Toward a Minimal Representation of Aerosols in Climate Models: Comparative Decomposition of Aerosol Direct, Semidirect, and Indirect Radiative Forcing

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ABSTRACT

The authors have decomposed the anthropogenic aerosol radiative forcing into direct contributions from each aerosol species to the planetary energy balance through absorption and scattering of solar radiation, indirect effects of anthropogenic aerosol on solar and infrared radiation through droplet and crystal nucleation on aerosol, and semidirect effects through the influence of solar absorption on the distribution of clouds. A three-mode representation of the aerosol in version 5.1 of the Community Atmosphere Model (CAM5.1) yields global annual mean radiative forcing estimates for each of these forcing mechanisms that are within 0.1 W m$^{-2}$ of estimates using a more complex seven-mode representation that distinguishes between fresh and aged black carbon and primary organic matter. Simulating fresh black carbon particles separately from internally mixed accumulation mode particles is found to be important only near fossil fuel sources. In addition to the usual large indirect effect on solar radiation, this study finds an unexpectedly large positive longwave indirect effect (because of enhanced cirrus produced by homogenous nucleation of ice crystals on anthropogenic sulfate), small shortwave and longwave semidirect effects, and a small direct effect (because of cancelation and interactions of direct effects of black carbon and sulfate). Differences between the three-mode and seven-mode versions are significantly larger (up to 0.2 W m$^{-2}$) when the hygroscopicity of primary organic matter is decreased from 0.1 to 0 and transfer of the primary carbonaceous aerosol to the accumulation mode in the seven-mode version requires more hygroscopic material coating the primary particles. Radiative forcing by cloudborne anthropogenic black carbon is only $-0.07$ W m$^{-2}$.

1. Introduction

Anthropogenic aerosol is thought to play an important role in driving climate change, but its role is so complex that uncertainty in estimates of radiative forcing of climate change is dominated by uncertainty associated with forcing by anthropogenic aerosol (Forster et al. 2007). This complexity arises because anthropogenic aerosol alters the planetary energy balance through a variety of mechanisms operating across a wide range of spatial scales: direct effects (Haywood and Boucher 2000; Myhre 2009), indirect effects (Lohmann and Feichter 2005), and semidirect effects (Hansen et al. 1997; Koch and Del Genio 2010). The term aerosol direct effects refers to the direct impact of anthropogenic aerosol particles on the planetary energy balance through scattering, absorption, and emission of radiation in the atmosphere, without consideration of the aerosol effects of the radiative heating on clouds. Aerosol indirect effects refer to the impact through the influence of anthropogenic aerosol on the optical properties of clouds by serving as the nuclei for droplets and ice crystals and thereby changing droplet and ice crystal number concentration, which changes cloud particle surface area, influences droplet collisions, and changes the accumulation of liquid water and ice in clouds, all of which affect the reflectivity and emissivity of clouds. Semi-direct effects are changes in the planetary energy balance as clouds respond to radiative heating by anthropogenic aerosols.

The radiative forcing depends on the size, composition, shape, and even internal structure of the collection of airborne particles constituting the aerosol, and these
aerosol properties result from a complex life cycle involving many processes from emissions to removal from the atmosphere. Representing this complexity in climate models is a major challenge because the radiative forcing changes with time and hence must be calculated throughout the duration of climate change simulations of a century or more. Given the many demands placed on climate models, computationally efficient representations of aerosol and essential effects of aerosol on climate are required. This requirement leads to the following question: what is the minimal level of complexity needed for a model to capture the essential effects of aerosol on climate? To answer this question we must first identify the requirements to accurately represent aerosol effects on climate. Then we can consider various approximations to determine the minimal level of complexity needed to achieve comparable accuracy.

An accurate representation of aerosol effects on climate begins with a realistic representation of the aerosol. A comprehensive representation of the aerosol would be free of any assumptions about the mixing state of the aerosol, permitting both internal and external mixtures of particles. This can be achieved in principle with a particle-resolved model (Riemer et al. 2009) but at a computational cost that is prohibitive for climate modeling. The first generation of climate models treating multiple aerosol species assumed external mixing of species, that is, each particle is composed of only one type of material (black carbon, organic carbon, sulfate, sea salt, or soil dust). A new generation of climate models takes the other extreme, assuming internal mixtures for particles of the same size. This permits representations that can be affordable for at least multiyear global simulations, and much longer if the distribution of number with size is represented efficiently. Although such an approximation can significantly bias the bulk optical and microphysical properties of the aerosol (Zaveri et al. 2010) in regions with external mixtures of fresh black carbon and other aerosol, Moffet and Prather (2009) provide evidence that aging is quite rapid, which suggests that external mixing is needed only very near black carbon sources. Although alternatives to the internal mixture assumption that distinguish between fresh primary and aged particles (Wilson et al. 2001; Vignati et al. 2004; Liu et al. 2005; Stier et al. 2005) are likely to be more accurate than the internal mixing assumption near black carbon sources, it is not clear if the added complexity of such treatments is necessary in global climate simulations.

