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Aerosols and Clouds in Chemical Transport Models and Climate Models

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Abstract

Clouds exert major influences on both short- and longwave radiation as well as on the hydrological cycle. Accurate representation of clouds in climate models poses a major problem because of the high sensitivity of radiative transfer and water cycle to cloud properties and processes, an incomplete understanding of these processes, and the wide range of scales over which these processes occur. Small changes in the amount, altitude, physical thickness, and/or microphysical properties of clouds that occur as a result of human influence can exert changes in Earth's radiation budget comparable to the radiative forcing by anthropogenic greenhouse gases, thus either partly offsetting or enhancing the warming due to these gases. Because clouds form on aerosol particles, changes in the amount and/or composition of aerosols affect clouds in various ways. The forcing of the radiation balance due to aerosol–cloud interactions (indirect aerosol effect) has large uncertainties because a variety of important processes are not well understood, precluding their accurate representation in models.

Introduction

Clouds are an extremely important element of Earth's climate system. They are highly reflective in the solar spectrum, yet strongly absorbing in the thermal infrared; consequently, they produce a large impact on Earth's radiation budget. This impact, termed cloud radiative forcing (CRF), has been quantified through satellite observations: globally, on average, clouds decrease the absorption of solar radiation by about 50 W m^{-2} (shortwave CRF) and decrease the upwelling thermal infrared radiation by 30 W m^{-2} (longwave CRF), thus exerting a net CRF of about –20 W m^{-2} (Kiehl and Trenberth 1997). Locally

and instantaneously, clouds can reduce absorbed shortwave radiation by as much as 700 W m^{-2}. In addition, clouds play a central role in Earth's hydrological cycle, which is coupled to the energy budget through the release of latent heat that results from water condensation or evaporation. This, in turn, influences atmospheric circulation on a variety of scales.

The nature and extent of these cloud processes may be expected to change in the future in response to changes in concentrations and properties of trace gases and aerosols and resulting changes in climate. Thus, it is imperative for clouds, as well as their radiative and hydrological properties, to be represented accurately in climate models. However, for a variety of reasons, accurate representation of clouds and cloud influences on radiation and hydrology in climate models remains particularly challenging. Key among these reasons are:

- the small fraction of the total water in the cloud that is present in condensed (solid or liquid) phase; this necessitates an accurate representation of both the total water content and temperature governing saturation vapor concentration;
- the complexities associated with the presence of several forms of condensed phase water (liquid, supercooled liquid, ice, mixed);
- the spatial and temporal diversity of cloud microphysical structure, as reflected in the number concentration and size distribution of cloud hydrometeors and the crystal habit of ice clouds; and
- the numerous varieties and morphologies of clouds as well as the resultant complexity of their three-dimensional structure on many scales (see Figure 23.1).

Small changes to macrophysical (coverage, structure, altitude) or microphysical properties (droplet size, phase) can exert substantial effects on climate. For example, a 5% increase of the shortwave cloud forcing, which could result from changes in the nature or amount of the atmospheric aerosol, would be enough to compensate for the increase in greenhouse gases between 1750–2000 (Ramaswamy et al. 2001). Recognition of this has stimulated the development of improved physically based representations of cloud processes, in general, and of aerosol influences on clouds, in particular, for inclusion in climate models. However, despite intensified research, the feedbacks on clouds and cloud processes that result from forcings by increasing greenhouse gases and aerosols remain among the greatest uncertainties in climate modeling projections of future and climate change (Randall et al. 2007). Similarly, understanding the radiative forcing by aerosols through their influences on clouds remains the greatest uncertainty in radiative forcing of climate change over the industrial period (IPCC 2007).

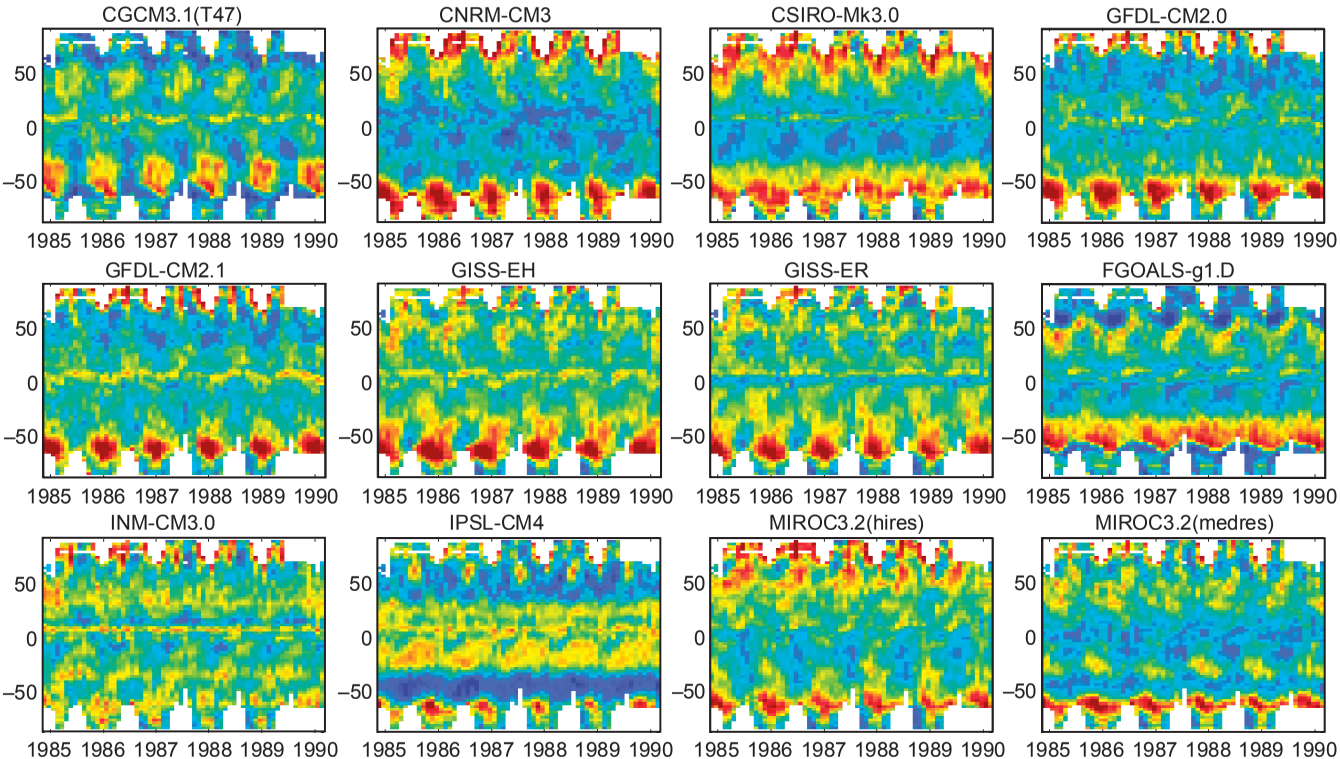
The principal tools for examining prospective consequences of future emissions of greenhouse gases and aerosols on Earth's climate are general circulation models (GCMs). The acronym GCM is also used to denote global climate model, and the terms are often used interchangeably. Global climate models are

Figure 23.1 Complexity of three-dimensional structure of clouds; note penetration of cumulonimbus clouds through thin cirrus layer (courtesy of Y.-N. Lee, Brookhaven National Laboratory).

not only the primary tool for simulating global climate change; they are also used to evaluate the regional effects of anthropogenic emissions on modifying precipitation amounts and distribution. By integrating atmospheric, radiative, oceanic, and land-surface processes on a global scale, global climate models can provide an indication of expected changes in the coupled system, including possible consequences of coupled increases in greenhouse gases and aerosols on atmospheric radiation, clouds, precipitation, and the climate system in general. Here we examine the current state of understanding of aerosol and cloud processes that must be represented in GCMs and the state of such representation. In addition, we identify recent advances and further developments that are needed.

