.

Thus, it is not the abundance of the aerosols but their efficacy that provides an estimate of the change in forcing for given aerosol burdens. Sato et al. (23) find that climate forcing from BC, regardless of its mixing state, can be $\sim$1 W m$^{-2}$, most of which is anthropogenic. The authors base their estimates on a climate model with BC climatologies tuned to match the results from retrievals based on AERONET (a network of calibrated sunphotometers distributed globally) optical depths for aerosol absorption. The method used by Sato et al. (23) to validate model BC climatology for two aerosol transport models (based on the information of multiwavelength multiangle measurements from AERONET and Mie scattering calculations, which are used to obtain the spectral characteristics of the aerosols of interest) indicates that model BC climatologies and inferred absorption are lower than measurements by a factor of 2 to 4. Because knowledge of the atmospheric burden of BC as well as its optical properties can be quite uncertain, the methodology presented by the authors provides a rather useful measure to infer the degree of absorption present in the atmosphere and evaluate the ability of climate models to account for that absorption. This can be used to refine model estimates that may be incorrect either due to inadequate representation of physical processes that distribute BC or due to poor emission sources.

## 3.2. Thermodynamical Effects

For absorbing aerosols, changes in surface forcing relative to values of forcing at the top of the atmosphere can have a significant impact on evaporation, latent heating, and the hydrologic cycle. This feeds back to the aerosol distribution due to changes in aerosol scavenging through precipitation. Here, we discuss changes in the monsoon that may be attributed to aerosol effects. We focus on the Asian monsoon because it is a large dynamical system that affects a region with one third of the world's population. There have been several studies that have attempted to quantify precipitation changes for the Asian monsoon from doubled $CO_2$ or increased sulfates. For a doubled $CO_2$ scenario, increased rainfall and greater warming over the Asian continent than over the Indian Ocean was related to increased water content (horizontal transport and precipitation efficiency) rather than to changes in

circulation (25), whereas a slight weakening (related to land-sea contrasts) for the Indian summer monsoon was found if sulfate aerosols were included (first indirect effect only) (26). Based on observational data, Xu (27) linked the southward shift of the Chinese monsoon to the direct radiative effects of sulfate aerosols over China. Observational evidence from the Indian Ocean Experiment (INDOEX) field study by Ramanathan et al. (6, 21) indicate a regional scale warming of approximately 1 K in the lower atmosphere over the tropical Indian Ocean and land surface cooling of approximately 0.5 to 1 K. These changes and redistribution of heat within the boundary layer were thought to have a strong impact on tropical circulation and interhemispheric precipitation patterns, and they were related to the presence of absorbing aerosols over the Indian subcontinent (9). Another study found that the trends in summer precipitation over China—North China droughts, South China floods—considered to be the largest observed in several decades, were related to the increased aerosol emissions observed over East Asia in recent years (10). These trends were attributed to changes in vertical temperature profiles, stability, and vertical velocity fields from absorbing aerosols, which were thought to increase/decrease cloud cover and precipitation in the south/north of China.

Also of relevance are changes in surface fluxes. Meehl (28) suggests that greater forcings over land (due to lowered surface albedo) can enhance land-sea temperature contrasts and result in a stronger monsoon. However, results from Srinivasan (29) indicate that the monsoon can be adequately represented by three parameters, net radiation at the top of the atmosphere, evaporation, and the vertical stability of the atmosphere, without any dependence on land-sea contrasts. These results were based on a simple thermodynamic model. Thus, because the absorptive properties of BC aerosols can influence the net surface radiation budgets (30), and because the surface energy budget is a primary energy source for the Asian summer monsoon (31), accounting for BC aerosol effects on surface fluxes is important.

Since much less is known about aerosol vertical distribution than the horizontal distribution, theoretical investigations to illustrate the importance of aerosol vertical profiles have to be relied on. This has implications for aerosols released from fossil fuel use (mostly confined to the lower boundary layer but may be transported to higher levels owing to deep convection) and from biomass burning (where emissions often reach the midtroposphere). We used model simulations following a similar strategy as Menon et al. (10) wherein climate changes are calculated using the GISS GCM for simulations with aerosols distributed over China and India/Indian Ocean on the basis of inferred measurements compared with simulations with aerosols that have $\sim$0.1 optical depth. For simplicity, a single scattering albedo of 0.85 for aerosols composed of sulfates, organics, and BC was used, as in Menon et al. (10). This simulation is referred to as Exp A. The aerosol vertical distribution is that from Koch (32) (obtained from an aerosol transport model coupled to the GISS GCM). To illustrate the impact of the vertical distribution of aerosols on climate, we chose experimental setups (Exp L1 to L8) in which aerosols were completely confined in a single model layer, beginning with the first and extending up to the eighth model layer, i.e., eight different setups were used.

**TABLE 2** Values of midlevel pressure and height for the different model vertical levels

| Level | Pressure (hPa) | Height (km) |
|---|---|---|
| 1 | 959 | 0.42 |
| 2 | 894 | 1.1 |
| 3 | 787 | 2.1 |
| 4 | 635 | 3.8 |
| 5 | 470 | 6.4 |
| 6 | 338 | 8.4 |
| 7 | 248 | 10.5 |
| 8 | 180 | 12.6 |

Values for pressure and height corresponding to model midlevels for the first eight layers are in Table 2. Results are confined to these levels because biomass burning emissions can extend up to 10–12 km (33) with peak values usually between 2–6 km.

Most of the Exp A results are in Menon et al. (10), and for brevity we describe the main results as applies to the intercomparison with Exp L1 to L8. We focus on Exp A, L1, L4, and L7 to illustrate key differences in simulations. Whereas surface radiation budgets and surface fluxes are negative for absorbing aerosols, top of the atmosphere fluxes are positive, and the atmospheric column gets heated ($\sim$0.3 K/day) (10). The atmosphere is unstable for aerosols confined to the lowest model layer, with stability increasing as aerosols get distributed to higher levels. Depending on the location of the absorbing aerosol radiative heating/cooling rates, vertical motions, cloud cover, and convection respond quite differently, as we demonstrate below.

Evaporation, land-sea contrasts, surface fluxes, moisture convergence, and convective activity control the precipitation field to a large extent. However, in our simulations, increased summer precipitation over southeastern China is not accompanied by increased evaporation, except in Exp L1 (implying soil moisture does not increase); land-sea contrasts do not play a major role [shown in (10)]. The change in net surface heat flux (defined as the sum of the net surface radiation, latent and sensible heat fluxes, and cooling caused by melting/sublimation) indicate less heat flux for southern China (18–34 N) compared with northern China (34–50 N), with net radiation being the dominant contributor (mainly from reduced surface shortwave radiation). Larger negative values for aerosols at higher levels ($<$550 hPa) (Exp L5 to L8) correspond to cases with the largest increase and decrease in precipitation (Figure 1). To aid the interpretation of the features in Figure 1, the change in the vertical profiles of cloud cover (Figure 2) and temperature are examined. Whereas the overall cloud cover increases in Exp A, for Exp L1 to L8 cloud cover decreases

in the layer where heating occurs [in conformation with the semidirect effect (34, 35)], except for aerosols at higher levels (<550 hPa). For Exp L1 and L2, reduced values of atmospheric heating and surface solar flux intensifies the low-level inversion that slows convection and cloud formation. For Exp L5 to L8, cloud cover decreases only in northern China, with overall increases in cloudiness at all levels for southern China. This is related to the strong updrafts (not shown) that dominate southern portions of China for aerosols at higher levels (<550 hPa), which also exhibit stronger heating gradients between the upper and lower atmospheres. Thus, strong convective activity appears to be the dominant mechanism that produces the precipitation patterns that are observed. Increased updrafts and the increased availability of moisture through circulation changes (not shown) result in increased vertical extent of convection, increased relative humidity, and increased precipitation. Observational evidence of changes in cloudiness due to smoke was also found for the 1991 Kuwait oil fire (36). The intense absorption of solar radiation by smoke induced convection and cloud formation, with roots at the top of the smoke layer that appeared to dominate the cloud evaporation caused by smoke-induced solar heating.

Values for the semidirect and direct effect (Exp A, L1, L4, and L7) over China and India are in Table 3. As can be seen for southern China, the magnitude of the semidirect effect increases as aerosols get distributed to higher levels, and the total forcing (semidirect + direct) for Exp A is $\sim -1.3$ W m$^{-2}$. Positive semidirect forcings are obtained for aerosols at lower levels and negative values for aerosols at higher levels (<550 hPa) for all regions. For northern China and India, the total forcing remains positive for all cases and is $\sim +2.2$ W m$^{-2}$ for Exp A (because cloud cover increase is not as much as over southern China). These findings agree with the results from Penner et al. (37), who found negative semidirect effects for aerosols confined at higher levels.

The above results imply that the strength of convection (caused by changes in the temperature profiles) and the enhanced BC absorption in the presence of clouds may compete with each other in a way that could either offset or enhance the presence of clouds, which in turn affects temperature and radiation. Observational or experimental verification of enhanced BC absorption either in clouds or when

**TABLE 3** Mean values of the semidirect and direct effects over China and India

| | Semidirect (W m$^{-2}$) | | | Direct (W m$^{-2}$) | | |
|---|---|---|---|---|---|---|
| Experiment | 2S–30N 60–100E | 18–34N 90–130E | 34–50N 90–130E | 2S–30N 60–100E | 18–34N 90–130E | 34–50N 90–130E |
| Exp A | −2.05 | −8.04 | −2.74 | 4.33 | 6.73 | 4.9 |
| Exp L1 | 2.35 | 1.62 | 0.32 | −1.20 | −0.36 | 0.24 |
| Exp L4 | −1.53 | −4.20 | −7.28 | 6.29 | 8.40 | 8.33 |
| Exp L7 | −7.97 | −33.8 | −5.9 | 17.1 | 17.9 | 12.6 |

BC is mixed with other aerosols is not well known, and therefore more efforts are needed to quantify these effects. Sato et al. (23) have found that existing BC inventories used in climate models to produce BC absorption optical depths do not match AERONET observations unless the absorption efficiency of BC is higher than currently thought, which suggests internally mixed BC particles.