A comprehensive representation of the aerosol would also account for the complex shape and internal structure of the particles. Fresh soot particles often resemble chained aggregates (Johnson et al. 2005) while aged particles are often composed of internal mixtures of black carbon coated with sulfates and other secondary material (Adachi and Buseck 2008). The impact of this complexity on radiative forcing has only recently been addressed (Adachi et al. 2010). Even if the particles can be considered spheres, the optical properties depend on whether the black carbon is randomly distributed within the particle or concentrated near the center (Bond et al. 2006), and these differences significantly affect the aerosol radiative forcing (Jacobson 2001). Optical properties are easier to calculate for a random distribution, but the shell-core model is more consistent with high-resolution images of aged particles.

It is well established that to simulate effects of anthropogenic aerosol on climate it is necessary to also simulate the major natural components of the aerosol (Menon et al. 2002; Kirkevag et al. 2008), including sea salt, sulfate from oxidation of dimethyl sulfide and volcanic sulfur dioxide, biogenic secondary organic aerosol, carbonaceous aerosol from wildfires, and mineral dust. Necessary anthropogenic aerosol components are sulfate, black carbon, and carbonaceous aerosol from fossil, biofuel, and biomass burning, but the necessity of predicting other components, such as nitrate and ammonium (which can be diagnosed from sulfate by assuming neutrality), is questionable. Given the variety of optical (Sokolik and Toon 1999) and cloud-nucleating (Hoosie et al. 2008) properties of materials comprising mineral dust, it might be necessary to distinguish between different mineralogy of dust particles.

The representation of the aerosol size distribution is also a key consideration. A sectional representation (Adams and Seinfeld 2002; Spracklen et al. 2005) is most accurate given a sufficient number of bins (Zhang et al. 2002) but is too expensive for representing all necessary aerosol components in century simulations. The modal method (Wilson et al. 2001; Herzog et al. 2004; Vignati et al. 2004), which predicts the number as well as the mass for each of multiple lognormal modes, offers more computational efficiency. Bulk methods prescribe the size distribution for each component (which are assumed to be externally mixed) and hence are inherently inappropriate because they do not distinguish between processes that affect number but not mass and those that affect mass but not number (Ghan et al. 2001). The quadrature method of moments (QMOM) (McGraw 1997; Wright et al. 2001; Yoon and McGraw 2004; Bauer et al. 2008) provides a computationally efficient statistical representation that makes no assumptions about the aerosol size distribution. Thus, both the modal and QMOM treatments are attractive; the remaining question is the optimal number of modes or moments for each approach.

Yet another consideration is the representation of aerosol attachment state, that is, whether the particles are
suspended in air or are attached to cloud or precipitation particles. Since wet scavenging is an important aerosol removal mechanism and aqueous chemistry is an important aerosol production mechanism, it is important to represent the aerosol attachment state realistically. A fully consistent representation would distinguish between interstitial aerosol and particles attached to cloud droplets, ice crystals, and precipitation particles, with explicit representation of the processes transferring particles between these states. But Ghan and Easter (2006) showed that for the purpose of characterizing aerosol radiative forcing it is sufficient to distinguish between interstitial aerosol and particles attached to cloud droplets, without treating transport of cloudborne particles.

Given a representation of the aerosol size distribution, mixing state, attachment state, composition, and internal structure, the next question is which processes in the aerosol life cycle need to be represented to accurately simulate the distribution and properties of the aerosol. Essential processes are primary emissions, emissions of precursor gases, oxidation of precursor gases, new particle formation, growth by condensation of oxidized precursor gases, coagulation, water uptake, activation to form cloud droplets, aqueous-phase reactions in clouds, resuspension when droplets evaporate, in-cloud and below-cloud precipitation scavenging, vertical transport by turbulence, gravitational settling, convective clouds, resolved winds, and dry deposition. Other processes that might be important are heterogeneous chemistry and oxidation of black carbon. Many choices about the representation of each of these processes must be made (Ghan and Schwartz 2007), and those choices should be consistent with the representation of the aerosol.

Given a simulated distribution of the aerosol, which mechanisms are necessary to estimate anthropogenic aerosol effects on climate? The literature on this subject is vast (Forster et al. 2007). Based on this literature, we consider absorption and scattering of solar radiation (including the subsequent effect of the absorption on clouds) and activation of the aerosol to form cloud droplets (and the subsequent effect on both droplet effective radius and cloud liquid water content for stratiform clouds) to be essential mechanisms. Aerosol effects on ice crystal nucleation might also be important (Lohmann and Feichter 2005; Hoose et al. 2008; Liu et al. 2009).

The minimum level of complexity needed to represent anthropogenic aerosol effects on climate cannot be determined and described in a single manuscript. Liu et al. (2012) describe and evaluate two representations of the aerosol, one suitable for century climate simulations, the other more detailed. In this study we focus on the sensitivity of the estimated aerosol forcing to simplifications in the representation of the aerosol in the model, with an emphasis on distinguishing between radiative forcing mechanisms (direct, semidirect, indirect). Section 2 describes the model used, including one relatively complete and one simplified representation of the aerosol. Section 3 compares the estimates of the radiative forcing by anthropogenic aerosol for the two representations and for each forcing mechanism. Conclusions are summarized in section 4.