When used to examine aerosol influences on clouds and precipitation, GCMs must accurately represent the macrophysical properties of clouds and precipitation, including their geographical and seasonal variation. Although GCMs have been used to examine the influence of widespread anthropogenic sources of cloud condensation nuclei (CCN) on global climate (i.e., aerosol particles that serve as the nuclei on which cloud droplets form), this has presented numerous problems. First is the issue of scales. Typically, GCM grid cells have a horizontal dimension of 150–250 km and a vertical dimension of hundreds to thousands of meters, over which there can be substantial spatial inhomogeneity. For example, clouds cover often only a small fraction of the volume of a grid cell, necessitating rather ad hoc parameterizations, and the average vertical velocities in a grid cell are very small (~0.01 m s^{-1}), whereas actual vertical velocities, which control cloud formation and the activation of aerosol particles to cloud droplets, might be 1 m s^{-1} or greater. The poor representation of convection is likely a major source of error in modeled liquid and solid water in clouds.

It is clear that there are major disparities among GCMs (see Figure 23.2), even in zonal averages of cloud albedo, which is a major determinant of Earth's


CGCM3.1(T47)
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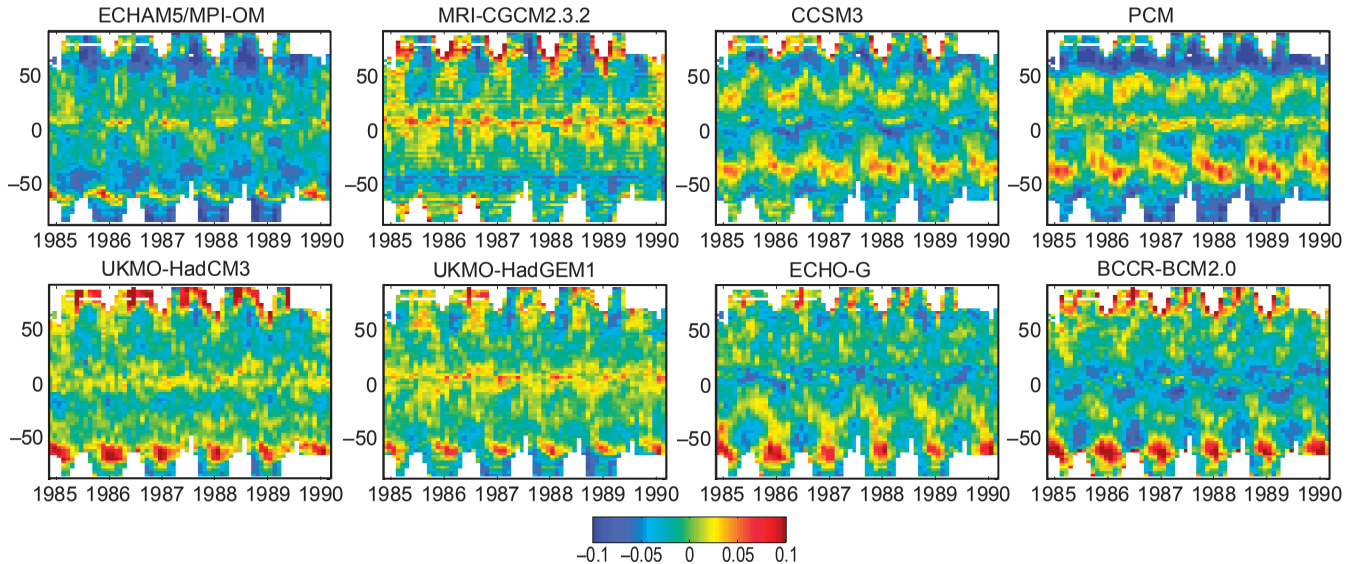

Figure 23.2 Difference between cloud albedo as determined by satellite measurements (ERBE) and twenty global climate models as a function of latitude and time (November 1984 to February 1990). Positive anomalies, where ERBE is higher, are indicated with red, and negative anomalies, where ERBE is lower, with blue colors. Courtesy of Frida Bender; modified from Bender at al. (2006).

radiation budget. In Figure 23.2, each panel corresponds to a different climate model. The model output was obtained from coordinated simulations with twenty different coupled ocean–atmosphere climate models, performed in support of the IPCC Fourth Assessment Report. Clearly these models cannot all be correct. Although space-based measurements can identify models that are doing better or worse, relative to this important cloud variable, such measurements are also difficult, although uncertainties in observations are smaller than intermodel differences.

Cloud microphysical properties are determined by processes such as droplet and crystal nucleation, condensation, evaporation, gravitational settling, and precipitation, all of which operate at the scale of the individual cloud particles or local populations. In contrast, the spatial distribution of clouds is determined by dynamic processes (e.g., turbulence, updrafts, downdrafts, and frontal circulations) and radiative cooling, which operate across meter to global scales. However, these scales are coupled by a variety of processes (e.g., microphysical influences on precipitation development), which in turn affect the release of latent heat below cloud that affects atmospheric stability and vertical motions. Treatment of these processes in climate models and the confidence in this treatment are limited by a lack of understanding and computational resources to represent these on all relevant scales; the latter necessitates development and application of parameterizations, which are inherently scale-dependent. The requirement of accurately representing the many roles of clouds in the climate system and more generally in the biogeochemistry of the planet applies not only to the present atmosphere but also to prior atmospheres (necessary for evaluation of performance of climate models over the instrumental record of the past 150 years or so) and to future atmospheres (necessary for evaluation of the influences of different projected emissions scenarios of greenhouse gases and aerosols). A concern is that each role's common dependence on many of the same cloud properties and processes suggests that errors in simulating one role would produce errors in other roles. Conversely, improving cloud treatment to reduce uncertainty in one role may also reduce uncertainty in other roles. Hence, improving representations and parameterizations of cloud processes in climate models will produce benefits well beyond the simulation of cloud feedbacks and aerosol indirect effects.

Representation of Aerosols in Global-scale Chemical Transport Models and Global Climate Models

Although there are many similarities between treatment of aerosol processes in chemical transport models (CTMs) and global climate models, it is useful to distinguish the two modeling approaches. Global climate models simulate their own meteorology and couple aerosol cycles with clouds, precipitation, and radiation transfer, thereby allowing the projection of future climate under

different emissions scenarios. Because climate modeling emphasizes long-term simulation of climate, treatment of aerosol processes in climate models must be greatly simplified. In contrast, with CTMs it is possible to treat aerosol processes and interactions between aerosols (and hydrometeors) and atmospheric chemistry in greater detail. CTMs are often driven by observed meteorology; in such models, the aerosol chemistry and physics do not feed back on the meteorology. CTMs and global climate models need to be driven by observed meteorology to capture detailed aerosol processes and to compare simulated aerosol fields with observations.

Since the pioneering study by Langner and Rodhe (1991), who used a coarse horizontal resolution CTM based on climatological meteorology to represent the global distribution of the mass concentration of sulfate aerosol (without explicit representation of aerosol microphysics), substantial advances have been made in the complexity of treatment of many key processes: aerosol precursor chemistry, aerosol microphysical processes, transport processes, and particle dry and wet deposition. Attempts have recently been undertaken to calculate the aerosol mass concentration as well as the particle number concentration by parameterizing aerosol formation and dynamic processes (e.g., Easter et al. 2004; Stier et al. 2005). An overview of the processes which must be understood and represented in models is given in Figure 23.3.

Most of the earlier studies concerned with the effect of aerosol particles on the climate system took only sulfate particles into account or considered sulfate to be a surrogate for all anthropogenic aerosols. Lately, most major global climate models include also carbonaceous particles, dust, and sea salt (for a synopsis of the state of model development, see Kinne et al. 2006 and the AeroCom model intercomparison project[1]). AeroCom has enabled a comparison of the results of aerosol simulations from more than a dozen modeling groups worldwide. Figure 23.4 provides an example of a comparison for global and annual mean aerosol optical depth and the vertical integral of aerosol extinction coefficient. Although fairly good agreement is demonstrated for most models, it is clear that there are substantial differences in the contributions of the several aerosol species.

