Chapter 27

ARM-Led Improvements in Aerosols in Climate and Climate Models

STEVEN GHAN

Pacific Northwest National Laboratory, Richland, Washington

JOYCE E. PENNER

University of Michigan, Ann Arbor, Michigan

1. Introduction

The climate science community has been aware for several decades of the role of aerosols in the climate system and of the potential influence of anthropogenic aerosols in driving climate change (Penner et al. 1994). In the Fifth Assessment Report of climate change by the Intergovernmental Panel on Climate Change (IPCC), Boucher et al. (2013) introduced new terminology that more clearly distinguishes the key mechanisms by which anthropogenic aerosols alter the energy balance of the earth. The term radiative forcing (RF) refers to the impact of anthropogenic aerosols on the shortwave and longwave radiative fluxes without considering the adjustment of clouds to the aerosol. RFari is the component of RF due to aerosol–radiation interactions, specifically scattering and absorption of radiation, while RFaci is the component of RF due to aerosol–cloud interactions, specifically aerosol effects on droplet and ice crystal number but not liquid water or ice mass concentration. Radiative forcing is the sum of RFari and RFaci. ERFari refers to the effective radiative forcing due to aerosol–radiative interactions, including the adjustment of clouds to the aerosol scattering and absorption as well as RFari. ERFaci refers to the effective radiative forcing due to aerosol–climate interactions, including the adjustment of cloud microphysical processes and properties to the aerosol. The sum of ERFari and ERFaci is the effective radiative forcing (ERF). RFari was formerly called aerosol direct radiative forcing and RFaci was called the cloud albedo effect. ERFari includes both the direct effect and what was formerly called the semidirect effect, in which clouds adjusted to the scattering and absorption by the aerosol. ERFaci includes both the cloud albedo effect (RFaci) and what was formerly called the cloud lifetime effect. Ghan (2013) showed how the partitioning of ERF into (i) RFari and (ii) the sum of ERFaci and the semidirect effect (total effective forcing due to aerosol effects on clouds) can be diagnosed from the radiation balance calculated with and without scattering and absorption by aerosols in the model calculation.

The DOE ARM Program has played a foundational role in efforts to quantify aerosol effects on climate, beginning with the early back-of-the-envelope estimates of RFari by anthropogenic sulfate and biomass burning aerosol (Penner et al. 1994). This led to one of the first aerosol model intercomparison studies (Penner et al. 2001). In this chapter we review the role that ARM has played in subsequent detailed estimates of RFari, RFaci, and ERFaci based on physically based representations of aerosols in climate models. Only recently have other DOE programs applied the aerosol modeling capability to simulate the climate response to the radiative forcing.

2. Aerosol challenges for climate models

All estimates of aerosol radiative forcing begin with emissions of aerosol and aerosol precursor gases and with a representation of the aerosol life cycle. In the simplest back-of-the-envelope estimates the aerosol life cycle is expressed in terms of a prescribed value for the aerosol lifetime in the atmosphere, which converts global emissions into a global mean atmospheric burden.

Corresponding author address: Steven Ghan, Pacific Northwest National Laboratory, 902 Battelle Blvd., P.O. Box 999, MSIN K9-24, Richland, WA 99352.
E-mail: steve.ghan@pnnl.gov

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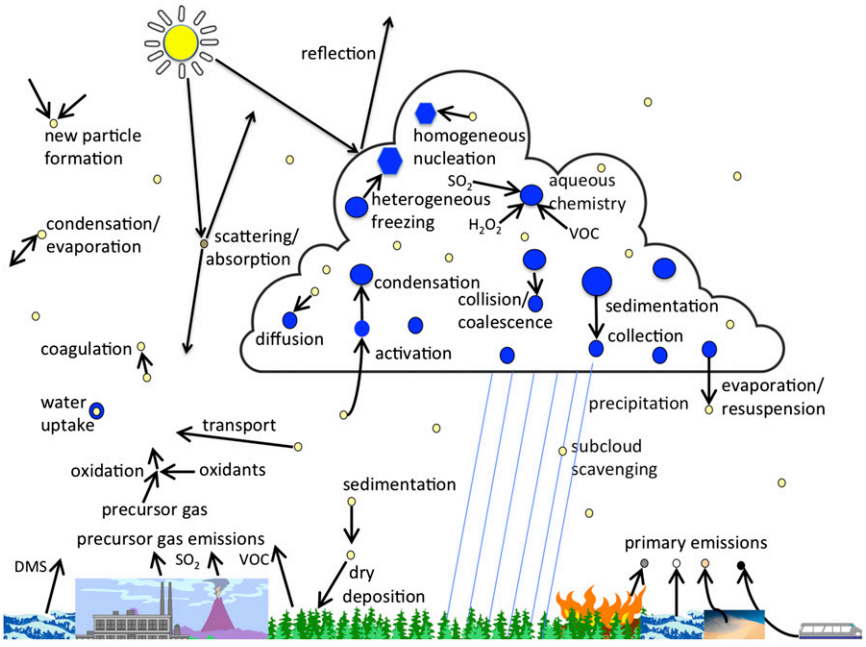

FIG. 27-1. Atmospheric processes important for producing aerosol effects on climate (after Ghan and Schwartz 2007).