Greater changes in simulated temperature, radiation, and precipitation for aerosols confined below ∼550 hPa (above ∼3700 m) suggest that climate effects of absorbing aerosols are stronger when they are located at higher levels in the atmosphere. This would apply to biomass emissions that are usually injected at higher altitudes, unless lower-level aerosol emissions get transmitted to higher levels through deep convection. High aerosol loadings at high altitudes observed over east Asia during the Aerosol Characterization Experiment (ACE)-Asia by Mader et al. (38) indicate that convective activity can enhance transport of the aerosols to higher levels, and because most of the Asian emissions are low-level fossil and biofuel generated (with a large proportion of BC) (39), they can have significant effects on climate. The role of convection is also of relevance in view of results from Fromm & Servranckx (40), who found that increased stratospheric aerosol amounts may arise from convective transport of boreal biomass burning. Model simulations of observed precipitation and temperature trends over China match observations mainly due to the effect of absorbing aerosols at higher levels, as was indicated by the series of simulations (Exp L1 to L8).

Thus, these results suggest that greater emphasis should be placed in obtaining realistic vertical profiles of aerosol distributions, which in turn determine the amount of energy and moisture available in the atmosphere.

## 3.3. Semidirect Effects

To differentiate between the direct (changes in radiation without cloud modification) and indirect (changes due to aerosol-cloud interactions) effect of aerosols on climate, we discuss two processes by which absorbing aerosols affect climate: (*a*) the effect that leads to the evaporation of clouds through solar heating of the boundary layer by absorbing aerosols such as BC, termed the semidirect effect (34), and (*b*) a process termed indirect soot forcing (17), which refers to the change in snow albedo due to BC aerosols. The semidirect aerosol effect has not been considered by IPCC (1) but may be quite significant based on the results of Hansen et al. (34) (who found an increase in radiation at the surface due to cloud evaporation that was important locally) and Ackerman et al. (35) (who found that daytime trade cumuli cloud cover could be reduced by almost one half due to solar absorption by aerosols). However, Penner et al. (37) point out that the negative longwave forcings of carbonaceous aerosol emissions (of which soot is a component) that are injected at higher atmospheric levels may cancel out the positive forcings usually associated with the semidirect forcing (because they can increase cloudiness at lower levels, which would decrease temperatures), and thus the semidirect effect associated with carbonaceous aerosols may not act to warm climate. To verify the

positive or negative forcing associated with the semidirect effect requires better information on aerosol and cloud vertical profiles, which is only now being obtained as is discussed in Section 4.

Another potentially important climate effect of soot is its impact on snow albedo. Using a general circulation model and empirical data on soot amount in snow, Hansen & Nazarenko (17) estimate that soot effects on snow and ice albedos result in a forcing of $\sim$0.3 W $m^{-2}$ for the Northern Hemisphere and find the efficacy of this forcing to be approximately twice as effective as $CO_2$ in altering global surface air temperatures. Furthermore, they suggest that this soot effect may have contributed to the past century's global warming, melting land ice and permafrost, early arrival of spring in the Northern Hemisphere, and thinning Artic sea ice. The estimated subjective uncertainty in soot albedo forcing effects (accounting for the uncertainty in snow albedo and soot content measurements, absorption efficiency for internally mixed snow and soot, and relative contribution of the anthropogenic part of soot) could be factor of two. However, the authors argue that the soot-snow albedo effects are large because they exclude other factors (effects of darker surfaces in late winter and spring when higher overhead sun may magnify the absorption efficiency and prolong melt season) that may magnify soot snow albedo effects. Furthermore, they suggest that to avoid dangerous anthropogenic interference (DAI) levels within the climate system, additional global warming should not exceed 1 K. Thus, steps taken to reduce soot emissions will not only reduce surface temperatures but will also restore snow albedos to pristine values, thereby increasing the negative forcing effects of snow (17).

## 3.4. Indirect Effects

For climate change estimates, evaluating the dynamical, microphysical, and radiative changes in cloud properties due to aerosol-cloud interactions remains challenging. Hygroscopic aerosols that serve as efficient cloud condensation nuclei can increase cloud droplet number concentrations (CDNC) and reduce cloud droplet sizes if cloud liquid water content (LWC) remains unchanged (41)—the first indirect effect. Measurements from various field studies (42–47) and satellite retrievals (48–52) corroborate these effects. A consequence of smaller droplet sizes is that they do not grow large enough to participate in cloud droplet coalescence processes, inhibiting precipitation formation and increasing cloud liquid water path (LWP) and cloud lifetime (53)—the second indirect effect. However, it is not yet clear if the cloud water increases, decreases, or stays constant with increasing CDNC (51, 54–56). Satellite data analyses by Han et al. (55) indicate seasonal and regional variabilities. If LWP does not stay constant, the cloud susceptibility [change in the reflectivity of the clouds as a function of CDNC (57)] gets reduced, implying less sensitivity to pollution (55). Because most satellite-based estimates of the indirect effect are related to the change in cloud droplet effective radius ($r_{eff}$) with pollution, understanding causes of variability between cloud droplet radius and cloud liquid water (which are more controlled by dynamical changes) may be

quite important when evaluating changes in $r_{eff}$ with pollution (58).

The increase in precipitation due to the presence of large particles that can enhance the amount of precipitation-sized droplets and increase coalescence, or the decrease in precipitation due to the presence of small particles that do not grow to larger sizes, can have several notable consequences when determining cloud water budgets. Rosenfeld et al. (59) find that although increased pollution can suppress precipitation over land, for polluted deep-ocean clouds with high surface winds the presence of larger sea salt particles can act to cleanse the cloud of pollution by triggering precipitation [also corroborated by findings from Rudich et al. (60) based on satellite retrievals, which suggest that large salt-containing dust particles from the Aral Sea increase cloud droplets to sizes that are likely to precipitate, thus increasing rainfall in these regions] and reducing CDNC and cloud albedo. Rosenfeld et al. (59) suggest that this mechanism could be the likely cause for larger $r_{eff}$ for the same aerosol index over land and ocean as reported by observational studies [such as that of Bréon et al. (61)].

Changes in LWP are an important consideration when evaluating the aerosol indirect effects. However, LWP and precipitation fields are often hard to predict because model parameterization of the autoconversion process (self-collection of cloud droplets) that determines the onset of precipitation is obtained from cloud-resolving models even though spatial scales vary dramatically. Multiscaling efforts required to treat these cloud physical processes in a GCM have not yet been explored adequately, leading to problems in most models when predicting precipitation fields (62).

Estimates of the indirect effects from the various climate models indicate a large range, from $-0.5$ to $-4.4$ W m$^{-2}$. Some of these higher values may be too large based on observed temperature changes over the past 150 years. Rotstayn & Penner (63) estimate climate sensitivity parameters for both the first and second indirect effect (from sulfates only) to be 0.78 and 0.7 K m$^2$ W$^{-1}$. These are comparable to the climate sensitivity parameters derived for the direct sulfate and $2 \times CO_2$ forcings of 0.69 and 1.01 K m$^2$ W$^{-1}$ from the same model, and therefore these indirect effects are quite critical for climate change studies. In addition to these radiative and hydrological effects, the indirect aerosol forcing was also thought to substantially alter low-latitude circulation and rainfall based on results from Rotstayn & Lohmann (8), who used a mixed-layer ocean model coupled to an atmospheric model. They found that the near-global quasi-hemispheric pattern of contrasting SST (sea surface temperature) anomalies associated with trends in the drying conditions found in the Sahel may have some contribution from the indirect effect, although they only include sulfate aerosols in their analysis.

The magnitude of the first indirect effect is quite sensitive to the change in droplet concentration (64–69), whereas that of the second indirect effect is sensitive to model treatment of the autoconversion or precipitation parameterizations (64, 66, 67, 69, 70). Lohmann & Lessins (71) suggest that the combined value of the first and second indirect effect of $-1.28$ W m$^{-2}$ over the oceans, predicted by their climate model, may be overestimated by as much as a factor of 1.3 based on slopes

of $r_{eff}$ and the aerosol index obtained from Polarization and Directionality of the Earth's Reflectance (POLDER) (61). They cite a new global value of $-0.8$ W m$^{-2}$ for the indirect effect. This is within the range of forcing (0 to $-1.2$ W m$^{-2}$) for the indirect aerosol effect based on observed historical surface temperature range or that derived from an analysis of observed changes and model predictions (72). However, these results can only be used to constrain indirect forcing estimates if they are based on physical mechanisms. Lohmann & Lessins (73) further point out that the difference in slopes over land and ocean that they obtained and corrected relative to satellite retrievals may be due to differences in cloud macrophysical properties (cloud LWP, cloud base, and thickness). Thus, using satellite retrievals to constrain model results is a worthy exercise provided caution is used when analyzing satellite retrievals, as is discussed in Section 5.3.

## 4. OBSERVATIONAL EVIDENCE

Analyses of estimates of changes in cloud properties based on satellite retrievals reveal several facets of the relationship that may exist between cloud top temperature, effective radii, CDNC, LWP, and cloud radiative properties such as cloud optical depth and cloud albedo.