A major source of uncertainty in present aerosol modeling is the lack of accurate time-resolved emission inventories. In particular, biogenic sources and emissions from biomass burning are highly uncertain. Both biogenic and biomass burning emissions depend on environmental conditions (e.g., weather) and exhibit high interannual variability, which has not been taken into account by climate studies. Probably the largest uncertainty is associated with organic aerosols, because current measurement techniques cannot identify the many organic species present in primary emissions (Kanakidou et al. 2004). A second issue is that the chemical pathways in the atmosphere are complex and not fully understood. Organic particles result both from primary emission and from

[1] http://nansen.ipsl.jussieu.fr/AEROCOM/

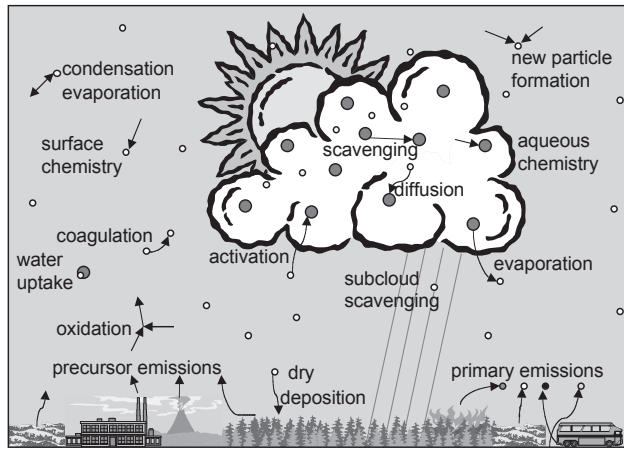

Figure 23.3 Important climate-influencing aerosol processes that must be accurately represented in climate models. Aerosol particles are directly emitted as primary particles and are formed secondarily by oxidation of emitted gaseous precursors. The formation of low-volatility materials results in new particle formation and condensation onto existing particles. Aqueous-phase oxidation of gas-phase precursors within cloud droplets accretes additional mass onto existing particles but does not form new particles. Particles age by surface chemistry, coagulation and condensation. The uptake of water with increasing relative humidity increases particle size, which affects the particle optical properties as well. As clouds form, some fraction of aerosol particles are "activated" to produce cloud droplets. Within clouds, interstitial particles can become attached to cloud droplets by diffusion, and activated particles are combined when cloud droplets collide and coalesce. If cloud droplets evaporate, the particles remain in the atmosphere; if the cloud precipitates, the particles are carried below the cloud to the surface, unless the precipitating particles evaporate completely. Aerosol particles below precipitating clouds can also be removed from the atmosphere through impaction by precipitating drops or through dry deposition to the surface (from Ghan and Schwartz 2007).

gas-to-particle conversion in the atmosphere (secondary production). The total source of these organic particles is therefore a major wildcard in simulations of future scenarios. Advances in measurement techniques for particles are thus of critical importance; one such recent advance is the aerosol mass spectrometer, which permits the development of a systematic measurement database to be developed of general aerosol composition and the identification of primary and secondary organic species (Zhang et al. 2007). Simulating nitrate particles remains problematic because of their semi-volatile nature. In addition to all of the difficulties that exist in developing an understanding of the chemical and microphysical processes, the simulation of aerosol processes in large-scale models is very CPU-time consuming.

There is increasing evidence that individual aerosol particles consist predominantly of a conglomerate of multiple internally mixed chemical substances. In contrast, most global climate models treat aerosols as external mixtures, because internal mixtures have more degrees of freedom, are more complex,

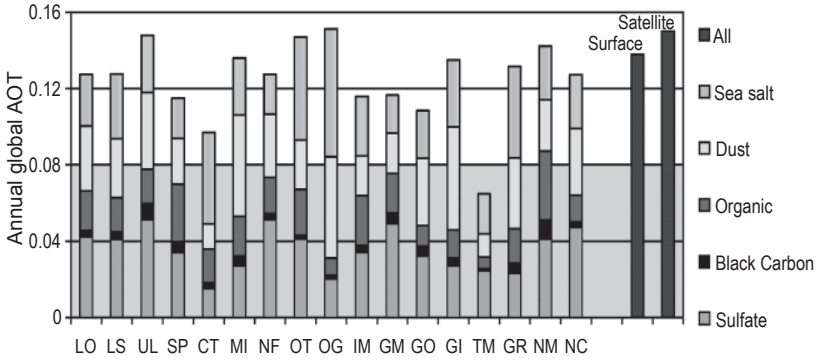

Figure 23.4 Simulated contributions of five aerosol components (seasalt, dust, organic, black carbon, and sulfate) to annual and global mean aerosol optical thickness (AOT), at 550 nm by 17 chemical transport models. For comparison, surface measurements taken by the AERONET network and a composite of satellite measurements are shown. Modified from Kinne et al. (2006).

and impose an additional computational burden. However, the mixing state of aerosol particles (externally vs. internally mixed) influences strongly their optical properties and ability to act as CCN. For example, a slight coating of a particle by only a moderately soluble organic species can drastically increase its ability to act as a CCN. Therefore, treating the degree of mixing properly is essential to represent accurately aerosol processing in global climate models, including aerosol–cloud interactions. Advanced aerosol modules in some global climate models have been expanded to include aerosol mixtures (see Lohmann and Feichter 2005 for references).

Representing the particle size distributions of aerosols and their evolution is also essential. Two kinds of aerosol dynamics models have been developed: modal schemes and bin schemes. Modal schemes represent each aerosol mode with a log-normal distribution of aerosol mass and possibly number. Bin schemes divide the aerosol spectrum into a large number of bins. Typically, modal representations of the aerosol size distribution evolve aerosol number concentration as well as mass concentration.

Representation of Cloud Microphysical Processes in Global Climate Models

Microphysics of Large-scale Clouds

Major improvements have recently been made in the description of cloud microphysics for large-scale models. Although early studies diagnosed cloud amount based on relative humidity, most current global climate models explicitly calculate cloud condensate in large-scale clouds. The degree

of sophistication varies from calculating the sum of cloud water and ice to calculating cloud water, cloud ice, snow, and rain as separate species (Lohmann and Feichter 2005). Because the aerosol indirect effect is based on the change in cloud droplet number concentration, some global climate models calculate explicitly cloud droplet number concentrations in addition to the cloud water mass mixing ratio using one of the above described physically based aerosol activation schemes as a source term for cloud droplets. Similarly, the number of ice crystals needs to be calculated in addition to the ice water mass mixing ratio to estimate the effect of aerosols on mixed-phase and ice clouds. Determining the size-dependent sedimentation rate of hydrometeors requires at least a two-moment scheme. Representing size-dependent sedimentation leads to important differences in the cloud vertical structure, cloud lifetime, and cloud optical properties. Two-moment schemes are superior to one-moment schemes provided that the second moment can be treated adequately. Major uncertainties remain, however, in terms of cloud droplet activation and precipitation formation, as discussed below. Theoretically, the best approach would be to use a size-resolved treatment of the cloud microphysics. However, using this approach in a global climate model would be questionable, because treatment of cloud dynamics, including entrainment and advection, is not accurate enough to warrant this level of detail.

Microphysics of Convective Clouds

There is currently a substantial discrepancy between the degree of sophistication in cloud microphysics in large-scale clouds and the very rudimentary treatment of cloud microphysics in convective clouds. This may reflect the fact that stratiform clouds are generally much more susceptible to indirect effects than convective clouds. Recently, however, evidence has emerged to show that biomass burning may affect convective clouds, thus necessitating improvements in the treatment of microphysical processes in convective clouds. In the first global study, Nober et al. (2003) accounted for this effect by decreasing the precipitation efficiency for warm cloud formation in convective clouds, and making it dependent on the cloud droplet number concentration. This approach was taken a step further by Lohmann (2007), who introduced the same microphysical processes (e.g., nucleation, autoconversion, freezing, aggregation) considered in large-scale clouds into convective clouds.

Another option, though considerably more computationally intensive, is to use so-called "super-parameterizations," in which cloud-resolving models (CRMs) are embedded within the normal GCM grid cells, but at only a small fraction of the area of the parent GCM grid cell (e.g., Randall et al. 2003). These models have the capability of calculating cloud-scale vertical velocities and liquid water content (LWC) and thus represent explicitly precipitation processes. They have yet, however, to be applied to aerosol effects on precipitation. If the representation of aerosol and clouds can be improved in such models,

or in others through new and innovative techniques for representing subgrid processes, this should increase the accuracy of calculations of the influence of aerosols on the amount and distribution of clouds and precipitation as well as on radiation.