2. Model description

The model used in this study is version 5.1 of the Community Atmosphere Model (CAM5.1). Liu et al. (2011) describe the treatment of aerosols, clouds, radiation, and turbulence in this model. Further details on CAM5.1 are available online (at http://www.cesm.ucar.edu/models/cesm1.0/cam/). For the sake of brevity here we only provide the most salient features of CAM5.1.

a. Aerosols

We have implemented two different modal representations of the aerosol in CAM5.1. For both representations the size distributions of the modes are assumed to be lognormal, with the number as well as the mass mixing ratios of internally mixed components predicted for each mode, and the width of the distributions prescribed. A seven-mode version of the modal aerosol model (MAM7) serves as a benchmark for further simplification. It includes Aitken, accumulation, primary carbon, fine dust and sea salt, and coarse dust and sea salt modes (Fig. 1). Nitrate is not included because it constitutes a significant fraction of the measured submicron aerosol mass in only limited regions and seasons (Jimenez et al. 2009); nitrate is known to be important in some industrial regions (e.g., East Asia) and could be important for radiative forcing in the future, when sulfur and carbonaceous emissions are reduced. Within each mode the mass mixing ratios of internally mixed aerosol components and the number mixing ratio of the particles are each predicted. Particles consisting of primary organic matter (POM) and black carbon (BC) are emitted to the primary carbon mode and are aged to the accumulation mode by coagulation with Aitken and accumulation modes and by condensation of H2SO4, NH3, and semivolatile organics. Cloudborne aerosols are predicted separately from interstitial aerosols. The total number of transported aerosol species is 31.

For long-term (up to multiple centuries) climate simulations a three-mode version of MAM (MAM3) has also been developed; it has only Aitken, accumulation and coarse modes, also illustrated in Fig. 1. The following assumptions are made for MAM3: 1) the primary
carbon mode is merged with the accumulation mode, so that all primary carbon is internally mixed with secondary aerosol; 2) the coarse dust and sea salt modes are merged into a single coarse mode based on the assumption that the dust and sea salt are geographically separated; 3) the fine dust and sea salt modes are both merged with the accumulation mode; and 4) sulfate is partially neutralized by ammonium in the form of ammonium bisulfate, so ammonium is effectively prescribed and NH$_3$ is not simulated. The total number of transported aerosol species is 15.

Both representations of the aerosol account for all of the important processes that influence the aerosol: nucleation, coagulation, condensational growth, gas- and aqueous-phase chemistry, emission, dry deposition and gravitational settling, water uptake, in-cloud and below-cloud scavenging, and production from evaporated cloud and rain droplets.
**b. Clouds**

As in most global models, CAM5.1 distinguishes between stratiform and cumulus clouds. Stratiform cloud microphysics predicts number and mass mixing ratios of droplets and ice crystals and diagnoses number and mass mixing ratios of rain and snow as described by Liu et al. (2007), Morrison and Gettelman (2008), and Gettelman et al. (2008, 2010). Stratiform cloud macrophysics is described by Gettelman et al. (2010) and S. Park et al. (2012, unpublished manuscript).

The treatment of shallow cumulus clouds is described by Park and Bretherton (2009). The Zhang and McFarlane (1995) parameterization of deep convective clouds has been modified as described by Neale et al. (2008).

**c. Radiation**

Longwave and shortwave radiative transfer are parameterized with the Rapid Radiative Transfer Model for GCMs (RRTMG), a broadband $k$-distribution radiation model developed for application to GCMs (Mlawer et al. 1997; Iacono et al. 2003; Iacono et al. 2008).

Aerosol optical properties are parameterized according to Ghan and Zaveri (2007). Refractive indices for most aerosol components are taken from the Optical Properties of Aerosols and Clouds (OPAC) dataset (Hess et al. 1998), but for black carbon the value (1.95, 0.79i) from Bond and Bergstrom (2006) is used.

Liquid cloud optics are based on Mie computations (Wiscombe 1979) for pure water droplets assuming gamma size distributions that vary with stratiform cloud droplet number and mass mixing ratio (Morrison and Gettelman 2008). The size distributions of cloud droplets for shallow and deep cumulus clouds are assumed to be the same as those for stratiform clouds if they exist in the same grid cell; otherwise, they are set to constant values. Contributions to cloud droplet absorption by cloudborne particles are neglected because the burdens of cloudborne aerosol are much lower than the burdens of the rest of the aerosol.

Ice cloud optics are expressed in terms of the parameters of a gamma size distribution and the ice particle mass and projected area-dimension power laws using the modified anomalous diffraction approximation (Mitchell 2000, 2002; Mitchell et al. 2006).

**d. Turbulence**

The treatment of turbulence in CAM.1 is described by Bretherton and Park (2009), and its performance is evaluated by Park and Bretherton (2009).

**e. Resolved transport**

Transport of water and other trace species is calculated by the Lin–Rood finite-volume core (Lin and Rood 1996; Lin 2004). P. J. Rasch et al. (2012, unpublished manuscript) describe the performance of this dycore at $1^\circ \times 1.25^\circ$ resolution with CAM5 physics. Here we use $1.9^\circ$ latitude $\times 2.5^\circ$ longitude resolution with 30 vertical layers, five of which are in the bottom 10% of the atmosphere, and 17 of which are in the bottom 80% of the atmosphere (by mass).

**f. Simulation details**

Each simulation is performed with the same ocean surface conditions, ozone concentrations, and greenhouse gas concentrations, all for year 2000. Emissions are from the Intergovernmental Panel on Climate Change (IPCC) Assessment Report (AR5) estimates (Lamarque et al. 2010). Simulations with emissions of aerosols and precursor gases for years 2000 and 1850 are run for six years for the MAM3 and MAM7 representations of the aerosols, with only the final five years analyzed. The year 2000 simulations of the aerosol have been evaluated by Liu et al. (2011). The mean climate simulated by CAM5 is described by P. J. Rasch et al. (2011, unpublished manuscript).