Early investigations of the aerosol indirect effect from analysis of satellite retrievals concentrated on statistical means between cloud droplet effective radii, cloud optical depth, and column number concentration (49, 50, 74). Slowly, the importance of changing LWP gained prominence (as discussed in Section 3.4) as some studies (75) suggested a violation of Twomey's definitions of the indirect effect (smaller droplet sizes implying greater reflectivity). This was mainly due to variations in LWP that were not found to be constant but were found to vary depending on locations and seasons (52, 55), although other studies did not report such implied variability in LWP (51, 54). Furthermore, other studies report changing cloud top temperature along with LWP with increasing aerosol number concentrations (76). Rosenfeld (5, 77) provides some evidence of changing $r_{eff}$ and cloud top temperatures for different cloud systems to link the increased pollution with smaller droplet sizes and decreased precipitation. Liu et al. (78) find that over the Indian Ocean region for similar values of cloud LWP, a north-south gradient in CDNC (a factor of $\sim$2.5 increase in CDNC for the north) is visible based on remotely sensed data collected by aircrafts during INDOEX, with decreases in radii of up to 2 $\mu$m for the north compared to the south. A more recent study from Sekiguchi et al. (79) does not find any correlation between cloud top temperature and aerosol number concentrations, although they do not preclude some correlation that may be more dependent on particle radius. They also found positive correlations between cloud optical thickness and cloud cover with aerosol number concentrations. Suzuki et al. (80), in their comparison of GCM cloud fields with that of Advanced Very High Resolution Radiometer (AVHRR) retrievals, found that the positive and negative correlations between LWP and aerosol concentration

($N_a$) may be balanced on a global scale due to the competing effects of increased cloud lifetime and increased aerosol scavenging, whereas LWP tends to have a negative correlation with $N_a$ when using a parameterization that does not incorporate the cloud lifetime effect (i.e., when the precipitation parameterization does not have a dependence on the aerosols).

These results from various groups listed above present some contrasting evidence regarding aerosol effects on cloud microphysics and optics that may arise due to cloud sampling issues (56): assumptions used in the analysis of diagnostics, seasons, location, circulation effects, etc. What is clearly needed is a careful evaluation of cloud properties sampled in similar meteorological settings for a few locations around the globe where meteorology effects may be minimized so that some signal due to aerosol effects on clouds may emerge. This will help establish the nature of the changes in cloud properties solely due to aerosols.

The relationship and competition between cloud lifetime and aerosol scavenging by clouds are manifested locally, thereby precluding general conclusions on a global scale. Regional variability in aerosol distribution results in regional changes that are difficult to capture when using coarse resolution models. Previously, longer time-change climate integrations required coarse grid models due to the computational expense when running long simulations. However, as computational efficiencies improve, models can now use finer grid scales, and therefore future climate simulations should concentrate on comparing regional-scale changes with observations rather than solely investigating global-scale features. Although field studies are useful to compare with models, global coverage from satellite data are needed to verify the global impacts. Satellite data are available from late 1978 [International Satellite Cloud Climatology Project (ISCCP), Total Ozone Mapping Spectrometer (TOMS)] and can be used to decipher impacts over the past two decades. However, global-scale retrievals are not always feasible because some of the present-era satellites (e.g., AVHRR, available since the mid-1980s) provide valuable information mainly over oceans due to the difficulty in obtaining optical properties of land surfaces. Penner et al. (81) also found that three different analyses of optical depths from AVHRR differ by $\sim$0.1 optical depth units mainly due to assumptions used in the retrievals. Cloud screening can also be an issue when analyzing satellite retrievals and could lead to different estimates (0.1 or larger for aerosol optical depths) for the same sets of retrievals (82). With the advent of polarization instrumentations, such as POLDER-1 and -2 (83) and the Research Scanning Polarimeter (RSP) (84) (which are able to provide more information regarding the size, shape, and refractive index of aerosol particles over land), accurate global-scale aerosol retrievals are becoming more feasible. Vertical information is usually more difficult to obtain, but for the first time vertical aerosol profiles will slowly be available through the NASA Geoscience Laser Altimeter System (GLAS) lidar measurements on the Ice Cloud and land Elevation Satellite (ICESat), which was launched in January 2003.

Encouragingly, an intercomparison of existing satellite products from MODIS (Moderate Resolution Imaging Spectroradiometer) or MISR (Multiple Imaging

Spectroradiometer) with AVHRR, TOMS, POLDER, ATSR (Along Track Scanning Radiometer), or GLI (Global Imager) can be performed to constrain aerosol climatology products (85). This is mainly possible due to the apparent lack of sensitivity of measurements to the time of the day (the lifetime of aerosols are of the order of a few days, and the relationship between aerosol loading and synoptic scale meteorological processes are largely independent of diurnal cycles) (85).

## 5. UNDERSTANDING UNCERTAINTIES

### 5.1. Emissions/Distribution

Accounting for sources, sinks, and transport of aerosol properties is complex, and therefore some uncertainty is bound to remain when estimating aerosol budgets, especially for carbonaceous aerosols. BC emissions are usually calculated from fuel consumption data differentiated by the utilization sectors and appropriate emission factors. Several compilations of sector-segregated BC emission factors have been published. Many of these emission factors were obtained from particulate mass and assumed BC mass fractions. Estimated uncertainties in these emission factors range from a factor of two to a factor of eight (86). Dickerson et al. (87) have found that BC estimated from fuel use and known emission factors yields values (0.7 Tg year$^{-1}$) that are much lower than those estimated from ambient concentrations (2–3 Tg year$^{-1}$). It is desirable to ascertain how realistic the emission factors are for BC emissions inventories. This can be accomplished by extensive direct source sampling and comparing model-derived and measured BC concentrations.

Directly measured BC emission factors have only recently been reported for a few related sources (88). These are, of course, characteristic of the technologies and locations where such measurements were done. It is difficult and perhaps even impossible to extend such measurements to all sources found in environments with different fuels and combustion technologies. Even if this were possible, the derived emissison factors would be applicable only to the present conditions and not those that have existed in the past. Several studies used BC emission inventories in global models to estimate BC concentrations and compared those with measured ambient values. Model-simulated and mean measured concentrations were generally within about a factor of two at some sites, with greater discrepancies at other sites (89). A more precise comparison is not feasible because of the lack of systematic BC concentration measurements worldwide because existing measurements were obtained at different time periods using different analytical methods.

To develop adequate mitigation strategies, consensus must be achieved regarding the uncertainty in aerosol radiative forcing estimates from either changes in aerosol distribution or aerosol physical process treatments. Considerable uncertainty exists in emission estimates for BC from biomass burning and can be even as high as $\pm 700\%$ (86). These uncertainties should be quantified separately among models that may use similar aerosol distribution to better separate the contribution to aerosol forcing from aerosol effects or from aerosol process treatment

effects. However, a major complexity that may remain is the determination of the contribution of carbonaceous aerosols to forcing. The OC versus BC fractions that come about from fossil fuel, biofuel, or biomass burning will be a crucial factor in determining forcing estimate changes for the future and in developing climate mitigation strategies (90). Guazzotti et al. (91) found a 74% biomass/biofuel contribution to submicrometer carbonaceous aerosols for the Indian subcontinent, whereas for air masses from the neighboring Arabian Peninsula, there was a 63% contribution to the carbonaceous aerosols from fossil fuel use. Novakov et al. (39) also found greater fossil fuel contribution to the aerosols over the Indian subcontinent based on a two-week study during the INDOEX field study and suggests that seasonality may play a strong role in determining the contribution of fossil fuel or biomass/biofuel to the total aerosol burdens. This variability in aerosol source type has also been reported by several observationally based studies (92–95). Differences in fuel use lead to different ratios of, for example, sulfate to BC because sulfate emissions from biomass/biofuel are typically smaller than that from fossil fuel sources. Emphasis on detailed gas and particle phase measurements is required globally and on a temporal basis to account for regional differences in aerosol emissions and particles that form from them.

## 5.2. Model Process Treatments

Current uncertainties in model treatments of the aerosol effects include limitations in representing cloud-scale processes on model grid scales that are typically several times larger; effects of cloud updrafts on the activation of aerosol particles; relative humidity effects that affect the aerosol size distribution; knowledge of the optical properties of internally mixed aerosols; lack of thorough understanding of aerosol effects on ice nucleation processes; cloud droplet size dispersion effects (96); dynamical influences that may mask aerosol effects on clouds, which require decoupling feedbacks from forcings; and analyzing the extent that anthropogenic perturbations can influence natural dynamical changes, such as the El Niño and North Atlantic Oscillations (97), to name a few.

Here, we focus on the indirect effect because it has the largest uncertainty and is perhaps one of the largest contributors to climate change estimates. In Table 4, we indicate values from different models for the first and second indirect effect for different aerosol species. They range from $-0.4$ to $-3.2$ W m$^{-2}$ depending on the process used in the models to represent the indirect effects. The difficulty in narrowing the wide range of the indirect effects predicted by the different models arises because model treatment of the coupling between aerosols and cloud droplet number, and in the parameterization of aerosol effects on precipitation, differ considerably. Differences exist between mechanistic and empirical treatments of cloud droplet number predictions (62), which may be partly related to inadequacies in model subgrid vertical velocity field diagnostics (which are usually derived from subgrid turbulence fields), although both methods are capable of providing an adequate treatment of cloud droplet number activated from aerosols.

**TABLE 4** Average values of the indirect effect in W $m^{-2}$ estimated by different models for the listed aerosol species and for all aerosols, given as the difference between simulations with present-day aerosols and that with preindustrial aerosols

| Reference | All | Sulfate | OC | BC |
|---|---|---|---|---|
| Ghan et al. (66) | — | −1.7 to −3.2 | — | — |
| Jones et al. (67) | — | −0.5 to −1.5 | — | — |
| Rotstayn (98) | — | −2.1 | — | — |
| *Kiehl et al. (68) | — | −0.4 to −1.78 | — | — |
| *Williams et al. (7) | — | −1.37 | — | — |
| *Chuang et al. (65) | −1.85 | −0.30 | OC + BC = −1.51 | — |
| Lohmann et al. (64) | −1.1 to −1.9 | −0.4 | OC + BC = −0.9 to −1.3 | — |
| Menon (This work) | −2.29 | −1.78 | −1.87 | −1.61 |

*models that simulate the first aerosol indirect effect only. OC and BC stand for organic and black carbon, respectively.