Cloud Droplet Formation

Linking aerosol particles to cloud droplets is a weak point in estimates of the indirect aerosol effects. Accurate treatment of cloud droplet formation requires knowledge of the particle number concentration and size-distributed chemical composition of the aerosol and the vertical velocity on the cloud scale. Parameterizations based on the Köhler theory (Köhler 1923) have been developed to describe cloud droplet formation for a multimodal aerosol. This approach has been extended to include kinetic effects that consider mass accommodation of water at the gas–liquid interface and account for the fact that the largest particles may not have time to grow to their equilibrium size and activate. Competition between natural and anthropogenic aerosol particles, such as between sulfate and sea salt, is also considered (Forster et al. 2007).

Organic carbon is an important constituent of CCN, especially if it is surface active. Facchini et al. (1999) indicate that by lowering the surface tension of surface-active organic particles (e.g., obtained from fog water samples) the cloud droplet number concentration and cloud albedo can be enhanced, leading to a negative forcing as large as ~ -1 W m^{-2}. In contrast, amphiphilic film-forming compounds may retard cloud droplet formation (Feingold and Chuang 2002). Delayed activation enables the growth of larger drops, which have formed earlier, and results in increased dispersion and enhanced drizzle formation. Chemical effects on cloud droplet formation, and thus on the indirect effect, may be as large as the effects of unresolved cloud dynamics (Lohmann and Feichter 2005). Whereas the effect of surface-active organics has recently been included in parameterizations of cloud droplet formation (Abdul-Razzak and Ghan 2004), other effects of organics, such as their film-forming ability have not yet been treated.

Application of parameterizations of cloud drop activation requires estimating cloud-scale vertical velocities in models which do not resolve these cloud scales. Recognizing that this information may not be available, some modelers assume an empirical relationship between modeled sulfate mass concentrations and droplet concentrations (e.g., Boucher and Lohmann 1995), which is equivalent to assuming there is only one single value of cloud updraft velocity for all clouds in the model. Others estimate vertical velocity based on turbulent kinetic energy calculated in boundary layer models (e.g., Lohmann et al. 1999). The latter represents a step in the right direction, but it does not account for the fact that cloudy updrafts are at the tail of the probability density function (PDF) of vertical velocity. Ghan et al. (1997), among others, assumed a normal distribution of vertical velocity with a mean value given by the

GCM grid point mean. They determined the velocity-weighted mean droplet concentration, taking into account the tails of their assumed PDF of vertical velocity. However, observed PDFs of vertical velocity in clouds in the boundary layer are multimodal and are better represented by double-Gaussian PDFs (Larson et al. 2001) with a mean that is a function of the root mean square vertical velocity rather than by a GCM grid point mean (Peng et al. 2005).

Precipitation Formation in Warm Clouds

The influences of precipitation and drizzle processes on cloud lifetime, cloud water content, and cloud radiative properties discussed above cannot be simulated well in current GCM cloud parameterization schemes. For example, the autoconversion rate, which is the rate at which cloud droplets collide and coalesce with each other to form precipitation size drops, is a nonlinear function of the total water condensate. Thus, the mean LWC from a GCM model grid box is essentially meaningless for the representation of precipitation production (e.g., Pincus and Klein 2000). Since the autoconversion bias attributable to horizontal heterogeneity has been found to scale strongly with cloud fractional coverage (Wood et al. 2002), it may be overcome using a parameterization that takes this bias into account. Alternatively, a PDF approach to subgrid modeling may be better in resolving these deficiencies. PDFs of subgrid quantities, such as vertical velocity and liquid water path, are determined from prescribed basis functions in which various moments of the basis functions are calculated in the models (e.g., Pincus and Klein 2000).

Autoconversion of cloud droplets to rain drops is a key process governing the amount and lifetime of clouds in the atmosphere, and must be represented accurately in models from the cloud-resolving to global scale. Even though the mass transfer rate of cloud drops to rain is dominated by accretion in most clouds (Wood 2005), autoconversion is the dominant process in most GCMs because rain is assumed to reach the surface within one model time step. Thus, developing parameterizations for autoconversion suitable for incorporation in large-scale models is an active area of research. Traditional parameterizations are either empirically or intuitively obtained (e.g., Kessler 1969 and Sundqvist 1978) or are derived by curve-fitting detailed microphysical models with simple functions, such as a power law (e.g., Berry 1968; Beheng 1994). These parameterizations lack, however, clear physical bases and have arbitrarily tunable parameters. Furthermore, parameterizations that calculate at least the cloud droplet number concentration in addition to the LWC would be expected to provide much better representation of cloud radiative influences and aerosol effects than existing one-moment schemes.

One promising scheme, which has been derived from theoretical considerations (see Liu et al. 2007 and earlier papers referenced therein), represents the autoconversion rate as the product of a rate function based on the collection efficiency of falling rain drops that describes the conversion rate after the

onset of the autoconversion process times a threshold function. The threshold function, unlike that of earlier parameterizations such as the widely used Kessler (1969) parameterization, does not increase abruptly at a critical value of mean droplet mass but instead increases gradually over a range of mean droplet masses that is dependent on the relative dispersion of the cloud droplet size distribution (ratio of standard deviation to mean radius). This dependence captures initiation of the autoconversion by large drops at the high end of the size distribution; its variation for differing values of relative dispersion encompasses prior empirical representations of threshold behavior. This approach yields a strong dependence of autoconversion rate on relative dispersion; for example, for liquid water volume content 0.3 g m^{-3} and cloud droplet number concentration 50 cm^{-3}, as the relative dispersion increases from 0.33 to 1 the characteristic time of autoconversion decreases from 10 hours to 0.1 hour. This parameterization has found application in modeling on regional (Gustafson et al. 2007) and global scales (Rotstayn and Liu 2005), modeling scavenging of soluble gases by precipitation (Garrett et al. 2006), and remote sensing of precipitation (Berg et al. 2006).

Aerosol Indirect Effects

Aerosol particles affect radiative fluxes by scattering solar radiation and absorbing solar and thermal radiation (direct effect). In addition, they interact with clouds and the hydrological cycle by acting as CCN and ice nuclei. For a given cloud LWC, a greater concentration of CCN increases cloud albedo (indirect cloud albedo effect) and is supposed to reduce the precipitation efficiency (indirect cloud lifetime effect), both of which are likely to result in a reduction of the global, annual mean net radiation at the top of the atmosphere (TOA). These effects may be partly offset through the evaporation of cloud droplets attributable to absorbing aerosols (semi-direct effect) and/or by more ice nuclei (glaciation effect). The influences of these processes on radiation at TOA and at the surface and on precipitation are summarized in Table 23.1. The following discussion is based on Denman et al. (2007), which also provides references to the studies noted.

Another aerosol influence on clouds and radiation that may be climatologically important is the enhancement of downwelling longwave radiation from Arctic haze (Blanchet and Girard 1994) and thin Arctic stratus whose longwave optical thickness is augmented by increased droplet concentration (Lubin and Vogelmann 2006).

In addition to raising the number concentration of aerosol particles, there is evidence that increased particle concentrations can broaden the cloud drop size distribution (Liu and Daum 2002). This would have the effect of decreasing aerosol influences on shortwave radiation and inhibiting precipitation development (e.g., Peng and Lohmann 2003).

Table 23.1 Overview of known aerosol indirect effects on net radiative flux (at TOA and at the surface) and on precipitation, and an assessment of level of current scientific understanding. Modified from Denman et al. (2007).