In climate models, the aerosol life cycle is explicitly simulated, accounting for transport, transformation, sedimentation, and removal by wet and dry deposition to produce simulations of atmospheric concentrations of the aerosol of interest. Figure 27-1 illustrates the processes that must be represented in physically based models of aerosol effects on climate.

Since anthropogenic aerosols affect the earth's energy balance through scattering and absorption of solar radiation, and through effects on droplet number and cloud albedo, the ARM Program first focused on those aspects of aerosols that determine these effects. Most anthropogenic aerosols are composed of sulfate and carbonaceous material, so the focus has been on radiative forcing by those aerosol components.

Early in the program it was recognized that effects of one aerosol component on cloud droplet formation depends upon competition with all other components that contribute significantly to cloud condensation nuclei (CCN) concentration, natural as well as anthropogenic (Ghan et al. 1998), and that the influence depends on the size distribution of particles as well as their mass concentration (Ghan et al. 1993, 1995). This led to efforts to simulate the size distribution of all important components of CCN (Chuang et al. 2002; Easter et al. 2004; X. Liu et al. 2005). Since a significant fraction of the CCN is produced by gas-to-particle conversion, the global aerosol model must simulate emissions and oxidation of the secondary aerosol precursor gases such as dimethyl sulfide (DMS), SO_2, and volatile organic compounds (VOC). In the earliest simulations, the chemistry included in the model was simplified considerably. During the calculation of sulfur oxidation, the ozone concentration was prescribed because the aerosol does not affect it significantly, while hydrogen peroxide concentrations were allowed to respond to the significant loss during aqueous phase oxidation of SO_2 dissolved in cloud droplets. However, it is now recognized that the contribution of VOC oxidation to secondary organic aerosol formation requires that a full treatment of chemistry be included (Lin et al. 2012, 2014).

The importance of realistically representing clouds, particularly cloud microphysics, was also recognized early in the ARM Program. Droplet number concentration is determined by a balance of processes that include droplet nucleation, droplet collision/coalescence, collection by drizzle and rain droplets, and evaporation. To account for all of these processes Ghan et al. (1997a,b) introduced prognostic droplet number into a climate model, and later Morrison et al. (2005) generalized the approach to represent all processes that control number and mass concentrations of rain, snow, and ice as well as cloud droplets. The initial focus of aerosol–cloud interactions has been on the microphysics of stratiform clouds, but more recently attention has been directed to aerosol impacts on the microphysics of convective clouds (Song and Zhang 2011).

Just as estimates of cloud feedback by climate models depend on the cloud parameterizations in the models, so do estimates of ERFaci. The effort of the ARM Program to improve cloud parameterizations is therefore relevant for ERFaci as well as cloud feedbacks. The representation of droplet collision/coalescence, which is sensitive to droplet size and hence droplet number, is particularly critical, as it is the gateway to cloud microphysical adjustments to the aerosol, which then influence cloud fraction and cloud liquid and ice water contents. The treatment of turbulence and entrainment are also important, as those processes play important roles in both aerosol activation (Ghan et al. 1997b; Barahona and Nenes 2007) and in the response of cloud liquid water path to aerosol-induced changes in sedimentation (Ackerman et al. 2004).

Climate and aerosol models developed and applied by the ARM Program have been characterized by physically based rather than empirical representations of aerosol and cloud properties and processes. Rather than relate droplet number concentration to sulfate mass or aerosol number concentration as in some models, ARM researchers developed physically based representations of droplet formation (Ghan et al. 1993, 1995; Chuang et al. 1997; Abdul-Razzak et al. 1998; Abdul-Razzak and Ghan 2000), ice nucleation (Liu and Penner 2005), as well as the freezing of liquid to ice in mixed-phase clouds (DeMott et al. 2010; Yun and Penner 2012). Such physically based schemes offered the generality necessary for application to models with complete representations of the aerosol composition and size distribution and also offer insights into uncertainties und biases introduced by approximations.