The IPCC (1) estimates almost a factor of three uncertainty in the indirect forcing estimate on the basis of the nucleation and growth partitioning of sulfate mass. Another difference between model estimates of the indirect forcing arises due to the way the background aerosol burden is prescribed. Model estimates of the aerosol indirect effect are based on simulations that distinguish between preindustrial and present-day aerosol emissions. Although this serves as a useful indicator of the relative contribution of the aerosol-cloud effects to the total climate forcing since industrialization, comparison of the estimates with different climate models is difficult in view of the wide range used to represent the preindustrial aerosol concentrations that produce wide ranges in forcings (64–67, 69). For example, simulations with the GISS GCM for the first indirect effect (with sulfates, sea salt, and OC aerosols) with a minimum CDNC = 10 $cm^{-3}$ was −2.1 W $m^{-2}$, whereas that for CDNC = 40 $cm^{-3}$ was −1.1 W $m^{-2}$. This difference of 1 W $m^{-2}$ is quite large and regionally can change the surface temperature response from a positive to a negative, as shown in Figure 3. At present, it is not possible to quantify what this background amount is, but observations can be used as a guideline to provide some constraints. During the second Aerosol Characterization Experiment (ACE-2), held in Tenerife in 1997, the minimum average CDNC values in adiabatic cloud parcels was ∼50 $cm^{-3}$, although values of ∼10 to 20 $cm^{-3}$ have been measured in pure marine background air with no wind (J.-L. Brenguier, personal communication). Although this may apply to the ACE-2 region in the Atlantic Ocean, satellite retrievals of baseline aerosols (99) may also be used to infer background values for CDNC on a global scale.

In addition to these uncertainties that may arise due to the treatment of the coupling between aerosols and CDNC/precipitation, certain other process treatments also can contribute to the aerosol indirect effects. Boers & Mitchell (100) describe a mechanism whereby the albedo enhancement for thin clouds may be

reduced, whereas that for an optically thick cloud can increase due to an absorption feedback effect that is based on the thermodynamic tendency of a cloud to stabilize itself against changes in absorption of solar radiation when cloud microphysics is affected through increased CDNC. This link between the convective and radiative nature of the cloud depends on the amount of convective mixing and the changes in the cloud thickness and therefore will depend on the extent to which the cloud thickness is diagnosed accurately. Most climate models diagnose a cloud thickness that is the same as the layer thickness of the model, i.e., the vertical extent of the cloud is assumed to be the same as the model layer height. For coarse resolution models this would be a drawback, as marine stratocumulus clouds, which cover a large portion of the Earth, are usually not as thick, with optical thickness ranging between 1 and 15 (100).

Most GCM estimates for the indirect effect are for warm nonconvective clouds. Parameterizations that represent aerosol effects on cold clouds are now being developed. The first such global results are from Lohmann et al. (101), who investigated volcanic aerosol effects on cirrus clouds and effects of soot on cirrus clouds (102). However, detailed observations of aerosol types that affect ice-cloud nucleation are still relatively rare, and mechanisms that promote homogeneous or heterogeneous freezing of ice nuclei are still being investigated.

The lack of schemes in GCMs that represent aerosol effects on all cloud types could also hamper comparison of model products with satellite products. As an example, Suzuki et al. (80) analyzed cloud $r_{eff}$ from a GCM and satellite and found that model results were closer to satellite retrievals over the ocean, but larger differences were evident over land locations and the tropics where deep and shallow convective clouds are more common. Because their treatment of the indirect effect was for warm nonconvective clouds, this difference over the tropics is not too surprising. So far, the only GCM estimate of the effect of aerosols on convection is that from Nober et al. (103), who investigated precipitation changes in convective clouds due to aerosol effects and found local changes in precipitation and latent heat release in areas where convective precipitation dominates (areas with strong biomass burning), which in turn affect convective heating, vertical temperature profiles, and global circulation.

Thus, omission of certain types of processes—aerosol effects on cold stratiform clouds and on all convective cloud types—and weakness in certain model diagnostics, such as updraft velocity and cloud thickness, can further exacerbate differences between various model aerosol indirect effect estimates, which makes it difficult to reconcile model cloud products with those from satellite retrievals.

## 5.3. Satellite Retrievals

Because aerosol-climate observations from field studies lack spatial detail, satellite retrieved observations over the past 20 years can be used to help reduce present uncertainties in aerosol climate effects. Once models and satellite data agree,

extrapolating to the past or predicting the future should be less ambiguous.

Changes in cloud properties over land and ocean due to aerosol effects are implied by observations. These are usually reproduced in model simulations but may be a result of assumed differences in background conditions used in models. Differences also arise from the treatment of supersaturation, size distribution, and hygroscopicity of the aerosol particles that affect CDNCs (104). To identify mechanisms that may cause the implied changes in cloud properties over land versus ocean, assumptions used in satellite retrievals need to be analyzed carefully. Factors such as meteorology, cloud LWP variability, decoupling of the boundary layer, types of aerosols, and the type of clouds sampled by particular satellite sensors [e.g., AVHRR data misses optically thin and broken clouds over land, whereas POLDER is biased toward optically thin and less turbulent clouds (105)] have to be considered. This could help generate a more useful data set from the various satellite sensors that can then be used to draw inferences regarding aerosol effects on clouds.

## 6. PREDICTING FUTURE IMPACTS

When projecting future climate change based on IPCC emission scenarios or any other future emission projections, the range in climate sensitivity is important. Climate sensitivity refers to the temperature increase if $CO_2$ were to double and the climate allowed to reach equilibrium. The negative climate forcing due to reflective aerosols and the aerosol-cloud effects could act to mitigate the temperature increase. Most climate models report climate sensitivities between 2.1 to 4.6 K (1). Although it is more difficult to obtain accurate climate sensitivities for short time spans (for a century or so), Knutti et al. (72) obtain a climate sensitivity of $\sim$4.6 K for surface temperature changes over the past century. Uncertainties of -1.5 to 1.5 K were attributed to model uncertainties and $-1.3$ to 2.6 K to uncertainties in the data. Additional uncertainties are thought to be present due to the uncertainties in radiative forcing. Knutti et al. (72) also find low estimates for climate sensitivity when ignoring the indirect aerosol effects and higher climate sensitivities when including a strong aerosol indirect effect. Using the 1906–1995 temperature records to constrain forcings during this time period, Forest et al. (106) estimate that the net forcing for aerosols whose forcing patterns are similar to that of sulfates (ozone, BC, OC, aerosol indirect effects) should be less than $-0.95$ W m$^{-2}$, much lower than IPCC (1) estimates. This net magnitude of $-0.95$ W m$^{-2}$, however, does not distinguish between positive and negative forcings, and thus does not provide sufficient constraints regarding the forcing efficacies for the various aerosols.

Key questions regarding the magnitude of future temperature or precipitation changes that affect climate and in turn influence policy changes are dependent on the certainty with which climate change due to certain aerosol species can be projected. Whereas the $SO_2$ emissions are expected to decline in the future, $NO_x$ as well as carbonaceous emissions are expected to increase (1). Although these are

mainly reflective aerosols, they could influence regional more than global climate because nitrates may be more prevalent in locations that do not have as much sulfate. Estimates of changes in radiative forcing for 2000–2050 based on the Hansen & Sato (107) trends in aerosol and greenhouse gas burdens versus IPCC estimates are in Figure 4. The figure indicates that the forcing from air pollutants can be greatly reduced (e.g., BC and ozone forcings) if BC and ozone emissions decline or do not exceed current levels, in contrast to the larger increases predicted by IPCC. This difference, coupled with the reduced forcing from $CO_2$ (flat $CO_2$ emissions in the "alternative scenario" of Hansen & Sato, versus, 4%/year growth estimated by IPCC) lead to a total forcing estimate of 0.85 W $m^{-2}$ for the next 50 years (from 2000) if the Hansen & Sato scenario emerges. Every 1 W $m^{-2}$ forcing can produce a 3/4°C change in temperature [based on the GISS GCM climate sensitivity (16)]. Every degree of warming is linked to a certain increase (subject to a large uncertainty) in sea-level rise [projected to be rising partly due to thermal expansion of the oceans due to warming and partly due to the melting of glaciers (1)]. This would imply that future projections of forcing estimates, which are used to predict changes in temperature, sea-level, etc., should be based on careful analysis of existing uncertainties and its range. Future aerosol-climate effect simulations should thus also include interactive feedbacks between aerosol processes that affect climate and the changing climate, which could in turn affect aerosol emissions.

## 7. POLICY NEEDS

Most integrated assessment models [e.g., the Mini-Climate Assessment Model of Edmonds et al. (108)] use only greenhouse gases and sulfate aerosols to obtain estimates of impacts of climate change on the economy or environment. This may be unrealistic (J. Sathaye, personal communication) in view of the larger projected impact of carbonaceous aerosols and the change in future ratios of reflective versus absorbing aerosols. The combined estimate and the uncertainty in the aerosol forcing effects implies that a more stringent analysis needs to be undertaken when making impact forecasts.