Effect	Cloud albedo effect	Cloud lifetime effect	Semi-direct effect	Glaciation indirect effect	Thermodynamic effect
Cloud types affected	All; greatest for clouds of intermediate optical thickness	All	All	Mixed-phase	Mixed-phase
Process	For same cloud water or ice content more but smaller cloud particles reflect more solar radiation	Smaller cloud particles decrease precipitation efficiency prolonging cloud lifetime	Absorption of solar radiation by absorbing aerosols evaporates cloud particles, increases static stability	Increase in ice nuclei increases precipitation efficiency	Smaller cloud droplets delay freezing and cause supercooled clouds to extend to colder temperatures
Change in net TOA irradiation	–	–	+/–	+	+/–
Potential magnitude	medium	medium	small	medium	medium
Scientific understanding	low	very low	very low	very low	very low
Change in surface irradiation	–	–	+/–	+	+/–
Potential magnitude	medium	medium	large	medium	medium
Scientific understanding	low	very low	very low	very low	very low
Change in precipitation	N/A	–	–	+	+/–
Potential magnitude	N/A	small	large	medium	medium
Scientific understanding	N/A	very low	very low	very low	very low

The increase in albedo of liquid water clouds attributable to anthropogenic aerosols has received much attention. Although uncertainties remain regarding the breadth of the cloud drop size distribution, more and probably larger uncertainties are related to aerosol effects on precipitation as well as on mixed- and ice-phase clouds, as discussed below.

Aerosol Effects on Water Clouds and Warm Precipitation

Aerosol particles are hypothesized to lengthen the lifetime of clouds because increased concentrations of smaller droplets lead to decreased drizzle production and reduced precipitation efficiency (Albrecht 1989). It is difficult to devise observational studies that can separate the cloud lifetime from the cloud albedo effect. Thus, observational studies provide estimates of the combined effects. Similarly, climate models cannot easily separate the cloud lifetime indirect effect once the aerosol scheme is fully coupled to a cloud microphysics scheme. Instead, they calculate the combined cloud albedo, lifetime, and semidirect effect.

GCM studies suggest that, in the absence of giant CCN and aerosol-induced changes in ice microphysics, anthropogenic aerosols suppress precipitation. It should be noted, however, that precipitation would also be suppressed in mixed-phase clouds in which the ice phase plays only a minor role. A decrease in the formation of precipitation leads to increased cloud processing of aerosols. CRM studies have shown that cloud processing can lead to either an increase or a decrease in precipitation in subsequent cloud cycles, depending on the size and concentration of activated CCN (e.g., Feingold and Kreidenweis 2002). When the actual cloud lifetime is analyzed in CRM simulations, an increase in aerosol concentration—from very clean to strongly anthropogenically influenced situations—does not increase cloud lifetime, even though precipitation is suppressed (Jiang et al. 2006). This results from competition between precipitation suppression and enhanced evaporation of the more numerous smaller cloud droplets at high cloud droplet concentration. Giant sea-salt nuclei, on the other hand, may override the precipitation suppression effect of the large number of small CCN.

Aerosol Impacts on Mixed-phase Clouds

GCM studies suggest that if, in addition to mineral dust, hydrophilic black carbon particles are assumed to act as ice nuclei at temperatures between 0° and –35°C, then increases in aerosol number concentration from preindustrial to present times may have resulted in greater glaciation of supercooled stratiform clouds and an increase in the amount of precipitation via the ice phase. This process could decrease the global mean cloud cover, leading to enhanced absorption of solar radiation. Whether the glaciation effect or the

warm cloud lifetime effect is larger depends on the chemical nature of the particles (Lohmann and Diehl 2006).

Simulations of precipitation from single-cell mixed-phase convective clouds suggest a reduction for various background aerosol concentrations when particle concentrations are increased. Khain et al. (2005) postulated that smaller cloud droplets, such as those affected by human activities, would change the thermodynamics of convective clouds. More but smaller droplets would reduce the production of rain in convective clouds. When these droplets freeze, the associated latent heat release would then result in more vigorous convection and more precipitation. In a clean cloud, on the other hand, rain would have depleted the cloud so that less latent heat is released when the cloud glaciates, resulting in less vigorous convection and less precipitation. For a thunderstorm in Florida, in the presence of Saharan dust, the simulated precipitation enhancement lasted only two hours, after which precipitation decreased as compared with clean conditions. This highlights the complexity of the system and indicates that the sign of the global change in precipitation attributable to aerosols is not yet known. Note that microphysical processes can only change the temporal and spatial distribution of precipitation, whereas the total amount of precipitation can only change if evaporation from the surface changes.

Subgrid-scale Variability and Radiative Transfer

Model inaccuracy can result from treating clouds as simple parallel homogeneous clouds. This would be the case if clouds are uniformly distributed over grid cells, as is conventional. Such treatment can overestimate the Twomey effect by up to 50% (Lohmann and Feichter 2005). One way to obviate this problem is to treat grid cells as being nonuniform. A PDF-based approach can be used here to account for subgrid-scale variability in cloud cover and cloud condensate in radiative transfer through inhomogeneous cloud fields (e.g., Pincus and Klein 2000).

Aerosol Impacts on Cirrus Clouds

The influence of aerosols from aircraft emissions on cirrus cloud extent and properties has received considerable attention because these particles are emitted at an altitude where clouds can exert a strong radiative influence. This subject is examined by Kärcher and Spichtinger (this volume) and Denman et al. (2007) and is thus not reviewed here.

Global Climate Model Estimates of the Total Anthropogenic Aerosol Effect

The total anthropogenic aerosol effect, as defined here, consists of the direct effect, semi-direct effect, indirect cloud albedo effect, and cloud lifetime effect for warm clouds. The total anthropogenic aerosol effect is obtained by calculating the difference between a multiyear simulation with present-day aerosol emissions and a simulation representative for preindustrial conditions, in which anthropogenic emissions are turned off. It should be noted that the representation of the cloud lifetime effect in global climate models is essentially one of changing the autoconversion of cloud water to rainwater.

The radiative forcing that results from the indirect cloud albedo effect attributable to anthropogenic aerosols is estimated from global models as –0.7 W m^{-2}, with a 90% confidence range of –0.3 to –1.8 W m^{-2} (Forster et al. 2007). Feedbacks that result from the cloud lifetime effect, semi-direct effect, or aerosol–ice cloud effects can either enhance or reduce the cloud albedo effect. Climate models estimate the total aerosol effect (direct plus indirect effects) on the TOA net radiation since preindustrial times to be –1.2 W m^{-2}, with a range of –0.2 to –2.3 W m^{-2} (Figure 23.5 and Denman et al. 2007). The range of the total aerosol effect from different models cannot easily be compared to the range of the indirect cloud albedo effect alone because different model simulations entered these various compilations.

All models agree that the total aerosol effect is larger over the northern hemisphere than over the southern hemisphere (Figure 23.5), consistent with emissions of anthropogenic aerosols and precursor gases being much greater in the northern hemisphere. This effect has not been seen, however, in satellite data (Han et al. 1998; Schwartz 1988), suggesting that either dynamic influences on the liquid water path mask such an effect or that the models do not represent aerosol–cloud interactions realistically. The values of the northern hemisphere total aerosol effect vary between –0.5 and –3.6 W m^{-2}; in the southern hemisphere they range between slightly positive to –1.1 W m^{-2}; and the average southern/northern hemisphere ratio is 0.3. Estimates of the ocean/land partitioning of the total aerosol effect vary from 0.03 to 1.8, with an average value of 0.7. Although the combination of ECHAM4 model results with POLDER satellite estimates suggests that the total aerosol effect should be larger over oceans, combined estimates of the LMD and ECHAM4 models with MODIS satellite data reach the opposite conclusion. The average total aerosol effect over the ocean of –1 W m^{-2} agrees with estimates between –1 to –1.6 W m^{-2} from AVHRR/POLDER (Denman et al. 2007).