For global climate simulations, it is not practical to represent the full complexity of aerosol and cloud properties and processes in climate models, so approximations were introduced to make the representation affordable. Ghan and Easter (1992), for example, introduced the diagnostic precipitation approximation, which made it possible to apply complex cloud microphysics schemes to climate models without excessive computational cost, although recent studies suggest the approximation adversely affects simulated aerosol effects on clouds (Lee and Penner 2010; Wang et al. 2012).

Similarly, the complexity of the distribution of size and composition of the aerosol population has been approximated by the use of a small set of aerosol modes, each mode composed of internal mixtures of aerosol components with the same proportionality for all particles within a mode (Easter et al. 2004). The distribution of number with size for each mode is usually represented by a lognormal distribution, with the width of the distribution prescribed and the mode radius either prescribed or diagnosed from the total volume concentration of the mode and a separate prediction of the total number concentration of the mode. Here again, though, simple internal mixture approximations may cause large differences in aerosol forcing compared to models that make fewer approximations (Zhou and Penner 2014).

The use of field and laboratory data to improve and test the representation of aerosol, cloud, and cloud–aerosol interaction processes is essential to increase confidence and reduce uncertainty in aerosol radiative forcing estimates. The strategy of founding and testing process models with field experiment data, applying the process models to regional and global models of the aerosol and cloud life cycles, using field experiment data to test the life cycle models, and using the models to estimate ERFari and ERFaci was laid out by Ghan and Schwartz (2007) and was followed during the development of all of the global models described in the next section. Retrievals of aerosol optical depth (AOD), spectral dependence of AOD, and single-scatter albedo from the surface at ARM and other surface sites and from satellite, as well as measurements of aerosol mass and CCN concentrations from aircraft have all been used to constrain and evaluate simulations of the present-day aerosol life cycle by global and regional aerosol models (Ghan et al. 2001a,b; Easter et al. 2004; X. Liu et al. 2005, 2007a; Wang et al. 2009). In parallel, simulated cloud–aerosol relationships have been evaluated using satellite retrievals of aerosol and clouds (Quaas et al. 2009; Wang et al. 2012).

Estimating anthropogenic aerosol impacts on climate also requires quantifying aerosol and aerosol precursor gas emissions and their life cycle and radiative and cloud impacts at preindustrial times. Evaluating aerosol simulations for preindustrial times is most directly done by comparing historical simulations with deposition rates determined from ice core measurements (which has not been addressed by the ARM Program), or by comparing simulations with measurements in remote regions (Penner et al. 2012), which the ARM Program can provide.

3. Process model development

To address the challenge of representing aerosol effects on climate in models adequately, it has been necessary to develop models of a variety processes involved in the link between emissions and climate impacts. As an atmospheric process science program, ARM has played a vital role developing models of several such processes.

It is straightforward to determine the optical properties from physical property measurements for externally

mixed aerosol composed of separate particle types. However, since most aerosol particles are known to be composed of internal mixtures, more difficulties arise. To determine the optical properties of internal mixtures of multiple hydrated aerosol components in aerosol modes with variable size distribution, Ghan et al. (2001a) and Ghan and Zaveri (2007) assumed particles are spheres and used the volume mean hygroscopicity and the kappa Kohler theory of Petters and Kreidenweis (2007) to diagnose the impact of water uptake on the wet mode radius of each mode. Parameterized Mie calculations then provide aerosol extinction, absorption, and the asymmetry parameter in terms of the wet mode radius and the volume mean wet refractive index of the aerosol components.

Optical properties and radiative impacts also are modified when mixing aerosol and cloud particles. To account for enhanced solar absorption by black carbon in cloud droplets, Chuang et al. (2002) used the effective medium approximation with the Maxwell Garnett mixing rule to diagnose the refractive index of black carbon-containing cloud droplets, and then used geometric optics to approximate the droplet single-scatter albedo in terms of droplet radius and the complex refractive index.

After the optical properties of the cloud and aerosol layers are determined, these have to be used in radiative transfer models. The radiative transfer in simulated atmospheres containing aerosol, clouds, and gases including interactions with the surface was made substantially more robust with the development of the Rapid Radiative Transfer Model for GCMs (RRTMG) (Mlawer et al. 1997; Iacono et al. 2003, 2008); the history of this model is described in Mlawer et al. (2016, chapter 15).