3. Radiative forcing

To estimate the aerosol radiative forcing we express it as a radiative flux perturbation (Haywood et al. 2009; Lohmann et al. 2010) calculated from the difference between simulations with the same ocean surface conditions but emissions for years 2000 and 1850. Contrasting such simulations allows clouds to adjust quickly to the aerosols while preventing slow feedbacks involving changes in ocean surface temperature. This permits estimate of the adjusted radiative forcing due to both the essentially instantaneous scattering, absorption and emission of shortwave and longwave radiation by the aerosols and clouds (before liquid water content changes), and the radiative impact of aerosol-induced changes in cloud liquid and ice water content.

In this section we first present the radiative forcing decomposition for MAM3, then compare the decompositions of MAM3 and MAM7, and conclude with some sensitivity experiments designed that explore differences between MAM3 and MAM7.

**a. Decomposition for MAM3**

CAM5.1 contains the physics to represent aerosol direct effects, indirect effects, and semidirect effects. Distinguishing these three radiative forcing mechanisms from CAM5.1 simulations requires multiple diagnostic calls to the radiation routine in addition to the calculation that is used to drive the simulations. These diagnostic
calculations, which do not affect the simulations, include shortwave and longwave fluxes at the top of the atmosphere (TOA) for clear-sky conditions with cloud absorption and scattering excluded and with aerosol absorption and scattering included \((S_{\text{clean}}\) and \(L_{\text{clean}}\), also called the clear-sky fluxes) and for all-sky conditions with cloud absorption and scattering included and aerosol absorption and scattering excluded \((S_{\text{clean}}\) and \(L_{\text{clean}}\)). The anthropogenic aerosol forcing is diagnosed from the difference \(\Delta\) between the TOA fluxes from the year 1850 and 2000 simulations \(\Delta S = S_{2000} - S_{1850}\). In addition, to distinguish between indirect and semidirect effects (which both involve changes in clouds), additional 1850 and 2000 simulations (noanthrad) for MAM3 and MAM7 are performed in which all absorption and scattering by aerosol species with anthropogenic sources (sulfate, black carbon, organic carbon) are neglected by setting their refractive indices to zero, so that the only aerosol forcing mechanism is the indirect effect. To ensure that the effects of anthropogenic aerosol on water uptake and aerosol scattering, which still influence \(\Delta S\) in the noanthrad simulations, do not influence the indirect forcing estimate, we base the estimate on the clean sky fluxes \(S_{\text{clean}}\) and \(L_{\text{clean}}\).

From these diagnostics the anthropogenic radiative forcing can be decomposed as follows:

\[
\text{DIRECT} = \Delta(S - S_{\text{clean}}),
\]

\[
\text{SW INDIRECT} = \Delta S_{\text{noanthrad,clean}}^i,
\]

\[
\text{SW SEMIDIRECT} = \Delta S - \text{DIRECT} - \text{SW INDIRECT},
\]

\[
\text{LW INDIRECT} = \Delta(L_{\text{noanthrad}} - L_{\text{noanthrad,clean}}),
\]

and

\[
\text{LW SEMIDIRECT} = \Delta(L - L_{\text{clear}}) - \text{LW INDIRECT},
\]

where \(S\) and \(L\) are the net TOA shortwave and longwave radiative flux. Note that the longwave direct forcing is assumed to be negligible (which is justified by the small size of anthropogenic particles compared to the wavelength), and we diagnose the longwave forcing from the change in the longwave cloud forcing \(L - L_{\text{clear}}\) to avoid the clear-sky longwave signature of changes in land surface temperature induced by the aerosol. The direct forcing can be further decomposed into anthropogenic contributions from individual species (sulfate, black carbon, primary and secondary organic carbon) by additional radiation calculations with concentrations of individual species set to zero (the direct forcing by species \(i\) estimated from \(\Delta(S - S_{\text{noi}})\), where \(S_{\text{noi}}\) is the shortwave flux calculated without species \(i\)); for each of these calculations the mean particle size and aerosol water are recalculated without the species that have been zeroed to ensure that the direct forcing is zero in the case of all species being set to zero.

Table 1 summarizes the decomposition of the global annual mean anthropogenic aerosol radiative forcing for MAM3. (The MAM7 and MAM7-aging experiments results are discussed in sections 3b and 3c, respectively.) The largest forcing is due to shortwave indirect effects, which at 2.0 W m\(^{-2}\) is larger than estimates by most recent models that simulate aerosol effects on cloud liquid water content as well as droplet effective radius (Lohmann et al. 2010). Semidirect forcing is not significantly different (at \(p = 95\%\)) from zero for both longwave and shortwave radiation.