Estimates of the total aerosol effect from global climate models are generally larger than those estimated from inverse approaches, which constrain the indirect aerosol effect to be between –0.1 and –1.7 W m^{-2} (Anderson et al. 2003; Hegerl et al. 2007). The estimated total anthropogenic aerosol effect is now lower than was stipulated in IPCC's Third Assessment Report and

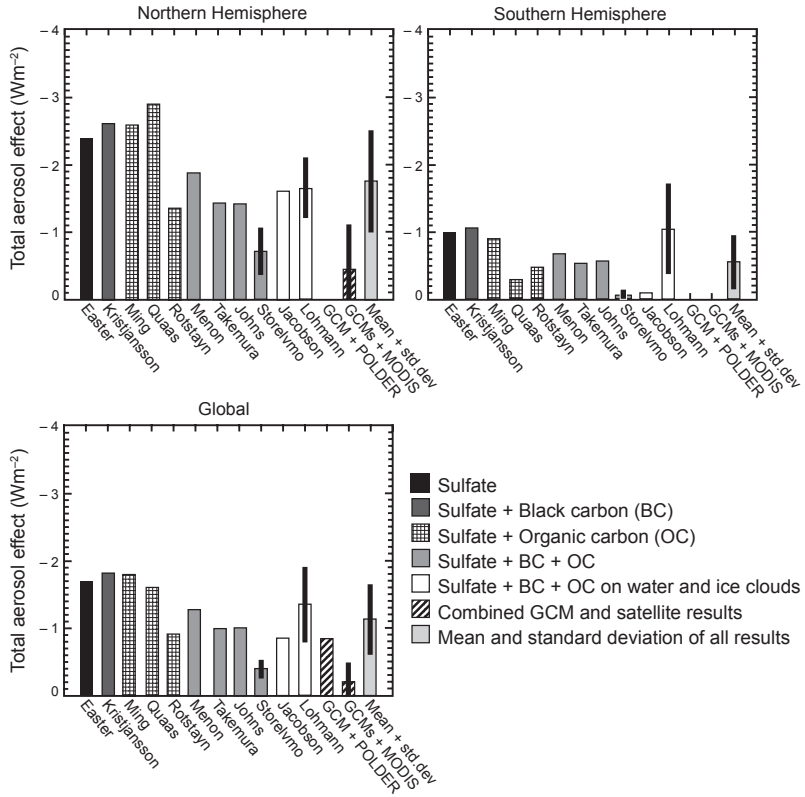

Figure 23.5 Total anthropogenic aerosol effect (direct, semi-direct and indirect cloud albedo and lifetime effects) in 12 global climate models and two determinations from satellite observations in global mean, over the northern and southern hemispheres, over oceans and over land, and the ratio over oceans/land. Anthropogenic aerosol effect is defined as the change in net radiation at TOA from preindustrial times to the present day resulting from anthropogenic emissions of aerosols and aerosol precursors. Patterns denote different anthropogenic species whose forcings were examined and the cloud types affected; all are for water clouds except as indicated.

this is attributable to improvements in cloud parameterizations. Still, large uncertainties remain.

The influence of aerosols on evapotranspiration and precipitation is also quite uncertain, with model results for the change in global average precipitation ranging from almost no change to a decrease of 0.13 mm day^{-1} (5 cm yr^{-1}), with much greater changes locally. Decreases in precipitation are larger when the atmospheric GCMs are coupled to mixed-layer ocean models, where the sea surface temperature and, hence, the evaporation from the ocean is also allowed to vary, than in models in which the sea surface temperature is held constant (Denman et al. 2007). The decrease in evapotranspiration results primarily from decreases in solar radiation at the surface, as a result of increased

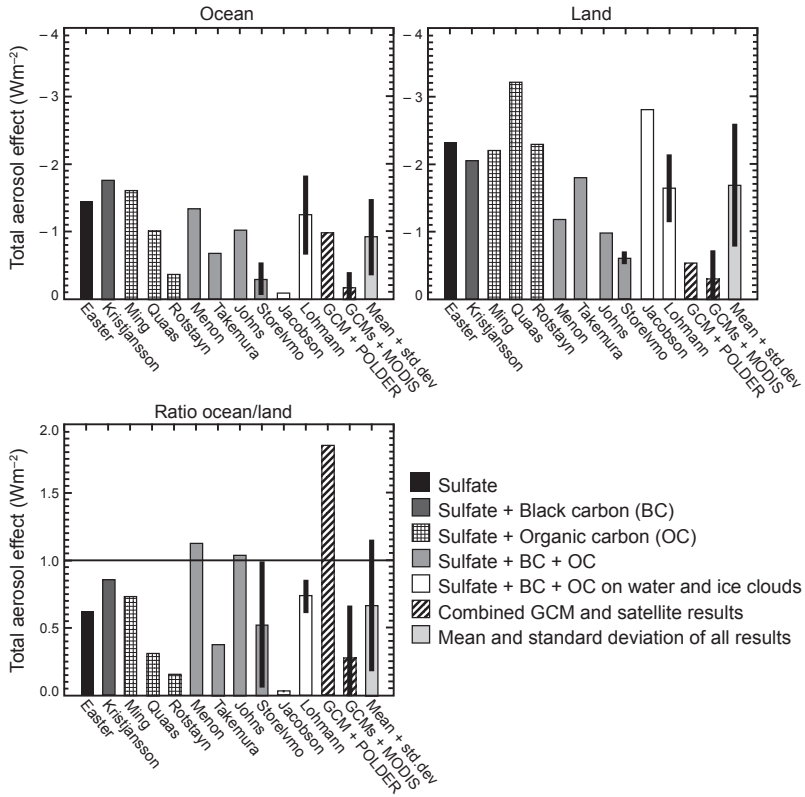

Figure 23.5 (continued) Vertical black lines denote ±1 standard deviation in cases of multiple simulations and/or results. Modified after Denman et al. (2007).Vertical black lines denote ±1 standard deviation in cases of multiple simulations and/or results. Modified after Denman et al. (2007).

aerosol optical depth and optically thicker clouds. The decrease in solar radiation at the surface is then partly balanced by a decreased latent heat flux, which results in a reduced global mean precipitation rate (e.g., Liepert et al. 2004).

Cloud Feedbacks in Climate Models and Their Influence on Climate Sensitivity

The energy balance model of Earth's climate system is useful to assess the influences of particular processes on global mean surface temperature (T) and to compare these influences across different climate models. Within this energy balance framework, the time-dependent change in heat content of the climate system, ΔQ, is related to radiative forcing, ΔF, and the change in T, ΔT, as:

$$\frac{d}{dt}(\Delta Q) = \Delta F - \frac{1}{\lambda}\Delta T, \tag{23.1}$$

where λ is the equilibrium climate sensitivity, as is readily seen by considering a system in a new equilibrium in response to a forcing ΔF, for which $d(\Delta Q)/dt = 0$. Hence,

$$\lambda = \frac{\Delta T_{eq}}{\Delta F}, \tag{23.2}$$

where ΔT_{eq} is the temperature difference between two equilibrium states. At present, climate sensitivity is not well constrained in climate models or in empirical analyses. According to the IPCC's Fourth Assessment Report (IPCC 2007), the likely range of global equilibrium temperature increase for the doubling of CO_2, ΔT_{2x}, lies between 2.0 and 4.5 K; values below 1.5 K are considered very unlikely. This sensitivity range is in agreement with values exhibited by current climate models (Figure 23.6). Since a doubling of CO_2 causes direct radiative forcing of about 3.7 W m^{-2} (Forster et al. 2007), the range of 2.0–4.5 K for a doubling of CO_2 corresponds to climate sensitivity between 0.54 and 1.22 K/(W m^{-2}). Roe and Baker (2007) point out that the upper range of the climate sensitivity is relatively insensitive to decreases in uncertainties associated with the underlying climate processes. In this context, we note that some recent experiments with CRMs embedded into climate models (Miura et al. 2005; Wyant et al. 2006) suggest a lower climate sensitivity, with values of 0.44 and 0.41 K/(W m^{-2}), respectively (ΔT_{2x} 1.6 and 1.5 K). These findings point to the strong influence of the treatment of clouds on modeled climate sensitivity.

Cloud feedback is the response of CRF to a change in global temperature. Key questions involve the nature and extent of CRF changes in a greenhouse-warmed world: As the climate warms, will longwave CRF increase (positive feedback) or decrease (negative feedback)? Similarly, will shortwave CRF increase (negative feedback) or decrease (positive feedback)? Current climate models produce a wide variety of cloud feedbacks ranging from weakly negative to strongly positive, depending on the relative magnitude of different cloud feedback mechanisms (Bony et al. 2006; Webb et al. 2006). As seen in Figure 23.6, much of the model-to-model difference in climate sensitivity results from differences in cloud feedback.