To treat aerosol indirect effects through the aerosol influence on droplet nucleation it was necessary to develop models of the aerosol activation process in terms of aerosol properties simulated by climate models. Twomey's (1959) expression for the number activated in terms of a power of supersaturation, while very popular for analysis of measurements, is not easily related to the aerosol quantities that can be predicted in climate models. Ghan et al. (1993, 1995) addressed this need by deriving an expression for the number activated in terms of the parameters of multiple lognormal aerosol size distributions. The parameters of the lognormal distribution can be related to aerosol mass and number concentrations for each mode, which can be simulated by climate models. Chuang et al. (1997, 2002) modified the Ghan et al. (1993) expression to account for different assumptions about the sulfate size distribution and aerosol mixing state over land and ocean. Abdul-Razzak et al. (1998) extended the generality of the Ghan et al. (1993) scheme by accounting for the influence of droplet growth after activation, and Abdul-Razzak and Ghan (2000) generalized the single-mode scheme to the multimode case. The latter scheme is used widely in climate models today. Abdul-Razzak and Ghan (2004) extended their scheme to treat the influence of surfactants on activation, and Abdul-Razzak and Ghan (2005) explored the role of slightly soluble components. Ghan et al. (2011) summarized the aerosol activation schemes developed since the Ghan et al. (1993) publication.

The droplet collision/coalescence process, also known as autoconversion in bulk cloud microphysics parameterizations, also has received considerable attention from the ARM Program because of its dependence on droplet size (and hence droplet number concentration) and because of its influence on cloud liquid water content (and hence cloud albedo). The ARM Program has developed two very different treatments of this process. Khairoutdinov and Kogan (2000) developed an empirical scheme using results from large-eddy simulations with size-resolved microphysics. The result, a simple expression proportional to droplet number concentration to the power −1.79, is used widely in cloud-resolving as well as general circulation models (GCMs).

A second treatment of droplet collision/coalescence was developed and described in a series of manuscripts by Y. Liu. Liu et al. (2004) began with the widely used Kessler (1969) autoconversion scheme, which is a discontinuous function of mean droplet radius, and used kinetic potential theory to derive an analytic expression for the critical droplet radius at the threshold between autoconversion and no autoconversion. Y. Liu et al. (2005, 2006a) related the scheme to earlier continuous ad hoc schemes, and Liu et al. (2006c) related the degree of continuity in the expression to the dispersion in the droplet size distribution.

In parallel, Y. Liu also developed representations of the droplet dispersion. Liu and Daum (2000) used in situ measurements to show that droplet dispersion is positively correlated with droplet number concentration, and Liu and Daum (2002) suggested that the correlation could diminish the aerosol cloud albedo effect; this was later quantified by Chen and Penner (2005). Liu et al. (2006b) provided theoretical support for the relationship in terms of activation of an aerosol size distribution, but employed Twomey's (1959) power law rather than lognormal size distributions.

As noted above, droplet nucleation and droplet collision/coalescence are two of numerous processes controlling cloud microphysics properties. The ARM Program has supported the development of packages of cloud microphysics that are well suited to quantifying aerosol indirect effects. Ghan et al. (1997a,b) introduced the droplet number balance to account for droplet collection, turbulent

transport, and evaporation as well as activation and collision/coalescence. This double-moment treatment of cloud droplets provided a physical basis for determining droplet number and the aerosol impact on the albedo of stratiform clouds. Morrison et al. (2005) applied the double-moment approach to cloud ice, rain, and snow as well as droplets, thus permitting treatment of aerosol effects on ice clouds and the potential for a more realistic treatment of aerosol resuspension as raindrops evaporate below clouds. Liu et al. (2007b) introduced parameterizations of ice nucleation mechanisms suitable for global models.

4. Development of a new generation of climate models

When the ARM Program started in 1990, the only climate models that simulated the aerosol life cycle were those designed first to study the climate impacts of aerosols produced by urban fires from a thermonuclear war. The GRANTOUR model (Walton et al. 1988) was a Lagrangian model consisting of 50 000 aerosol-containing air parcels coupled to meteorology simulated first by the Oregon State University two-level general circulation model (Ghan et al. 1988) and subsequently by the Community Climate Model (CCM1; Penner et al. 1991). Coupling was only through the radiative heating by the aerosol and through transport and removal of the aerosol by winds and precipitation, respectively. Chuang et al. (1997) first applied GRANTOUR to tropospheric sulfate aerosol for present-day and preindustrial emissions to estimate direct and indirect effects of anthropogenic sulfate. The preindustrial sulfate aerosol size distribution was prescribed, and two different treatments of anthropogenic sulfate were considered: (i) an internally mixed treatment, in which anthropogenic sulfate mass condenses on the preindustrial aerosol; and (ii) an externally mixed treatment, in which the anthropogenic sulfate aerosol has a different prescribed size distribution. The influence of water uptake on scattering by sulfate particles was introduced following the Köhler and Mie theory. Droplet number was diagnosed from an elaboration of the Ghan et al. (1993) scheme. Penner et al. (1998) then applied GRANTOUR to carbonaceous as well as sulfate aerosol, and Chuang et al. (2002) extended it to simulate all climatically important tropospheric aerosol (sulfate, black carbon, organic carbon, mineral dust, and sea salt), with a treatment of solar absorption by black carbon in cloud droplets. Chuang et al. (2002) used surface measurements to evaluate the aerosol concentrations simulated by GRANTOUR.