Jacobson (109) presents a quantitative analysis on controls of BC and OC versus greenhouse gases in controlling warming and the time period over which measures would be effective. He finds that reduction in emissions of fossil fuel BC and associated OC may slow warming more than any reductions in $CO_2$ and $CH_4$ for a specific period. To eliminate 20%–45% of net warming within 3–5 years, all fossil fuel BC and OC would have to be eliminated, and to achieve similar reductions in warming, approximately one third of $CO_2$ emissions would have to be eliminated, although the reduced warming would not come into effect for 50–200 years. Jacobson also calculates the warming due to gasoline versus diesel engines, and he finds that tax laws that promote diesel use may actually increase warming unless stringent emission controls are applied to diesel engines, which may shorten the warming period. Although this study provides useful measures

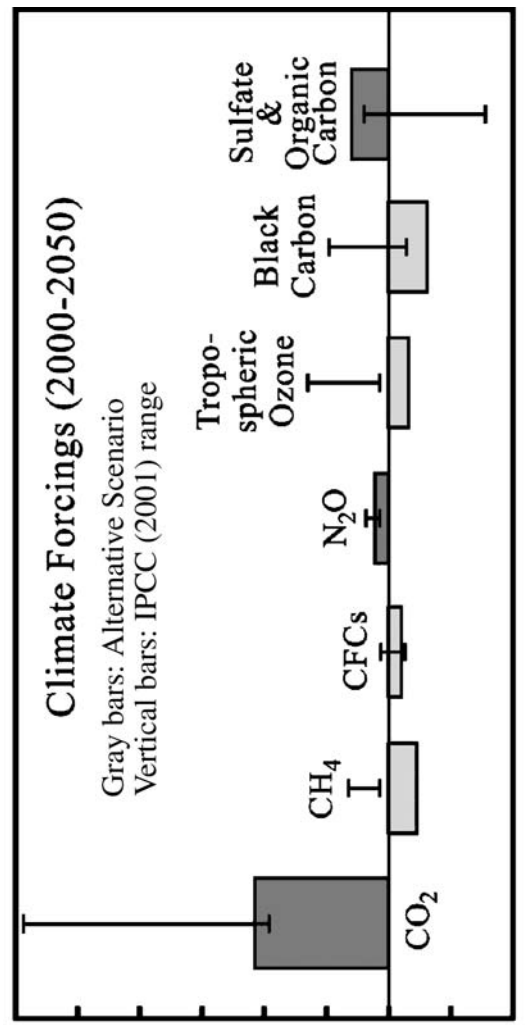


**Figure 4** Climate forcings for 2000–2050 based on the "alternative scenario" of Hansen & Sato (107) and from the IPCC scenario (1). [Reprinted with permission of Hansen & Sato (107).]

on impacts of fossil fuel BC and OC versus greenhouse gases, the results and assumptions used to derive the results need to be further quantified and assessed by other models so that appropriate measures may be taken to evaluate the impacts of diesel fuel use (110–112).

Diesel engine emissions should be quantified not only by weight but also by particle number and chemical content. Limitations of emissions by weight alone may make it relatively easy for engine manufactures to eliminate the largest and heaviest particles, whereas it is the smallest particles with the longest lifetimes that are more harmful to human health (113). Even though efficient particle traps for vehicles may reduce aerosol particles emitted, without fuel efficiency, controls on $CO_2$ will remain unchecked. Because effects from the unrealized warming [estimated to be approximately 0.5 K (3, 114)] of accumulated concentrations of long-lived greenhouse gases persist for several decades, attention should still be focused on $CO_2$ and other greenhouse gases. On the short term, benefits from reduction in aerosols—human health, visibility, acid rain, agriculture, etc.—may outweigh those from controls on $CO_2$. However, future temperatures may increase if aerosol levels are reduced and greenhouse gas levels remain unchanged.

To reduce harmful anthropogenic influences on climate, a global response on several different levels is needed. For example, incentives to coal manufacturers to help them reduce coal emissions and switch to cleaner-burning technologies may be a better alternative to costly litigations that may ensue once public concern with air pollution gains more prominence in the coming years. Issues related to air quality and health may thus be a primary motivating factor that may help reduce emissions.

## 8. SUMMARY

Although the quantification of the effect of anthropogenic activity on climate is uncertain, observational evidence related to aerosol effects on climate suggests that this aerosol effect is real. To avoid harmful impacts on the environment—melting of glaciers, sea-level rise, droughts, floods, change in growing season, reduction in visibility, air pollution, etc.—remedial action ought to be taken to control the human contribution to these impacts. These include application of appropriate standards in industrial activity; agricultural practices; coal, biofuel, and dung burning; and transportation. Because these are undoubtedly tied to economic activity, mitigation efforts to minimize human influence on climate have to be taken with great care. To guide policy experts on the range of steps that would be needed to reduce emissions, accurate climate change scenarios are needed.

To arrive at a reasonable consensus regarding the actual magnitude of anthropogenic influence on climate requires a more integrated understanding of how the processes that affect climate are connected to each other. This process is still under development in most climate models, as models are getting more and more sophisticated in their process treatments. For example, predictions on the effects

of aerosols and greenhouse gases on the hydrological cycle were usually made through separate projections, and rarely were the two effects combined. Even when the effects of greenhouse gases and aerosols are combined, both aerosol indirect effects are usually not incorporated. Because the prediction of rain and LWP budgets by models remain challenging, adequate treatment of the precipitation process is necessary before we can make improved predictions on changes in water budgets (both atmospheric and surface). Also relevant is the aerosol-temperature feedback that results in lower aerosol burdens in a warmer climate due to the linkage between temperature changes and the hydrological cycle (115).

Present estimates of aerosol influence on climate—through the direct, semidirect, thermodynamical, indirect effects, and associated feedbacks—range from $+0.8$ to $-2.4$ W m$^{-2}$, with an implied value of $-1.0$ W m$^{-2}$ (range from $-0.5$ to $-2$ W m$^{-2}$) for the aerosol indirect effect (107). Here, we use a value of $-2.1$ W m$^{-2}$ as an upper bound to the indirect forcing estimate (based on the last two estimates given in Table 4 because they include most aerosol types and both the indirect effects). Combining the positive and negative forcings gives us values of $-1.6$ or $-2.7$ W m$^{-2}$ (for implied values of $-1.0$ or $-2.1$ W m$^{-2}$, respectively, for the indirect effect) for the net aerosol effect. Placing these magnitudes in the context of other forcings, such as greenhouse gases, land use, solar activity, and volcanic aerosol effects, which add up to $\sim$3.35 W m$^{-2}$ (107), results in a contribution of $\sim$50% to 80% from the aerosol effect to the forcing from others.

However, as we have summarized in this review, this estimate can be uncertain (*a*) due to the various processes that affect aerosol distributions—both horizontal and vertical—and (*b*) due to the transformation of the emissions to particles that in turn affect the radiation and other climate diagnostics through the direct, semidirect, thermodynamcial, and indirect effect processes represented in models. In this regard, a multiplatform-based approach that makes use of satellite, meteorological, and process-based observations (field and laboratory) and the integration of these results in climate models are clearly needed to resolve various diagnostics used to decipher climate trends. Integrated assessments can then be reliably made regarding climate change impacts.

## ACKNOWLEDGMENTS

The author thanks Jim Hansen for several insightful comments and guidance on the various aspects of this work, Tica Novakov for his valuable contribution in understanding and elucidating the role of carbonaceous aerosols on climate both historically and in this work, and Tony Del Genio for his thought provoking comments on the aerosol indirect effect. The author also thanks Larrisa Nazarenkov for contributions to the model simulations/results pertaining to the thermodynamical effects of aerosols. The author acknowledges support from the following NASA projects: Global Water and Energy Cycle Project under M. Jasinski and the Global Modeling and Analysis Program under T. Lee, and the Laboratory Directed Research and Development Program of Lawrence Berkeley National

Laboratory.

**The *Annual Review of Environment and Resources* is online at http://environ.annualreviews.org**

## LITERATURE CITED


1. IPCC. 2001. In *Climate Change 2001: The Scientific Basis*, ed. TJ Houghton, Y Ding, DJ Griggs, M Noguer, pp. 289–348. Cambridge, UK: Cambridge Univ. Press
2. Hansen J, Ruedy R, Sato M, Lo K. 2002. Global warming continues. *Science* 295: 275
3. Karl TR, Trenberth KE. 2003. Modern global climate change. *Science* 302:1719–23
4. Bauer E, Claussen M, Brovkin V, Huenerbein A. 2003. Assessing climate forcings of the Earth system for the past millennium. *Geophys. Res. Lett.* 30:1276
5. Rosenfeld D. 1999. TRMM observed first direct evidence of smoke from forest fires inhibiting rainfall. *Geophys. Res. Lett.* 26: 3105–8
6. Ramanathan V, Crutzen PJ, Lelieveld J, Mitra AP, Althausen D, et al. 2001. The Indian Ocean Experiment: An integrated analysis of the climate forcing and effects of the great Indo-Asian haze. *J. Geophys. Res.* 104:2223–31
7. Williams KD, Jones A, Roberts DL, Senior CA, Woodage MJ. 2001. The response of the climate system to the indirect effects of anthropogenic sulfate aerosol. *Clim. Dyn.* 17:845–56
8. Rotstayn LD, Lohmann U. 2002. Tropical rainfall trends and the indirect aerosol effect. *J. Clim.* 15:2103–16
9. Chung CE, Ramanathan V, Kiehl JT. 2002. Effects of south Asian absorbing haze on the northeast monsoon and surface-air heat exchange. *J. Clim.* 15: 2462–76
10. Menon S, Hansen J, Nazarenko L, Luo Y. 2002. Climate effects of black carbon aerosols in China and India. *Science* 297: 2250–53
11. Haywood J, Boucher O. 2000. Estimates of the direct and indirect radiative forcing due to tropospheric aerosols: A review. *Rev. Geophys.* 38:513–43
12. Kirchstetter TW, Novakov T. 2004. Differences in the wavelength dependence of light absorption by aerosols emitted from urban sources and burning biomass. *Atmos. Environ.* In review
13. Haywood JM, Ramaswamy V. 1998. Global sensitivity studies of the direct radiative forcing due to anthropogenic sulfate and black carbon aerosols. *J. Geophys. Res.* 103:6043–58
14. Myhre G, Stordal F, Restad K, Isaksen I. 1998. Estimates of the direct radiative forcing due to sulfate and soot aerosols. *Tellus B* 50:463–77
15. Jacobson MZ. 2000. A physically-based treatment of elemental carbon optics: Implications for global direct forcing of aerosols. *Geophys. Res. Lett.* 27:217–20
16. Hansen J, Sato M, Nazarenko L, Ruedy R, Lacis A, et al. 2002. Climate forcings in GISS SI2000 simulations. *J. Geophys. Res.* 107(D18):4347
17. Hansen J, Nazarenko L. 2004. Soot climate forcing via snow and ice albedos. *Proc. Natl. Acad. Sci. USA* 101:423–28
18. Novakov T, Ramanathan V, Hansen JE, Kirchstetter TW, Sato M, et al. 2003. Large historical changes of fossil-fuel black carbon aerosols. *Geophys. Res. Lett.* 30(6):1324
19. Eck TF, Holben BN, Reid JS, O'Neill NT, Schafer JS, et al. 2003. High aerosol optical depth biomass burning events: A comparison of optical properties for different source regions. *Geophys. Res. Lett.*