The aerosol direct forcing, \(-0.02\) W m\(^{-2}\), is much smaller in magnitude than the clear-sky estimate \(\Delta(S_{\text{clear}} - S_{\text{noanthrad,clean}})\), \(-0.35\) W m\(^{-2}\), because scattering aerosols increase the planetary albedo more for a clear atmosphere than a cloudy atmosphere. The global mean direct forcing is also smaller in magnitude than most other estimates (Forster et al. 2007). This result is due to enhanced warming by black carbon (compared with most other estimates) because CAM5.1 treats enhanced absorption when black carbon (BC) is internally mixed with soluble aerosol components and water, and the MAM3 treatment in CAM5.1 assumes BC is always internally mixed with other components in the accumulation mode. In addition, CAM tends to put a larger fraction of BC in the upper troposphere/lower stratosphere than other models do (Schwarz et al. 2010; Liu et al. 2011), which can increase the direct forcing by BC by 20\% (Zarzycki and Bond 2010).

To understand the direct forcing better, Table 2 lists the decomposition of the global annual mean direct forcing.
Table 2. Decomposition of global mean direct forcing (W m⁻²) for year 1850–2000.

<table>
<thead>
<tr>
<th></th>
<th>MAM3</th>
<th>MAM7</th>
<th>MAM7-aging</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ammonium sulfate</td>
<td>-0.176</td>
<td>-0.173</td>
<td></td>
</tr>
<tr>
<td>BC from fossil fuel and biofuel</td>
<td>0.196</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total black carbon</td>
<td>0.260</td>
<td>0.260</td>
<td>0.341</td>
</tr>
<tr>
<td>Secondary organic aerosol</td>
<td>-0.012</td>
<td>-0.009</td>
<td></td>
</tr>
<tr>
<td>Primary organic from biofuel</td>
<td>-0.021</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total organic matter</td>
<td>-0.030</td>
<td>-0.023</td>
<td>0.001</td>
</tr>
<tr>
<td>Biomass burning</td>
<td>0.035</td>
<td></td>
<td></td>
</tr>
<tr>
<td>All carbonaceous</td>
<td>0.218</td>
<td>0.228</td>
<td></td>
</tr>
<tr>
<td>Soil dust</td>
<td>0.014</td>
<td>0.034</td>
<td></td>
</tr>
<tr>
<td>Sea salt</td>
<td>0.012</td>
<td>0.010</td>
<td></td>
</tr>
<tr>
<td>Sum of contributions</td>
<td>0.068</td>
<td>0.099</td>
<td></td>
</tr>
<tr>
<td>Total direct</td>
<td>-0.017</td>
<td>-0.002</td>
<td>0.084</td>
</tr>
<tr>
<td>Difference</td>
<td>0.085</td>
<td>0.101</td>
<td></td>
</tr>
</tbody>
</table>

According to species. The decomposition is achieved with additional radiation calculations that neglect scattering and absorption by each selected species, so that the direct forcing by BC, for example, is determined from $\Delta(S - S_{\text{noBC}})$. The dominant contributions are cooling by sulfate and somewhat larger warming by BC (primarily from fossil and biofuel combustion). Although the sum of the global mean direct forcing by all components is positive (0.05 W m⁻²), the global mean total direct forcing is slightly negative; the difference is 0.085 W m⁻². This difference is much larger than the standard error of the direct forcing (0.01 W m⁻²) and is due to nonlinear interactions between the direct forcing by each component.

To understand this difference, Fig. 2 shows the spatial distribution of each component of the direct forcing for MAM3. Most of the spatial distribution of the total direct forcing is explained by the contributions from each component, with strong cooling over the Mediterranean Sea due to sulfate aerosol over the cloud-free water, and strong warming by fossil fuel black carbon over India and China and by biomass burning aerosol over clouds off the western coast of Africa (Chand et al. 2009). However, the sum of the contributions from all components is systematically greater than the total direct forcing nearly everywhere there is anthropogenic direct forcing. Potential explanations for the difference are as follows:

- reduction of planetary albedo by BC below sulfate increases radiative cooling by sulfate;
- increase in upward solar by sulfate below BC enhances radiative warming by BC;
- reduction in downward solar by BC above sulfate reduces radiative cooling by sulfate;
- reduction in downward solar by sulfate above BC reduces radiative warming by BC; and
- sulfate and water on BC particles enhances solar absorption by BC.

All of these nonlinear mechanisms are supported by the strong relationship between the direct forcing difference and the product of the burdens of BC and sulfate, shown in Fig. 3. Quantifying each of these mechanisms is difficult, but an upper bound on the last mechanism can be determined from the ratio of the BC absorption optical depth (calculated from the difference between estimates of total absorption optical depth with and without BC) at wavelength 0.55 μm to the BC absorption optical depth calculated without sulfate, organic carbon, sea salt, or the water associated with the hygroscopic aerosol, shown in Fig. 4. Absorption is strongly enhanced by the presence of the hygroscopic aerosol and water internally mixed with BC (Jacobson 2003). The enhancement factor is near one in regions dominated by dust but exceeds a factor of 2 in all regions influenced by sulfur and biomass burning emissions. The global mean BC absorption optical depth for year 2000 emissions is 0.0024 with the enhancement and 0.0014 without, for a global mean enhancement factor of 1.7. This suggests that the direct forcing imbalance is primarily due to absorption enhancement because of colocation of BC, sulfate, organic carbon, and water, rather than differences in the vertical distribution of absorbing and scattering aerosol.