CRF depends largely on the spatial and temporal distribution of clouds and their radiative properties, which are determined by their microphysical characteristics such as size distribution of droplets and ice crystals. These cloud properties are controlled by cloud-forming processes (e.g., cooling through rising air or radiative cooling) and by cloud-dissipating processes (e.g., precipitation, sinking motion, and mixing with dry air). Thus, cloud feedbacks involve changes in the spatial distribution of clouds and their microphysical properties that result from alterations in processes that form and dissipate

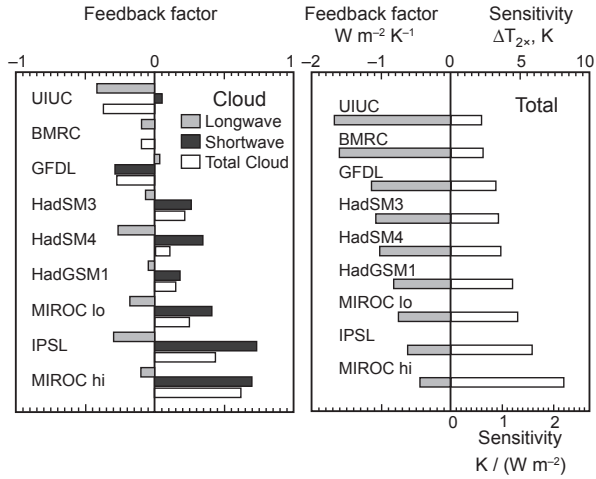

Figure 23.6 Influence of cloud feedback on total feedback and sensitivity of current GCMs. Left panel shows total cloud feedback and long- and shortwave components in nine current climate models. Right panel shows total feedback factor and sensitivity expressed as the inverse of the total feedback factor, in units of K/(W m^{-2}), and as the equilibrium increase in global mean surface temperature that would result from a doubling of CO_2, $\Delta T_{2\times}$, evaluated as –3.7 W m^{-2} upon the total feedback factor. Modified from Webb et al. (2006).

clouds. Uncertainty arises because it is not clear how cloud microphysical properties and cloud horizontal/vertical distributions respond to alterations in controlling variables as the climate changes. In contrast to CRF, cloud feedback cannot be measured directly; it can only be determined from GCM simulations. Consequently, confidence in estimates of cloud feedback can only be assessed by using observations to evaluate simulations of cloud microphysical and radiative properties, cloud distribution, and radiative forcing in a variety of conditions that span the range expected under climate change. Therefore, atmospheric process research on cloud feedbacks focuses on how these cloud features depend on processes that form and dissipate clouds under a variety of conditions.

Closely related to cloud feedback is water-vapor feedback. Water vapor is the most important greenhouse gas in Earth's atmosphere. Consistent with basic physics, all climate models show an increase in the amount of atmospheric water vapor with rising global mean surface temperature. However, the amount and spatial distribution of the resultant radiative forcing differ considerably. This water-vapor feedback is strongly connected to the cloud feedback because water vapor is the source of condensed phase water in clouds, and clouds remove water from the atmosphere when they precipitate. Clouds also influence evapotranspiration, which is the source of water vapor to the atmosphere.

Conclusions and Outlook

Clouds play a crucial role in determining Earth's energy balance. This must be accurately represented in climate models, if the models are to be used with confidence to project future climate change. Much of the variation in climate sensitivity observed in current climate models can be attributed to differences in the treatment of clouds, as evidenced by model-to-model variation in cloud feedback. Microphysical processes are now recognized to exert strong influences on cloud dynamics and that the influences of clouds on short- and longwave radiation must be understood and accurately represented in climate models. In addition, the strong coupling between aerosols and clouds has large influences on climate and climate change. An increased atmospheric aerosol burden alters the microphysical properties of clouds and influences short- and longwave radiation and the locus and intensity of precipitation.

One of the better understood influences of aerosols on clouds is a reduction of the amount of solar radiation absorbed by Earth–atmosphere system, as quantified by net shortwave radiation at the TOA and a similar decrease in shortwave radiation reaching the surface. The negative radiative forcing of anthropogenic aerosols competes with greenhouse gas warming as a forcing of climate change and in altering evaporation and precipitation. Although much has been learned about these effects, they are not understood be well enough to be fully represented in climate models. None of the transient climate model simulations conducted thus far accounts for all of the known aerosol–cloud interactions; thus the net effects of aerosols on clouds and climate deduced from global climate models cannot be considered conclusive. Therefore, the cloud feedback and sensitivity of Earth's climate system remain highly uncertain. One reason is that aerosol–cloud interactions take place on microscale and thus are at best crudely represented in GCMs.

In terms of aerosols and clouds, the principal areas for future development in global climate models are twofold. The treatment of clouds themselves requires improvement in all aspects, if models are to represent accurately cloud feedbacks in a greenhouse-warmed world. A good representation of cloud dynamics, including entrainment, is especially important for the representation of convective clouds and boundary layer clouds. Cloud microphysical processes are important for the conversion of cloud particles into precipitation-size particles. The treatment of aerosol–cloud interactions needs to be improved as well, if aerosol radiative forcing is to be accurately quantified.

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References

Abdul-Razzak, H., and S. J. Ghan. 2004. Parameterization of the influence of organic surfactants on aerosol activation, *J. Geophys. Res*. **109**:D03205.

Albrecht, B. 1989. Aerosols, cloud microphysics, and fractional cloudiness. *Science* **245**:1227–1230.

Anderson, T. L., R. J. Charlson, S. E. Schwartz et al. 2003. Climate forcing by aerosols: A hazy picture. *Science* **300**:1103–1104.

Beheng, K. D. 1994. A parameterization of warm cloud microphysical conversion processes. *Atmos. Res*. **33**:193–206.

Bender, F. A. M., H. Rodhe, R. J. Charlson, A. M. L. Ekman, and N. Loeb. 2006. 22 views of the global albedo: Comparison between 20 GCMs and two satellites. *Tellus* **58**:320–330.

Berg, W., T. L'Ecuyer, and C. Kummerow. 2006. Rainfall climate regimes: The relationship of regional TRMM rainfall biases to the environment. *J. Appl. Meteor. Climatol*. **45(3)**:434.

Berry, E. X. 1968. Modification of the warm-rain process. Proc. 1st. Natl. Conf. Weather Modification, pp. 81–88. Albany: *Amer. Meteor. Soc*.

Blanchet, J.-P., and E. Girard. 1994. Arctic greenhouse effect. *Nature* **371(6496)**:383.

Bony, S., R. Colman, V. M. Kattsov et al. 2006. How well do we understand and evaluate climate change feedback processes? *J. Climate* **19**:3445–3482.

Boucher, O., and U. Lohmann. 1995. The sulfate-CCN-cloud albedo effect: A sensitivity study using two general circulation models. *Tellus* **47B**:281–300.

Denman, K. L., G. Brasseur, A. Chidthaisong et al. 2007. Couplings between changes in the climate system and biogeochemistry. In: Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the IPCC, ed. Solomon, S., D. Qin, M. Manning et al., pp. 499–588. New York. Cambridge Univ. Press.

Easter, R. C., S. J. Ghan, Y. Zhang et al. 2004. MIRAGE: Model description and evaluation of aerosols and trace gases. *J. Geophys. Res.* **109**:D20210.

Facchini, M. C., M. Mircea, S. Fuzzi, and R. J. Charlson. 1999. Cloud albedo enhancement by surface-active organic solutes in growing droplets. *Nature* **401**:257–259.

Feingold, G., and P. Y. Chuang. 2002. Analysis of the influence of film-forming compounds on droplet growth: Implications for cloud microphysical processes and climate. *J. Atmos. Sci*. **59**:2006–2018.

Feingold, G., and S. M. Kreidenweis. 2002. Cloud processing of aerosol as modeled by a large eddy simulation with coupled microphysics and aqueous chemistry. *J. Geophys. Res.* **107(D23)**:4687.

Forster, P., V. Ramaswamy, P. Artaxo et al. 2007. Radiative forcing of climate change. In: Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the IPCC, ed. by S. Solomon, D. Qin, M. Manning et al., pp. 129–234. New York: Cambridge Univ. Press.