A second global aerosol model developed with ARM funding was the Model for Integrated Research on Atmospheric Global Exchanges (MIRAGE) developed at the Pacific Northwest National Laboratory (PNNL). MIRAGE (Easter et al. 2004) represented the aerosol-size distribution as the sum of four lognormal distributions, with the width of each distribution prescribed but the number and mass concentrations predicted from expressions for emissions, oxidation, new particle formation, condensation, coagulation, sedimentation, transport by resolved winds, turbulence and convection, aqueous-phase oxidation, and wet and dry removal. Predicting both number and mass for each mode is important for distinguishing between the effects of processes that primarily affect aerosol number (nucleation, coagulation) from those that primarily affect aerosol mass (condensation, aqueous production). All aerosol components (sulfate, organic carbon, black carbon, mineral dust, and sea salt) are assumed to be internally mixed within each mode, and concentrations of the cloud-borne as well as interstitial aerosol were predicted separately to ensure consistency of nucleation scavenging and cloud processing of the aerosol. The aerosol in MIRAGE interacted in each time step with the meteorology in CCM1 through absorption and scattering of solar radiation, droplet nucleation (which influenced cloud liquid water content as well as cloud albedo), aqueous chemistry, and removal by nucleation scavenging and impaction scavenging. Droplet nucleation was parameterized by the Abdul-Razzak and Ghan (2000) physically based scheme, and the optical properties of the internally mixed hydrated aerosol in each mode were parameterized in terms of the wet mode radius and volume mean wet refractive index (Ghan and Zaveri 2007). Droplet number was determined from the full droplet number balance (Ghan et al. 1997a,b). Easter et al. (2004) used surface and airborne measurements to evaluate the aerosol simulated by MIRAGE. Ghan et al. (2001a) used ARM and other surface-based and satellite retrievals to evaluate the simulated AOD and RFari, while Ghan et al. (2001b) evaluated the CCN concentration and cloud properties using aircraft measurements and satellite retrievals.

A third ARM-sponsored model is the Integrated Massively Parallel Atmospheric Chemical Transport (IMPACT) model (Liu and Penner 2002; X. Liu et al. 2005; Wang et al. 2009) developed jointly by Lawrence Livermore National Laboratory (LLNL) and the University of Michigan. IMPACT can be run as both an offline modal aerosol model driven by meteorology from the Goddard Earth Observing System (GEOS1.3; X. Liu et al. 2005) or inline together with the Community Atmosphere Model, versions 3 and 5 (CAM3 or

CAM5; Wang et al. 2009; Zhou and Penner 2014). Like MIRAGE, it predicts total number and mass concentrations of all important aerosol species in multiple aerosol modes (Herzog et al. 2004), but also includes a pure sulfate aerosol component, causing large differences in predicted sulfate number. It also does not explicitly predict the cloud-borne aerosol. The offline mode of calculation neglects the feedback on clouds and therefore does not include the cloud lifetime effect of the aerosol, but it simplifies estimates of RFaci because natural variability is not introduced (Wang and Penner 2009). Feng and Penner (2007) added nitrate and ammonium aerosols to the basic aerosol representation used by Chuang et al. (2002) in GRANTOUR. X. Liu et al. (2005) used surface measurements to evaluate the aerosol mass concentrations simulated by IMPACT, and Wang et al. (2009) used surface and aircraft observations to evaluate the simulated aerosol number concentration and size distribution in the coupled CAM3/IMPACT model.