Returning to the decomposition of the total forcing, Fig. 5 shows the spatial distributions of the year 1850–2000 annual mean total aerosol forcing from MAM3 decomposed into contributions from direct forcing and from shortwave and longwave indirect and semidirect forcing. With the exception of the direct forcing, all other forcing distributions vary widely between regions, from regional cooling exceeding 10 W m⁻² to warming exceeding 5 W m⁻². Semidirect effects are statistically insignificant almost everywhere, while there is a tendency for compensation between shortwave and longwave indirect forcing. Much of the spatial variability in shortwave indirect forcing is associated with spatial variability in changes in liquid water path (LWP) because of indirect effects, shown in Fig. 6. The strongest radiative cooling is associated with increased LWP in Southeast Asia because of indirect effects, as higher cloud condensation nuclei (CCN) (Fig. 7) and hence droplet number concentrations allow accumulation of more liquid water before clouds precipitate. Changes in LWP because of semidirect effects are insignificant everywhere.

Longwave indirect forcing is surprisingly large (+0.5 W m⁻² global mean). It is not associated with increases in liquid water path, as most liquid water clouds have sufficient LWP to emit as blackbodies at infrared wavelengths. Longwave indirect forcing is due to increases in upper-tropospheric cirrus through enhanced ice crystal nucleation. Figure 8 shows the vertical profile of the change in global annual mean cloud fraction and
cloud ice water content. Both increase in the upper troposphere, with the largest cloud ice increase at 200 hPa and the largest cloud fraction increase at 150 hPa. The changes in upper-tropospheric cloud ice are driven by increased homogenous nucleation and immersion freezing of droplets (also shown in Fig. 8), enhancing crystal nucleation that initiates ice formation and increases in cloud ice (Lohmann et al. 2004). Changes in all other terms in the cloud ice balance are much smaller. The increased homogeneous nucleation is due to higher concentrations of aerosol that form ice crystals at temperatures below $-40^\circ$C (Liu and Penner 2005). The increased droplet freezing is due to higher concentrations of upper-tropospheric CCN that increase the number concentration of droplets when cloud water is detrained from deep convective clouds. The nucleation peaks in the upper troposphere because of the strong temperature dependence of nucleation. The detrainment itself changes little between present 1850 and 2000 conditions. Surprisingly, homogeneous freezing of droplets plays little role, probably because it is calculated after immersion freezing, and most droplets freeze by that process. Further investigation of the sensitivity to the time integration sequence is needed.

The positive longwave indirect forcing differs from the large longwave cooling found by Penner et al. (2009), who attributed the cooling to heterogeneous nucleation on soot particles and found little evidence of sulfate effects on homogeneous nucleation. Carbonaceous aerosol particles in CAM5.1 are assumed to be poor heterogeneous ice nuclei, and the small changes in dust (an effective ice nuclei) are poorly correlated with the changes in crystal number concentration because the ice nucleation changes are dominated by changes in homogeneous nucleation and immersion freezing of droplets. This suggests that conflicting laboratory evidence of the ability of soot to nucleate ice crystals (Diehl and Mitra 1998; Gorbunov...
et al. 2001; Friedman et al. 2011), which leads to very different treatments of soot activation in climate models, needs to be reconciled.

b. MAM3 versus MAM7 forcing

Tables 1 and 2 summarize the decomposition of the global annual mean anthropogenic aerosol radiative forcing for MAM3 and MAM7. The estimates are remarkably similar, with all components agreeing to within 0.1 W m$^{-2}$ and the direct forcing decompositions for MAM3 and MAM7 agreeing to within 0.02 W m$^{-2}$ for each component. This suggests that MAM3 provides a useful approximation to the global mean forcing that MAM7 produces.

We now consider latitudinally resolved differences between the aerosol radiative forcing estimated with MAM3 and MAM7. Figure 9 shows many similarities, but also differences exceeding one standard error, between the zonal mean total, direct, shortwave indirect, shortwave semidirect, longwave indirect, and longwave semidirect forcing estimated by MAM3 and MAM7. MAM3 and MAM7 estimates agree quite well for direct,
shortwave indirect, and longwave indirect forcing and fairly well for longwave semidirect forcing. The most notable difference in the MAM3 and MAM7 total forcing is the stronger cooling by MAM3 near latitude 20\degree N. This difference is almost entirely due to the larger liquid water path response over South Asia (Fig. 10), as the droplet number changes (Fig. 10) are nearly identical in the MAM3 and MAM7 simulations. Consistent with this is the larger contribution of differences in semidirect forcing (through the liquid water path response) than indirect forcing to the difference between MAM3 and MAM7 forcing, for both longwave and shortwave.

The larger role of semidirect forcing in explaining MAM3 and MAM7 differences in radiative forcing suggests that differences in aerosol absorption optical depth might play a role. The 1850–2000 zonal mean change in aerosol absorption optical depth (Fig. 10) is not the same for MAM3 and MAM7 between latitudes 20\degree and 40\degree N. This disagreement is not due to the different treatments of black carbon in MAM3 and MAM7; Liu et al. (2011) find little difference between black carbon concentrations and burdens simulated by CAM5.1 with MAM3 and MAM7, and the MAM3–MAM7 difference in the change in BC burden (not shown) explains little of the difference in the change in aerosol absorption optical depth. Figure 11 shows that the disagreement is explained almost entirely by the different 1850–2000 change in column burden of mineral dust in the MAM3 and MAM7 simulations (the MAM3–MAM7 difference in the change in dust burden also explains most of the

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**Fig. 6.** Years 1850–2000 change in annual mean liquid water path due to indirect effects (above) and semidirect effects (below) from MAM3. Areas where change is insignificant (p = 95%) are not colored.