30:2035
20. Kaufman YJ, Tanre D, Boucher O. 2002. A satellite view of aerosols in the climate system. *Nature* 419:215–23
21. Ramanthan V, Crutzen PJ, Kiehl JT, Rosenfeld D. 2001. Aerosols, climate and the hydrological cycle. *Science* 294: 2119–24
22. Chylek P, Hallett J. 1992. Enhanced absorption of solar radiation by cloud droplets containing soot particles in their surface. *Q. J. R. Meteorol. Soc.* 118:167–72
23. Sato M, Hansen J, Koch D, Lacis A, Ruedy R, et al. 2003. Global atmospheric black carbon inferred from AERONET. *Proc. Nat. Acad. Sci. USA* 100:6319–24
24. Nakajima T, Sekiguchi M, Takemura T, Uno I, Higurashi A, et al. 2003. Significance of direct and indirect radiative forcings of aerosols in the East China Sea region. *J. Geophys. Res.* 108:8658
25. Douville H, Royer K-F, Polcher J, Cox P, Gedney N, et al. 2000. Impact of doubling $CO_2$ on the Asian summer monsoon: Robust versus model-dependent responses. *J. Meteor. Soc. Jpn.* 78:421–39
26. Roeckner EL, Bengtsson L, Feichter J, Lelieveld J, Rodhe H. 1999. Transient climate change simulations with a coupled atmosphere-ocean GCM including the tropospheric sulfur cycle. *J. Clim.* 12:3004–32
27. Xu Q. 2001. Abrupt change of the mid-summer climate in central east China by the influence of atmospheric pollution. *Atmos. Environ* 35:5029–40
28. Meehl GA. 1994. Influence of the land surface in the Asian summer monsoon external conditions. *J. Clim.* 7:1033–49
29. Srinivasan J. 2001. A simple thermodynamic model for seasonal variation of monsoon rainfall. *Curr. Sci.* 80:73–77
30. Satheesh SK, Ramanathan V. 2000. Large differences in the tropical aerosol forcing at the top of the atmosphere and Earth's surface. *Nature* 405:60–63
31. Iwasaki T, Kitagawa H. 1998. A possible link of aerosol and cloud radiation to Asian summer monsoon and its implications in long-range numerical weather prediction. *J. Meteorol. Soc. Jpn.* 76:965–82
32. Koch D. 2001. The transport and direct radiative forcing of carbonaceous and sulfate aerosols in the GISS GCM. *J. Geophys. Res.* 106:20311–32
33. Anderson BE, Grant WB, Gregory GL, Browell EV, Collins JE, et al. 1996. Aerosols from biomass burning over the tropical South Atlantic region: Distribution and impacts. *J. Geophys. Res.* 101: 24117–37
34. Hansen J, Sato M, Ruedy R. 1997. Radiative forcing and climate response. *J. Geophys Res.* 102:6831–64
35. Ackerman AS, Toon OB, Steven DE, Heymsfield AJ, Ramanathan V, Welton EJ. 2000. Reduction of tropical cloudiness by soot. *Science* 288:1042–48
36. Rudich Y, Sagi A, Rosenfeld D. 2003. Influence of the Kuwait oil fires plume (1991) on the microphysical development of clouds. *J. Geophys. Res.* 108: 4478
37. Penner JE, Zhang SY, Chuang CC. 2003. Soot and smoke aerosol may not warm climate. *J. Geophys. Res.* 108:4657
38. Mader BT, Flagan RC, Seinfeld JH. 2002. Airborne measurements of atmospheric carbonaceous aerosols during ACE-Asia. *J. Geophys. Res.* 107:4704
39. Novakov T, Andreae MO, Gabriel R, Kirchstetter TW, Mayol-Bracero OL, Ramanathan V. 2000. Origin of carbonaceous aerosols over the Indian Ocean: Biomass burning or fossil fuels? *Geophys. Res. Lett.* 27:4061–64
40. Fromm MD, Servranckx R. 2003. Transport of forest fire smoke above the tropopause by supercell convection. *Geophys. Res. Lett.* 30:1542
41. Twomey S. 1977. The influence of pollution on the shortwave albedo of clouds. *J. Atmos. Sci.* 34:1149–52

42. Leaitch WR, Banic CM, Isaac GA, Couture MD, Liu PSK, et al. 1996. Physical and chemical observations in marine stratus during 1993 NARE: Factors controlling cloud droplet number concentration. *J. Geophys. Res.* 101:29123–35
43. Borys RD, Lowenthal DH, Wetzel MA, Herrera F, Gonzalez A, Harris J. 1998. Chemical and microphysical properties of marine stratiform cloud in the north Atlantic. *J. Geophys. Res.* 103:22073–85
44. Brenguier J-L, Pawlowska H, Schuller L, Preusker R, Fischer J, Fouquart Y. 2000. Radiative properties of boundary layer clouds: Droplet effective radius versus number concentration. *J. Atmos. Sci.* 57: 803–21
45. Menon S, Saxena VK. 1998. Role of sulfates in regional cloud-climate interactions. *Atmos. Res.* 4748:299–315
46. Menon S, Saxena VK, Durkee P, Wenny BN, Nielsen K. 2002. Role of sulfate aerosols in modifying the cloud albedo: A closure experiment. *Atmos. Res.* 61:169–87
47. Feingold G, Eberhard WL, Veron DE, Previdi M. 2003. First measurements of the Twomey aerosol indirect effect using ground-based remote sensors. *Geophys. Res. Lett.* 30(6):1287
48. Kaufman YJ, Fraser RS. 1997. Control of the effect of smoke particles on clouds and climate by water vapor. *Science* 277:1636–39
49. Han Q, Rossow WB, Chou J, Welch RM. 1998. Global survey of the relationships of cloud albedo and liquid water path with cloud droplet size using ISCCP data. *J. Clim.* 11:1516–28
50. Han Q, Rossow WB, Chou J, Welch RM. 2000. Near-global survey of cloud column susceptibility using ISCCP data. *Geophys. Res. Lett.* 27:3221–24
51. Coakley JA, Walsh CD. 2002. Limits to the aerosol indirect radiative effect derived from observations of ship tracks. *J. Atmos. Sci.* 59:668–80
52. Schwartz SE, Harshvardsan, SE, Benkovitz CM. 2002. Influence of anthropogenic aerosol on cloud optical depth and albedo shown by staellite measurements and chemical transport modeling. *Proc. Natl. Acad. Sci. USA* 99:1784–89
53. Albrecht BA. 1989. Aerosols, cloud microphysics, and fractional cloudiness. *Science* 245:1227–30
54. Nakajima T, Higurashi A, Kawamoto K, Penner JE. 2001. A study of correlation between satellite-derived cloud and aerosol microphysical parameters. *Geophys. Res. Lett.* 28:1171–74
55. Han Q, Rossow WB, Zeng J, Welch RM. 2002. Three different behaviors of liquid water path of water clouds in aerosol-cloud interactions. *J. Atmos. Sci.* 59:726–35
56. Ackerman AS, Toon OB, Steven DE, Coakley JA Jr. 2003. Enhancement of cloud cover and suppression of nocturnal drizzle in stratocumulus polluted by haze. *Geophys. Res. Lett.* 30:1381
57. Twomey S. 1991. Aerosols, clouds and radiation. *Atmos. Res.* 65:109–28
58. Brenguier J-L, Pawlowska H, Schuller L. 2002. Cloud microphysical and radiative properties for parameterization and satellite monitoring of the indirect effect of aerosol on climate. *J. Geophys. Res.* 108:8632
59. Rosenfeld D, Lahav R, Khain A, Pinsky M. 2002. The role of sea spray in cleansing air pollution over ocean via cloud processes. *Nature* 297:1667–70
60. Rudich Y, Khersonsky O, Rosenfeld D. 2002. Treating clouds with a grain of salt. *Geophys. Res. Lett.* 29:2060
61. Bréon F-M, Tanre D, Generoso S. 2002. Aerosol effect on cloud droplet size monitored from satellite. *Science* 295:834–38
62. Menon S, Brenguier J-L, Boucher O, Davison P, Del Genio AD, et al. 2003. Evaluating aerosol/cloud radiation process parameterizations with single-column models and ACE-2 cloudy column observations. *J. Geophys. Res.* 108:

4762
63. Rotstayn LD, Penner JE. 2001. Indirect aerosol forcing, quasi forcing and climate response. *J. Clim.* 14:2960–74
64. Lohmann U, Feichter J, Penner J, Leaitch R. 2000. Indirect effect of sulfate and carbonaceous aerosols: A mechanistic treatment. *J. Geophys. Res.* 105:12193–206
65. Chuang CC, Penner JE, Prospero JM, Grant KE, Rau GH, Kawamoto K. 2002. Cloud susceptibility and the first aerosol indirect forcing: Sensitivity to black carbon and aerosol concentrations. *J. Geophys. Res.* 107:4564
66. Ghan S, Easter R, Hudson J, Bréon F-M. 2001. Evaluation of aerosol indirect forcing in MIRAGE. *J. Geophys. Res.* 106:5317–34
67. Jones A, Roberts DL, Woodage MJ, Johnson CE. 2001. Indirect sulphate aerosol forcing in a climate model with an interactive sulphur cycle. *J. Geophys. Res.* 106:20293–310
68. Kiehl JT, Schneider TL, Rasch PJ, Barth MC, Wong J. 2000. Radiative forcing due to sulfate aerosols from simulations with the National Center for Atmospheric Research Community Climate Model Version 3. *J. Geophys. Res.* 105:1441–57
69. Menon S, Del Genio AD, Koch D, Tselioudis G. 2002. An evaluation of the sulfate and carbonaceous aerosol indirect effect using the GISS coupled chemistry-climate model and satellite observations. *J. Atmos. Sci.* 59:692–713
70. Rotstayn LD. 2000. On the tuning of autoconversion parameterizations in climate models. *J. Geophys. Res.* 105:15495–507
71. Lohmann U, Lesins G. 2002. Stronger constraints on the anthropogenic indirect aerosol effect. *Science* 298:1012–15
72. Knutti R, Stocker TF, Joos F, Plattner G-K. 2002. Constraints on radiative forcing and future climate change from observations and climate model ensembles. *Nature* 416:719–23
73. Lohmann U, Lesins G. 2003. Comparing continental and oceanic cloud susceptibilities to aerosols. *Geophys. Res. Lett.* 30:1791
74. Chameides WL, Luo C, Saylor R, Streets D, Huang Y, et al. 2002. Correlation between model-calculated anthropogenic aerosols and satellite-derived cloud optical depths: Indication of indirect effect? *J. Geophys. Res.* 107:4085
75. Schuller L, Brenguier J-L, Pawlowska H. 2003. Retrieval of the microphysical, geometrical, and radiative properties of marine stratocumulus from remote sensing. *J. Geophys. Res.* 108:8631
76. Harshvardhan, H. Schwartz SE, Benkovitz CM, Guo G. 2002. Aerosol influence on cloud microphysics examined by satellite measurements and chemical transport modeling. *J. Atmos. Sci.* 59:714–25
77. Rosenfeld D. 2000. Suppression of rain and snow by industrial pollution. *Science* 287:1793–96
78. Liu GS, Shao HF, Coakley JA, Curry JA, Haggerty JA, Tschudi MA. 2003. Retrieval of cloud droplet size from visible and microwave radiometric measurements during INDOEX: Implication to aerosols' indirect effect. *J. Geophys. Res.* 108:4006
79. Sekiguchi M, Nakajima T, Suzuki K, Kawamoto K, Higurashi A, et al. 2003. A study of the direct and indirect effects of aerosols using global satellite data sets of aerosol and cloud parameters. *J. Geophys. Res.* 108:4699
80. Suzuki K, Nakajima T, Numaguti A, Takemura T, Kawamoto K, Higurashi A. 2004. A study of the aerosol effect on a cloud field with simultaneous use of GCM modeling and satellite observation. *J. Atmos. Sci.* 61:179–94
81. Penner JE, Zhang SY, Chin M, Chuang C, Feichter J, et al. 2002. A comparison of model and satellite derived aerosol optical depth and reflectivity. *J. Atmos. Sci.* 59: 441–60
82. Mischenko MK, Geogdzhayev I, Cairns B, Rossow WB, Lacis AA. 1999. Aerosol retrievals over the ocean using channel 1

and 2 AVHRR data: A sensitivity analysis and preliminary results. *Appl. Opt.* 38: 7325–41
83. Bréon F-M, Goloub P. 1998. Cloud droplet effective radius from spaceborne polarization measurements. *Geophys. Res. Lett.* 25:1879–82
84. Chowdhary J, Cairns B, Mishchenko M, Travis L. 2001. Retrieval of aerosol properties over the ocean using multispectral and multiangle photopolarimetric measurements from the Research Scanning Polarimeter. *Geophys. Res. Lett.* 28:243–46
85. Kaufman YJ, Holben BN, Tanre D, Slutsker I, Smirnov A, Eck TF. 2000. Will aerosol measurements from Terra and Aqua polar orbiting satellites represent the daily aerosol abundance and properties. *Geophys. Res. Lett.* 27:3861–64
86. Streets DG, Yarber KF, Woo J-H, Carmichael GR. 2003. Biomass burning in Asia: Annual and seasonal estimates and atmospheric emissions. *Glob. Biogeochem. Cycl.* 17:1099
87. Dickerson RR, Andreae MO, Campos T, Mayol-Bracero OL, Neususess C, Streets DG. 2002. Analysis of black carbon and carbon monoxide observed over the Indian Ocean: Implications for emissions and photochemistry. *J. Geophys. Res.* 107:8017
88. Bond TC, Streets DG, Yarber KF, Nelson SM, Woo J-H, Klimont Z. 2004. A technology-based global inventory of black and orgnaic carbon emissions from combustion. *J. Geophys. Res.* 109:doi: 101029/2003JD003697
89. Cooke WF, Ramaswamy V, Kasibhatla P. 2002. A general circulation model study of carboncaeous aerosol distribution. *J. Geophys. Res.* 107:4279
90. Hansen JE, Bond T, Cairns B, Gaeggeler H, Liepert B, et al. 2004. Carbonaceous aerosols in the industrial era. *AGU EOS Newsl.* 85(25):1
91. Guazzotti SA, Suess DT, Coffee KR, Quinn PK, Bates TS, et al. 2003. Characterization of carbonaceous aerosols outflow from India and Arabia: Biomass/biofuel burning and fossil fuel combustion. *J. Geophys. Res.* 108:4485
92. Mayol-Bracero OL, Gabriel R, Andreae MO, Kirchstetter TW, Novakov T, et al. 2002. Carbonaceous aerosols over the Indian Ocean during the Indian Ocean Experiment (INDOEX): Chemical characterization, optical properties and probable sources. *J. Geophys. Res.* 107:8030
93. Salam A, Kassin K, Ullah SM, Puxbaum H. 2003. Aerosol chemical characteristics of an island site in the Bay of Bengal (Bhola-Bangladesh). *J. Environ. Monit.* 5: 1–9
94. Salam A, Bauer H, Kasisn K, Ullah SM, Puxbaum H. 2003. Aerosol chemical characteristics of a mega-city in Southeast Asia (Dhaka-Bangladesh). *Atmos. Environ.* 37:2517–28
95. Alfaro SC, Gaudichet A, Rajot JL, Gomes L, Maille M, Cachier H. 2003. Variability of aerosol size-resolved compostion at an Indian coastal site during the INDOEX intensive field phase. *J. Geophys. Res.* 108:4235
96. Liu Y, Daum PH. 2002. Indirect warming effect from dispersion forcing. *Nature* 419:580–81
97. Chung CE, Ramanathan V. 2003. South Asian haze forcing: Remote impacts with implications to ENSO and AO. *J. Clim.* 16:1791–806
98. Rotstayn LD. 1999. Indirect forcing by anthropogenic aerosols: A global climate model calculation of the effective-radius and cloud-lifetime effects. *J. Geophys. Res.* 104:9369–80
99. Kaufman YJ, Smirnov A, Holben B, Dubovik O. 2001. Baseline maritime aerosols: Methology to derive the optical thickness and scattering properties. *Geophys. Res. Lett.* 28:3251–54
100. Boers R, Mitchell RM. 1994. Absorption feedback in stratocumulus clouds, influence on cloud top albedo. *Tellus A* 46:229–41
101. Lohmann U, Karcher B, Timmreck C.

2003. Impact of the Mt. Pinatubo eruption on cirrus clouds formed by homogenous freezing in the ECHAM4 GCM. *J. Geophys. Res.* 108:4568

102. Lohmann U. 2002. Possible aerosol effects on ice clouds via contact nucleation. *J. Atmos. Sci.* 59:647–56
103. Nober FJ, Graf H-F, Rosenfeld D. 2003. Sensitivity of the global circulation to the suppression of precipitation by anthropogenic aerosols. *Glob. Planet. Change* 37:57–80
104. Feingold G, Remer LA, Ramaprasad J, Kaufman YJ. 2001. Analysis of smoke impact on clouds in Brazilian biomass burning regions: An extension of Twomey's approach. *J. Geophys. Res.* 106: 22907–22
105. Rosenfeld D, Feingold G. 2003. Explanation of discrepancies among satellite observations of the aerosol indirect effects. *Geophys. Res. Lett.* 30:1776
106. Forest CE, Stone PH, Sokolov AP, Allen MR, Webster MD. 2002. Quantifying uncertainties in climate system properties with the use of recent climate observations. *Science* 295:113–17
107. Hansen J, Sato M. 2001. Trends of measured climate forcing agents. *Proc. Natl. Acad. Sci. USA* 98:14778–82
108. Edmonds J, Wise M, Pitcher H, Richels R, Wigley T, MacCracken C. 1997. An integrated assessment of climate change and the accelerated introduction of advanced energy technologies. *Mitig. Adapt. Strateg. Glob. Change* 1:311–39
109. Jacobson MZ. 2002. Control of fossil-fuel particulate black carbon and organic matter, possibly the most effective method of slowing global warming. *J. Geophys. Res.* 107:4410
110. Chock DP, Song QY, Hass H, Schell B, Ackermann I. 2003. Comment on "Control of fossil-fuel particulate black carbon and organic matter, possibly the most effective method of slowing global warming" by M.Z. Jacobson. *J. Geophys. Res.* 108:4769
111. Feichter J, Sausen R, Grassl H, Fiebig M. 2003. Comment on "Control of fossil-fuel particulate black carbon and organic matter, possibly the most effective method of slowing global warming" by M.Z. Jacobson. *J. Geophys. Res.* 108:4767
112. Penner JE. 2003. Comment on "Control of fossil-fuel particulate black carbon and organic matter, possibly the most effective method of slowing global warming" by M.Z. Jacobson. *J. Geophys. Res.* 108:4771
113. Acid News. 2003. Stricter standards proposed. 4:18–19
114. Hansen JE. 2002. A brighter future. *Clim. Change* 52:435–40
115. Feichter J, Roeckner E, Lohmann U, Liepert B. 2004. Nonliner aspects of the climate response to greenhouse gas and aerosol forcing. *J. Clim.* 17(12):2384–98