Much of the aerosol and cloud physics developed for GRANTOUR, MIRAGE, and IMPACT was incorporated in the CAM5 (Liu et al. 2012). Two different modal configurations were introduced: a ''benchmark'' version with seven modes, and a simple version with only three modes. This model adopts an internally mixed representation of the aerosol. Total number concentration and mass concentrations of all important aerosol components are predicted for each mode, and interstitial as well as cloud-borne aerosol are predicted separately (though cloud-borne aerosol is not transported by the large-scale winds). Water uptake is based on Köhler theory using the volume-mean hygroscopicity, optical properties are expressed in terms of the wet mode radius and volume mean refractive index (Ghan and Zaveri 2007), and droplet nucleation is parameterized in terms of subgrid updraft velocity and the size distribution and volume-mean hygroscopicity of all aerosol modes (Abdul-Razzak and Ghan 2000). In addition, treatments of droplet collision/coalescence (Khairoutdinov and Kogan 2000) and homogeneous and heterogeneous ice nucleation (Liu and Penner 2005) were introduced as part of a double-moment cloud microphysics parameterization (Morrison and Gettelman 2008; Gettelman et al. 2008) based on the Morrison et al. (2005) double-moment cloud microphysics scheme developed with ARM funding. Liu et al. (2007b) introduced a treatment of ice supersaturation and vapor deposition on ice in CAM, and compared simulations with ARM retrievals of ice water content and in situ measurements of ice supersaturation. All of these capabilities are needed to quantify aerosol indirect effects through their role as droplet and crystal nuclei. Other ARM contributions to CAM5 include the RRTMG solar and infrared radiative transfer scheme (Iacono et al. 2003, 2008) and a shallow cumulus scheme (McCaa and Bretherton 2004; Bretherton et al. 2004; Park and Bretherton 2009; Bretherton and Park 2009). Liu et al. (2012) used ARM and other surface observations and satellite retrievals to evaluate the aerosol mass concentrations and AOD simulated by CAM5. Estimates of ERFari and ERFaci by CAM5 are described by Ghan et al. (2012). CAM5 was used as part of the Community Earth System Model (CESM) in simulations of anthropogenic climate change for the IPCC Fifth Assessment Report, and as a community model is widely used for studies of aerosol effects on climate.

5. Estimates of aerosol radiative forcing

Aerosol radiative forcing estimates have come a long way since the first back-of-the-envelope estimates by Penner et al. (1994), both in terms of aerosol speciation and processes represented. Estimates of the global mean ERF have converged slowly from a wide range between -0.4 and $-3\,\mathrm{W\,m^{-2}}$ in the early estimates to between -0.5 and $-2.4\,\mathrm{W\,m^{-2}}$ (Lohmann et al. 2010) as additional forcing mechanisms are introduced and estimates are constrained more effectively with data. In this section, we summarize what has been learned from aerosol radiative forcing studies funded by the ARM Program.

Penner et al. (1998) estimated RFari of carbonaceous and sulfate aerosol using GRANTOUR driven by meteorology from CCM1. Carbonaceous aerosol was assumed to be an internal mixture of black carbon and organic carbon, with refractive indices determined from volume mixing. Internal mixing of sulfate and carbonaceous aerosol was neglected. The radiative forcing by carbonaceous aerosol was found to be smaller than $0.1\,\mathrm{W\,m^{-2}}$, with compensation between $0.2\,\mathrm{W\,m^{-2}}$ radiative warming and $-0.2\,\mathrm{W\,m^{-2}}$ cooling by carbonaceous aerosol from fossil fuel and biomass burning aerosol, respectively. Consequently, total RFari was dominated by radiative cooling from anthropogenic sulfate aerosol, estimated to be -0.5 to $-0.8\,\mathrm{W\,m^{-2}}$.

Boucher et al. (1998) compared sulfate aerosol RFari estimates by different radiative transfer models and found that the estimates of forcing normalized by sulfate column burden agreed to within about 10% for most conditions. However, Boucher et al. did not consider the influence of clouds on the radiative forcing. More recent work (Stier et al. 2013) using prescribed aerosols with clouds simulated by climate models found much larger diversity for absorbing aerosol arising from differences in the simulated cloud distributions.

Chuang et al. (1997) produced the first estimates of RFaci of anthropogenic sulfate with a physically based aerosol activation scheme, using the GRANTOUR model. Estimates ranged from -0.4 to $-1.6\,W\,m^{-2}$, depending on assumptions about how sulfate is distributed on preindustrial aerosol. Chuang et al. (2002) extended the model to include indirect forcing by carbonaceous aerosol, and simulated the life cycles of natural aerosol (sulfate, organic matter, sea salt, mineral dust) as well as anthropogenic aerosol. The total RFaci was estimated to be $-1.85\,W\,m^{-2}$, with most of that from biomass burning. Significant dependence of anthropogenic forcing on emissions of natural aerosol was found because of the nonlinear dependence of droplet nucleation on aerosol concentration. Much smaller RFaci are calculated for nitrate and ammonium aerosols (i.e., $-0.09\,Wm^{-2}$) (Xu and Penner 2012).

The radiative influence of the correlation between droplet dispersion and mean droplet number was explored by Rotstayn and Liu (2003, 2009). When the Liu and Daum (2000) empirical relationship between dispersion and droplet number was applied to the Commonwealth Scientific and Industrial Research Organisation (CSIRO) GCM, the estimated RFaci was reduced by 12%–35%. When a more general empirical relationship between dispersion and the ratio of liquid water content/droplet number (Liu et al. 2008) was used, the cloud albedo effect was reduced by 42%.