**Fig. 7.** Years 1850–2000 change in annual mean cloud condensation nuclei concentration at 0.1% supersaturation near pressure 900 hPa, from MAM3.

**Fig. 8.** Vertical profiles of years 1850–2000 change in global annual mean (right) cloud fraction, (middle right) cloud ice mixing ratio, and cloud ice production from (middle left) homogeneous nucleation and (left) immersion freezing from MAM3 (black curve) and MAM7 with POM hygroscopicity = 0 and slow aging (red curve).
difference in the change in aerosol extinction optical depth—not shown). This does not mean that dust is controlling the 1850–2000 changes in absorption optical depth, which as shown in Fig. 12 are dominated by the 1850–2000 BC changes, for both MAM3 and MAM7. Rather, the MAM3-MAM7 dust differences are controlling the MAM3-MAM7 differences in the 1850–2000 changes to absorption optical depth; However, most of the regional differences in the direct forcing estimates by MAM3 and MAM7 (not shown) are not statistically significant at $p = 0.05$ because the dust differences are small.

c. Sensitivity experiments

Although these comparisons suggest that the different treatments of aerosol mixing state and aging in MAM3 and MAM7 yield little difference in aerosol radiative forcing, the results depend on how aging is represented. If the hygroscopicity of POM is set to 0 (which is more appropriate for POM derived from fossil fuel combustion) rather than 0.1 (which is more representative of POM from biomass burning), and if the transfer of POM and BC from the primary mode in MAM7 to the accumulation mode is delayed by the requirement of eight rather than three monolayers of sulfate and equivalent secondary organic material, then larger differences between MAM3 and MAM7 emerge. Table 3 lists the global annual mean increase in black carbon column burden and absorption optical depth for MAM3, MAM7, and with these changes (denoted here by MAM7-aging). Compared with MAM7, the BC burden for MAM7-aging increases by 26%, but the BC absorption optical depth increases by only 20% because the BC mass absorption cross section is reduced from 14.2 to 13.6 m$^2$ g$^{-1}$ for MAM7-aging as less of the BC is internally mixed with sulfate, organic, and water (as discussed in the next paragraph, coatings on BC enhance absorption of sunlight). As listed in Tables 1 and 2, the increase in BC absorption optical depth increases the global mean direct forcing by BC from 0.26 to 0.34 W m$^{-2}$ and decreases the anthropogenic aerosol longwave warming from 0.46 to 0.27 W m$^{-2}$ as the weaker enhancement of upper tropospheric cloud ice by homogeneous nucleation (Fig. 8) is supplemented by upper-tropospheric warming because of enhanced BC concentrations and absorption. Most of the decrease in the longwave warming is due to indirect forcing (decreasing from 0.54 to 0.40 W m$^{-2}$) but a significant contribution is from the significantly stronger semidirect forcing (increasing in strength from $-0.08$ to $-0.14$ W m$^{-2}$) in MAM7-aging. The shortwave semi-indirect forcing is also stronger, increasing from $-0.01$ to 0.18 W m$^{-2}$, but this is compensated by a stronger shortwave indirect forcing. Thus, the total radiative forcing by MAM7 goes from $-1.54$ W m$^{-2}$ for POM hygroscopicity = 0.1 and fast aging to $-1.66$ W m$^{-2}$ in the slow aging experiment. Although the MAM7-aging experiment might be unrealistic, because eight monolayers are more than is necessary to transform the hydrophobic primary carbonaceous aerosol to hygroscopic aerosol, the simulated BC surface
concentrations are more realistic in most regions (Liu et al. 2011).

Although absorption of shortwave radiation by BC is known to be enhanced when BC is coated with water (Danielson et al. 1969; Chyžek et al. 1984, 1996; Jacobson 2003; Li et al. 2011), CAM5 neglects all absorption by cloudborne aerosol, on the grounds that the vast majority (97% for the whole atmosphere in CAM5 simulations) of BC aerosol is not cloudborne. To quantify the radiative impact of neglecting the absorption, we have performed another set of simulations with MAM7 in which shortwave absorption by cloudborne BC is treated, including enhancement by cloud water. Cloudborne aerosol is already simulated by CAM5 (Liu et al. 2011), so the only change needed is to treat the radiative effects of the cloudborne BC. The treatment follows that of Chuang et al. (2002, hereafter C2002), except that to maintain consistency with the treatment of droplet single-scatter albedo in CAM5 (Morrison and Gettelman 2008), the C2002 method is not used directly to determine the droplet single-scatter albedo. Instead, the C2002 expression for single-scatter albedo is applied to both the mixture of cloud water and cloudborne BC and to pure cloud water, and the difference in the single-scatter albedo between those two estimates is added to the Morrison and Gettelman (2008) estimate for pure water. As in C2002, the BC is assumed to be randomly distributed within the cloud droplets. The Li et al. (2011) analytic solution for the real and imaginary parts of the refractive index from...
the square of the complex refractive index eliminates the 
need for iterations. The direct forcing by the cloudborne BC 
is estimated by differencing two MAM7 estimates of 
aerosol direct forcing, one with and one without the treat-
ment of cloudborne aerosol absorption. We have found that 
the global annual mean direct forcing by cloudborne BC is 
negligible, that is, smaller than 0.01 W m$^{-2}$. However, 
semidirect forcing is larger because the absorption is col-
located with the cloud water. The shortwave semidirect 
forcing decreases by 0.02 ± 0.04 W m$^{-2}$ with cloudborne 
absorption, and longwave semidirect forcing decreases by 
another 0.05 ± 0.05 W m$^{-2}$, for a total forcing of −0.07 ± 
0.05 W m$^{-2}$. This forcing is perhaps not negligible but is 
much smaller than the uncertainty in other forcing mecha-
nisms. To the best of our knowledge, these are the first es-
imates of the radiative forcing due to absorption by the 
cloudborne BC.