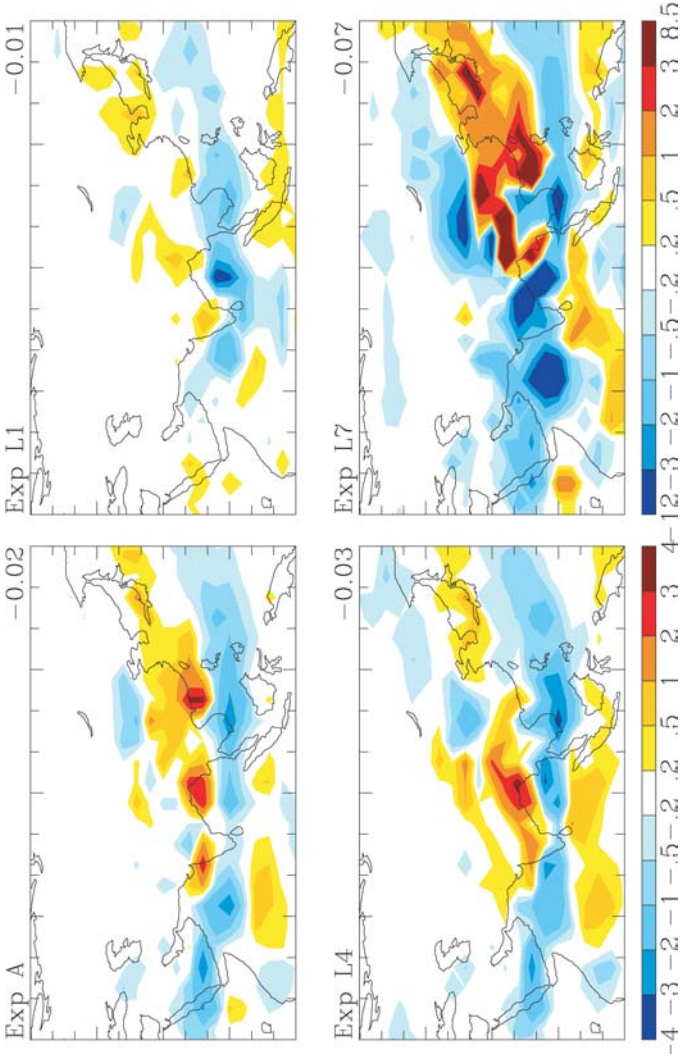


**Figure 1** Simulated changes in precipitation fields (millimeter/day) for June-July-August for Exp A, L1, L4, and L7.

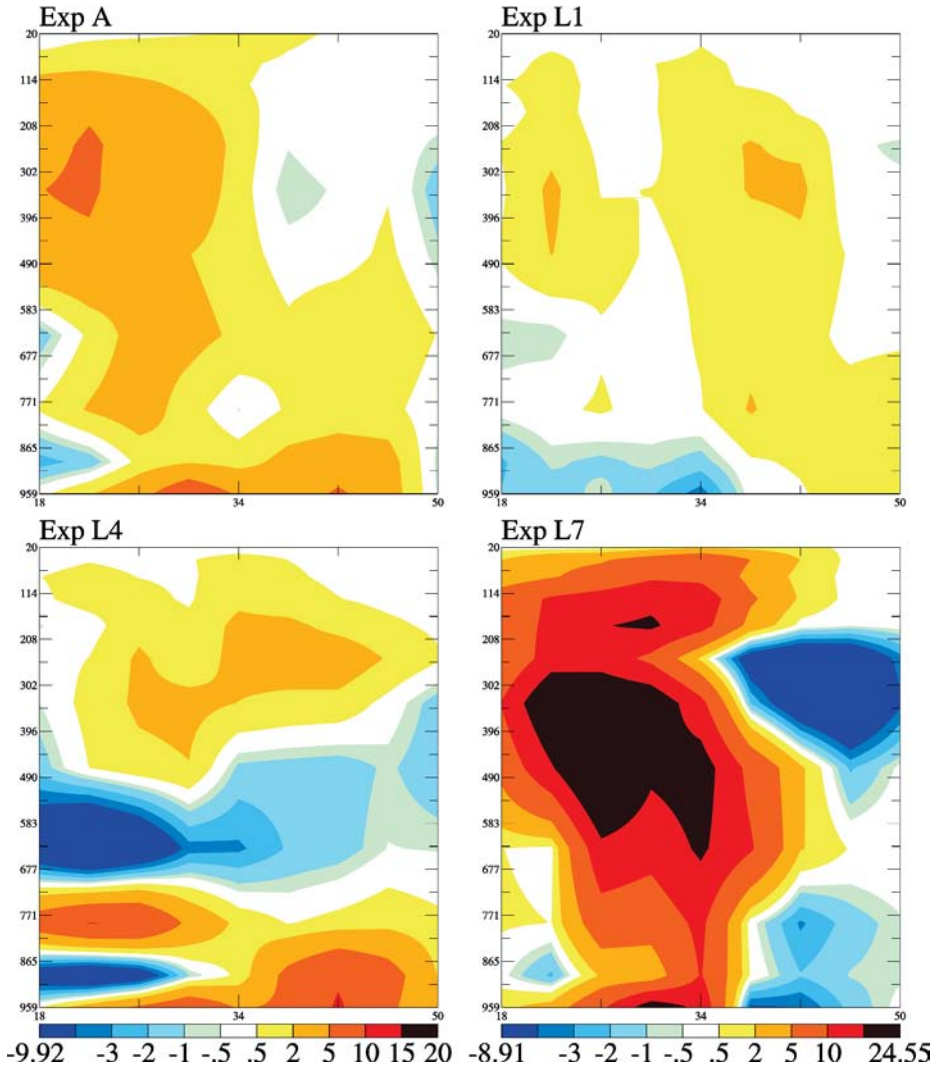


**Figure 2** Similar to Figure 1 but for vertical profiles of total cloud cover change (%). Values extend from 2S to 62N and are averaged from 90 to 130E.

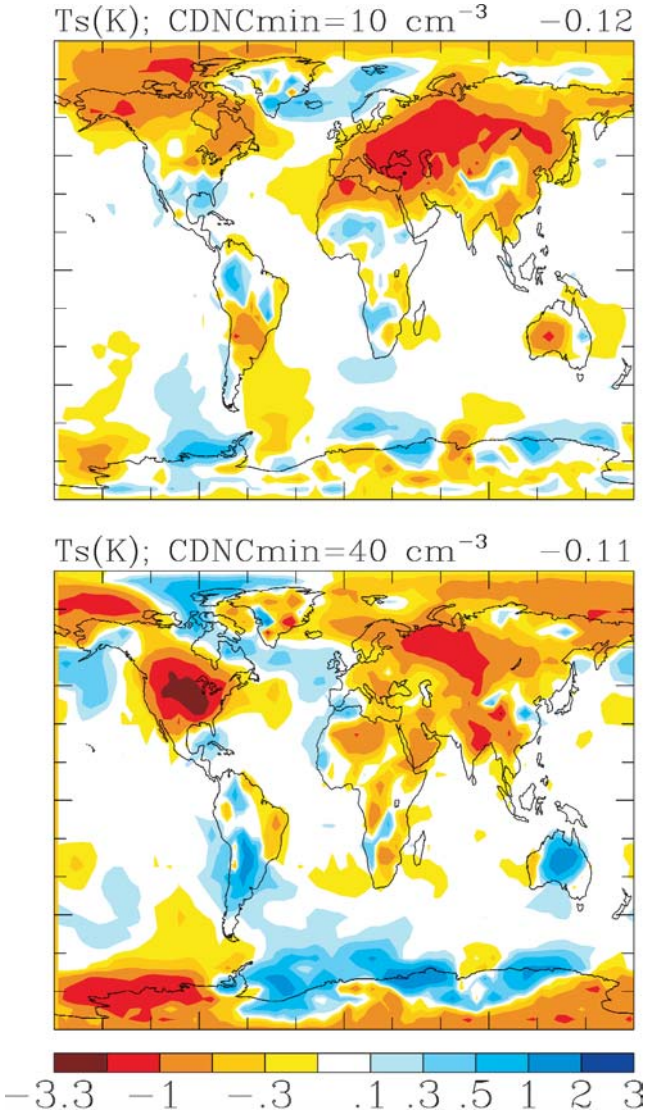


**Figure 3** Changes in simulated surface temperature (Ts) for the first indirect effect for a minimum value of cloud droplet number concentration (CDNC) = 10 $cm^{-3}$ and when CDNC = 40 $cm^{-3}$. Changes are for present-day versus preindustrial aerosol distributions. Global means are on the upper right-hand corner.