Uncertainty in RFaci was systematically explored by Chen and Penner (2005) using prescribed aerosol, analyzed relative humidity, parameterizations of cloud fraction and cloud liquid water path, and an offline radiative transfer model.

From a reference estimate of $-1.3\,W\,m^{-2}$, they found the following:

- 20% uncertainty in RFaci due to anthropogenic emissions, preindustrial aerosol, and aerosol activation parameterization.
- 30%–60% uncertainty due to aerosol mass concentration.
- 50% uncertainty due to cloud fraction.
- 15%–40% uncertainty due to droplet dispersion.
- 5% uncertainty due to cloud liquid water path, updraft velocity, and aerosol mode radius and standard deviation.

The dependence of RFaci on the treatment of new particle formation was addressed with the IMPACT model by Wang and Penner (2009). Interestingly, when an empirical treatment of new particle formation replaces the binary homogeneous nucleation treatment in the boundary layer, RFaci changes little, from -1.55 to $-1.49\,W\,m^{-2}$, when primary emissions of sulfur are neglected, but changes much more, from -2.03 to $-1.65\,W\,m^{-2}$, when primary emissions of sulfate are included. As might be expected, both the total RFaci from sulfur and the enhancement when primary sulfur emissions are included, are weaker when the empirical nucleation scheme is added, because the empirical scheme produces more natural CCN in the boundary layer.

One of the first estimates of indirect forcing that included the cloud lifetime effect was by MIRAGE (Ghan et al. 2001c). The ERFaci by anthropogenic sulfate was estimated to be $-1.7\,W\,m^{-2}$, but a much larger estimate, $-3.2\,W\,m^{-2}$, was found when a treatment of autoconversion that is a discontinuous function of droplet number was used. ERFaci also was about 30% larger when the aerosol number was diagnosed from the simulated mass and a prescribed radius for each mode, rather than simulated independently. This sensitivity is to be expected as much of the sulfate condensation should go into making particles larger rather than making more particles. Such large negative estimates exceed the radiative warming by increasing greenhouse gases and hence are inconsistent with the observed warming of the earth over the last 150 years, but indicate the magnitude of uncertainty in bottom-up estimates of the radiative forcing.

The strong dependence on the parameterization of autoconversion was further explored in an ARM-funded AeroCom intercomparison of indirect effects estimates by different climate models (Penner et al. 2006). Even when the distribution of aerosol concentration was prescribed, the addition of a common autoconversion scheme that depends on droplet number yielded a wide range of responses of cloud lifetime effect by different climate models, with ERFaci increasing by as little as 22% to as much as a factor of 3 compared with estimates without the cloud lifetime effect. The particular choice of autoconversion scheme also yielded large differences, with the total indirect effect differing by just 10% when the same autoconversion scheme is used in each model but differing by a factor of 3 when different autoconversion schemes are used in each model. As might be expected, the one model that used an autoconversion scheme that is a continuous function of droplet number produced the smallest estimates of ERFaci.

Most global studies of aerosol effects on clouds have focused on stratiform clouds and have neglected aerosol effects on the microphysics of cumulus clouds. Menon and Rotstayn (2006) applied an empirical relationship between aerosol number and droplet number to cumulus clouds in two different global models, and added a

simple treatment of the dependence of precipitation formation on droplet number. They found very different responses of cloud liquid water path and radiative forcing to similar treatments of aerosol effects on cumulus clouds, with the CSIRO climate model producing a large liquid water path and radiative forcing response and the Goddard Institute for Space Studies (GISS) model producing a negligible response. This suggests considerable dependence of aerosol–cumulus interactions on details of the cumulus cloud parameterizations.

Semidirect aerosol effects, in which solar absorption by anthropogenic aerosol influences the energy balance through changes in clouds, were estimated by Chuang et al. (2002) and Penner et al. (2003) using GRANTOUR. Chuang et al. (2002) estimated the semidirect effect of cloud-borne black carbon on the planetary energy balance to be 0.07 W m^{-2}. Penner et al. (2003) found similar values for the solar semidirect effect, but found larger radiative cooling from the impact of the cloud changes on the longwave energy balance as cloud dissipation enhanced longwave emission to space.