Garrett, T. J., L. Avey, P. I. Palmer et al. 2006. Quantifying wet scavenging processes in aircraft observations of nitric acid and cloud condensation nuclei. *J. Geophys. Res.* **111**:D23S51.

Ghan, S. J., L. R. Leung, R. C. Easter, and H. Abdul-Razzak. 1997. Prediction of cloud droplet number in a general circulation model. *J. Geophys. Res.* **102**:21,777–21,794.

Ghan, S. J., and S. E. Schwartz. 2007. Aerosol properties and processes: A path from field and laboratory measurements to global climate models. *Bull. Amer. Meteor. Soc.* **88**:1059–1083.

Gustafson, W. I., Jr., E. G. Chapman, S. J. Ghan, R. C. Easter, and J. D. Fast. 2007. Impact on modeled cloud characteristics due to simplified treatment of uniform cloud condensation nuclei during NEAQS 2004. *Geophys. Res. Lett.* **34**:L19809.

Han, Q., W. B. Rossow, J. Chou, and R. M. Welch. 1998. Global survey of the relationships of cloud albedo and liquid water path with droplet size using ISCCP. *J. Climate* **11**:1516–1528.

Hegerl, G. C., F. W. Zwiers, P. Braconnot et al. 2007. Understanding and attributing climate change. In: Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the IPCC, ed. S. Solomon, D. Qin, M. Manning et al., pp. 665–744. New York: Cambridge Univ. Press.

IPCC. 2007. Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, ed. S. Solomon, D. Qin, M. Manning et al. New York: Cambridge Univ. Press.

Jiang, H., H. Xue, A. Teller, G. Feingold, and Z. Levin. 2006. Aerosol effects on the lifetime of shallow cumulus. *Geophys. Res. Lett.* **33(14)**:L14806.

Kanakidou, M., J. Seinfeld, S. Pandis et al. 2004. Organic aerosol and global climate modelling: A review. *Atmos. Chem. Phys.* **4**:5855–6024.

Kessler, E. 1969. On the Distribution and Continuity of Water Substance in Atmospheric Circulation. Meteor. Monogr., vol 10. Boston: Amer. Meteor. Soc.

Khain, A. P., D. Rosenfeld, and A. Pokrovsky. 2005. Aerosol impact on the dynamics and microphysics of convective clouds. *Q. J. Roy. Meteor. Soc*. **131(611)**:2639–2663.

Kiehl, J. T., and K. E. Trenberth. 1997. Earth's annual global mean energy budget. *Bull. Amer. Meteor. Soc.* **78**:197–208.

Kinne, S. et al. 2006. An AeroCom initial assessment: Optical properties in aerosol component modules of global models. *Atmos. Chem. Phys.* **6**:1815–1834.

Köhler, H. 1923. Zur Kondensation des Wasserdampfes in der Atmosphäre, erste Mitteilung. *Geophys.* **2**:1–15.

Langner, J., and H. Rodhe. 1991. A global three-dimensional model of the global sulfur cycle. *J. Atmos. Chem*. **13**:225–263.

Larson, V. E., R. Wood, P. R. Field et al. 2001. Small-scale and mesoscale variability of scalars in cloudy boundary layers: One-dimensional probability density functions. *J. Atmos. Sci.* **58**:1978–1996.

Liepert, B. G., J. Feichter, U. Lohmann, and E. Roeckner. 2004. Can aerosols spin down the water cycle in a warmer and moister world? *Geophys. Res. Lett.* **31**:L06207.

Liu, Y., and P. H. Daum. 2002. Indirect warming effect from dispersion forcing. *Nature* **419**:580–581.

Liu, Y., P. H. Daum, R. McGraw, M. Miller, and S. Niu. 2007. Theoretical formulation for autoconversion rate of cloud droplet concentration. *Geophys. Res. Lett.* **34**:L116821.

Lohmann, U. 2007. Global anthropogenic aerosol effects on convective clouds in ECHAM5-HAM. *Atmos. Chem. Phys.* **7**:14,639–14,674.

Lohmann, U., and K. Diehl. 2006. Sensitivity studies of the importance of dust ice nuclei for the indirect aerosol effect on stratiform mixed-phase clouds. *J. Atmos. Sci.* **63**:968–982.

Lohmann, U., and J. Feichter. 2005. Global indirect aerosol effects: A review. *Atmos. Chem. Phys.* **5**:715–737.

Lohmann, U., J. Feichter, C. C. Chuang, and J. E. Penner. 1999. Predicting the number of cloud droplets in the ECHAM GCM. *J. Geophys. Res.* **104**:9169–9198.

Lubin, D., and A. M. Vogelmann. 2006. A climatologically significant aerosol longwave indirect effect in the Arctic. *Nature* **439**:453–456.

Miura, H., H. Tomita, T. Nasuno et al. 2005. A climate sensitivity test using a global cloud resolving model under an aqua planet condition. *Geophys. Res. Lett.* **32**:L19717.

Nober, F. J., H. F. Graf, and D. Rosenfeld. 2003. Sensitivity of the global circulation to the suppression of precipitation by anthropogenic aerosols. *Global Planetary Change* **37**:57–80.

Peng, Y., and U. Lohmann. 2003. Sensitivity study of the spectral dispersion of the cloud droplet size distribution on the indirect aerosol effect. *Geophys. Res. Lett.* **30(10)**:1507.

Peng, Y., U. Lohmann, and W. R. Leaitch. 2005. Importance of vertical velocity variations in the cloud droplet nucleating process of marine stratus clouds. *J. Geophys. Res.* **110**:D21213.

Pincus, R., and S. A. Klein. 2000. Unresolved spatial variability and microphysical process rates in large scale models. *J. Geophys. Res.* **105**:27,059–27,065.

Ramaswamy, V., O. Boucher, J. Haigh et al. 2001. Radiative forcing of climate change. In: Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the IPCC, ed. J. T. Houghton, Y. Ding, D.J. Griggs et al., pp. 349–416. New York: Cambridge Univ. Press.

Randall, D., M. Khairoutdinov, A. Arakawa, and W. Grabowski. 2003. Breaking the cloud parameterization deadlock. *Bull. Amer. Meteor. Soc.* **84**:1547–1564.

Randall, D. A., R. A. Wood, S. Bony et al. 2007. Climate models and their evaluation. In: Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the IPCC, ed. S. Solomon, D. Qin, M. Manning et al., pp. 591–662. New York: Cambridge Univ. Press.

Roe, G. H., and M. B. Baker. 2007. Why is climate sensitivity so unpredictable? *Science* **318**:629–632.

Rotstayn, L. D., and Y. Liu. 2005. A smaller global estimate of the second indirect aerosol effect. *Geophys. Res. Lett.* **32**:L05708.

Schwartz, S. E. 1988. Are global cloud albedo and climate controlled by marine phytoplankton? *Nature* **336**:441–445.

Stier, P., J. Feichter, S. Kinne et al. 2005: The aerosol–climate model ECHAM5–HAM. *Atmos. Chem. Phys.* **5**:1125–1156.

Sundqvist, H. 1978. A parameterization scheme for non-convective condensation including prediction of cloud water content. *Q. J. Roy. Meteor. Soc.* **104**:677–690.

Webb, M. J., C. A. Senior, D. M. H. Sexton et al. 2006. On the contribution of local feedback mechanisms to the range of climate sensitivity in two GCM ensembles. *Climate Dyn.* **27**:17–38.

Wood, R. 2005. Drizzle in stratiform boundary layer clouds. Part II: Microphysical aspects. *J. Atmos. Sci.* **62**:3034–3050.

Wood, R., P. R. Field, and W. R. Cotton. 2002. Autoconversion rate bias in stratiform boundary layer cloud parameterizations. *Atmos. Res.* **65**:109–128.

Wyant, M., M. Khairoutdinov, and C. Bretherton. 2006. Climate sensitivity and cloud response of a GCM with a superparameterization. *Geophys. Res. Lett.* **33**:L06714.

Zhang, Q., J. L. Jimenez, M.R. Canagaratna et al. 2007. Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced northern hemisphere midlatitudes. *Geophys. Res. Lett.* **34**:L13801.