4. Conclusions

We have shown that a three-mode representation of 
the aerosol composition and size distribution yields an 
estimate of anthropogenic aerosol radiative forcing that 
is globally to within 0.1 W m$^{-2}$ of that from a more com-
plex and computationally expensive seven-mode repre-
sentation and captures much of the spatial distribution of 
the total radiative forcing and its decomposition into 
direct, shortwave and longwave indirect, and shortwave 
and longwave semidirect forcing terms. The different 
assumed mixing state of freshly emitted black carbon in 
MAM7 (externally mixed) versus MAM3 (internally mixed 
with accumulation mode species) produces 10% differences 
in direct radiative forcing by black carbon near black carbon 
 sources and smaller differences elsewhere. About 90% 
(globally) of the MAM7 BC is aged and in the accumulation 
mode. However, this result is found to be dependent on the 
assumed hygroscopicity of the POM that is emitted with 
BC—a value of zero combined with a slower treatment 
of BC aging produces larger differences between MAM3 
and MAM7 estimates of BC direct and semidirect forcing. 
The largest source of differences between MAM3 and 
MAM7 with POM hygroscorticity = 0.1 is found to be due 
to differences in the dust simulation, which influences 
direct and semidirect forcing. However, the role of dust 
is probably overestimated because the refractive index 
chosen for dust is based on the OPAC model (Hess et al. 
1998), which overestimates absorption by a factor of 
about 3 for Saharan dust (Sinyuk et al. 2003). Since the 
three-mode model costs only 60% of the seven-model 
 model, the three-mode model is clearly recommended 
for simulations when computation time is limited.

Although many studies have quantified anthropogenic 
aerosol indirect forcing and others have quantified aerosol 
semidirect forcing, only recently have both indirect and 
semidirect forcing been quantified with the same model. 
Takeamura and Uchida (2011) performed two pairs of 
simulations to separate indirect and semidirect effects 
and estimated semidirect shortwave forcing to be +0.06 
W m$^{-2}$, Bauer and Menon (2012) also estimated the 
semidirect forcing as a residual, from simulations with 
the Goddard Institute for Space Studies (GISS)/ 
Multiconfiguration Aerosol Tracker of Mixing State 
(MATRIX) model and found the global mean to be 
−0.1 W m$^{-2}$, mostly owing to shortwave forcing. We have 
quantified indirect and semidirect forcing for both short-
wave and longwave radiation and find that both longwave 
and shortwave semidirect forcing are smaller in magnitude 
than 0.1 W m$^{-2}$, and that both globally and regionally 
semidirect forcing is statistically insignificant because of 
the natural variability in clouds. Although Lohmann and 
Feichter (2001) did not quantify semidirect forcing, they 
also found radiative warming effects on clouds to be 
smaller than effects through droplet nucleation. However, 
semidirect effects are significantly stronger in simulations 
that account for absorption by cloudborne aerosol and/or 
that increase BC concentrations (because of a lower POM 
hygroscopicity and a slower BC aging process).
We also have found the direct forcing by anthropogenic aerosol (sulfate, black carbon, organic carbon) to be smaller (−0.02 W m⁻² globally) than most previous estimates, probably because of enhancement of solar absorption by black carbon coated with scattering material, which nearly compensates for scattering by sulfate and organic carbon. Nonlinear interactions between the aerosol components also play a role.

In addition, both MAM3 and MAM7 produce a large positive longwave indirect forcing (0.5 W m⁻²), primarily as a result of homogeneous nucleation of ice crystals on anthropogenic sulfate and freezing of droplets detrained from deep convection. The response is likely to be sensitive to the representation of subgrid vertical velocity and to the representation of heterogeneous nucleation on black carbon, which is ignored in these simulations because recent laboratory studies (Friedman et al. 2011) suggest soot is ineffective as an ice nuclei.

The MAM7 has adopted some approximations that should be removed to provide more comprehensive benchmark estimates of radiative forcing. Nitrate should be included because it is important in some regions and its global importance is expected to increase in the future as sulfur and carbonaceous emissions are reduced. To accommodate the addition of nitrate and to treat deliquescence more realistically, water uptake should be determined with a more comprehensive thermodynamic model than the Kohler theory used in CAM5.1. The hygroscopicity of primary organic matter from fossil fuel should be reduced from 0.1 (the value used in these simulations) to a much lower value reflecting the hydrophobic nature of primary organic matter emitted from fossil fuel combustion, but it should not be reduced for POM from biomass burning. Absorption of sunlight by cloudborne aerosol material has been shown to produce a small influence on semidirect forcing (−0.07 W m⁻²) but negligible direct forcing. Comparison with MAM3 simulations could be used to determine the importance of these properties and processes for estimating aerosol radiative forcing for future as well as recent emissions scenarios.

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