In the last decade the ARM Program has supported efforts to quantify anthropogenic effects on ice clouds. Liu et al. (2009) coupled the IMPACT aerosol model to CAM3 using the Liu and Penner (2005) ice nucleation scheme and quantified radiative impacts of anthropogenic aerosol via homogeneous and heterogeneous ice nucleation. Anthropogenic sulfate was estimated to produce 0.5 W m^{-2} of radiative warming through homogeneous nucleation, with positive contributions from both shortwave and longwave radiation. Anthropogenic black carbon, which Liu and Penner assumed to be capable of serving as ice nuclei through the immersion nucleation mechanism, increased crystal number and cloud ice, making shortwave cloud forcing 1.5 W m^{-2} more negative and longwave cloud forcing 1.5 W m^{-2} more positive, for a small net forcing. The anthropogenic aerosol effect on longwave cloud forcing was estimated to be 0.5 W m^{-2} in CAM5 (Ghan et al. 2012) with ~0.3 W m^{-2} resulting from homogenous ice nucleation of anthropogenic sulfate and the remainder from immersion freezing of droplets detrained from deep convection. This estimate agrees well with a more recent estimate of 0.27 W m^{-2} for ERFaci through ice clouds (Gettelman et al. 2012). Yun and Penner (2012) and Yun et al. (2013) examined forcing in mixed-phase clouds, finding values between 0.16 and 0.93 W m^{-2}. Total forcing using the IMPACT aerosol model was −2.36 W m^{-2}.

Climate models are designed for future projections of climate change, and those projections depend on future emission and radiative forcing. Menon et al. (2008) explored changes in aerosol radiative forcing between years 1980, 1995, and 2030 using the GISS ModelE and projected emissions for the IPCC A1B scenario. The aerosol radiative cooling was projected to be greatest for the year 2030 with the projected larger sulfur emissions from China.

6. Cooperating with other programs

While the ARM Program played an instrumental role in collecting the data, developing and evaluating the process models and cloud and aerosol life cycle models, and using the models to estimate aerosol radiative forcing, other federal programs also played essential roles.

The DOE Atmospheric Science Program supported the development of empirical parameterizations of aerosol nucleation (Kuang et al. 2008), of the Y. Liu autoconversion parameterization (Liu and Daum 2004; Liu et al. 2006a), and of the MOSAIC aerosol thermodynamics model (Zaveri et al. 2005a,b; 2008) that have been or are being applied to global aerosol models. The DOE Science Discovery through Advanced Chemistry (SciDAC) program funded the development of CAM5, which incorporated many of the advances described in this chapter.

The National Aeronautics and Space Administration (NASA) Interdisciplinary Science Program supported the development of MIRAGE. The NASA Global Aerosol Climatology Project and Radiation Science Program partly funded development and application of GRANTOUR. The NASA Atmospheric Chemistry Modeling and Analysis Program (ACMAP) partly funded the AeroCom intercomparison and the development and application of the IMPACT model.

The National Science Foundation provided partial funding to couple the IMPACT model and the CAM model as well as to incorporate new treatments of ice and mixed-phase aerosol/cloud microphysics.

7. Summary and future progress

The ARM Program has played a vital role in the emergence of global modeling of the aerosol life cycle and aerosol effects on the earth's energy balance. Key insights into aerosol radiative forcing mechanisms and ways to represent them efficiently in climate models have been supported by ARM. The importance of representing all aerosol components, natural as well as anthropogenic, was established by ARM-funded scientists. This understanding led to the development of the treatments of the aerosol life cycle and cloud–aerosol interactions in the CESM1 used for simulations that

contributed to the IPCC Fifth Assessment Report (Boucher et al. 2013).

However, the uncertainty in estimates of aerosol radiative forcing still drives an almost threefold uncertainty in total radiative forcing of climate change. Although the relative uncertainty in future forcing will decline as carbon dioxide accumulates in the atmosphere and anthropogenic aerosol emissions are reduced, a threefold uncertainty in radiative forcing between present-day and preindustrial conditions renders the observed warming of little use in constraining estimates of climate sensitivity.

How then, to reduce that uncertainty? Ghan and Schwartz (2007) outline a strategy that involves greater complexity in process models constrained with more complete, accurate, and sensitive measurements of the aerosol. Missing aerosol components, such as nitrate, need to be added, and crudely represented components, such as secondary organic aerosol, need to be improved. A more general representation of thermodynamic equilibrium that is suitable for all important aerosol components is needed. The influence of aging by condensation on the aerosol mixing state needs to be represented more realistically, as well as emissions of primary black carbon and organic carbon and secondary organic carbon. Better understanding of new particle formation is needed as well. The treatment of wet removal of aerosol needs to be constrained better with measurements. The microphysical effects on cumulus clouds need to be represented, and better understanding and representation of heterogeneous ice nucleation on dust and black carbon are also needed. ARM can play a central role in each of these process studies.

Finally, while most of the emphasis of aerosol research during the last 20 years has focused on the impact of anthropogenic aerosol on the planetary energy balance, studies of the influence on precipitation and snow albedo are just beginning.

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