

REPRESENTING AEROSOLS IN GLOBAL MODELS: FROM MICROMETERS TO MEGAMETERS

Stephen E. Schwartz

Environmental Chemistry Division, Brookhaven National Laboratory, Upton NY 11973 USA

Anthropogenically induced climate change is of great current interest because of increases in atmospheric loading of infrared active (greenhouse) gases over the past 150 years and the inferred resultant increase in the flux of infrared radiation in the troposphere. In parallel with the increase in greenhouse gas loadings there has been an increase in loadings of tropospheric aerosols derived from industrial activities. These aerosols scatter shortwave (solar) radiation leading to a decrease in the shortwave radiation absorbed by the earth-atmosphere system, thereby exerting a cooling influence on climate, the direct aerosol effect. These aerosols are thought also to modify the microphysical and radiative properties of clouds, enhancing their albedo and perhaps suppressing precipitation, thereby enhancing cloud lifetimes, both of which effects would also exert a cooling influence, the indirect aerosol effect.

Present estimates suggest that the magnitude of the global average climate forcing by these aerosol effects is comparable to the longwave (thermal infrared) forcing by greenhouse gases, that is one to a few watts per square meter. However the estimates, which are based entirely on model calculations of aerosol loading, are considered quite uncertain. If the magnitude of aerosol forcing is at the low end of the currently estimated uncertainty range, aerosols negate only a small fraction of the greenhouse forcing, but at the high end of the uncertainty range, aerosols could be negating virtually all of the present greenhouse forcing. There is thus great uncertainty in net climate forcing over the industrial period, mainly because of uncertainty in aerosol forcing. This situation urgently requires resolution in view of the implications of this overall uncertainty in climate sensitivity to greenhouse gas forcing and the necessity for decision making on possible strategies to limit greenhouse gas forcing.

Light scattering by aerosols is most efficient (on a mass basis) for particles of diameter comparable to the wavelength of visible light, *ca.* 0.5 μm . Particles in this size range result mainly from gas-to-particle conversion of low-volatility gaseous species, mainly sulfuric acid and nitric acid and their ammonia neutralization products and partially oxidized organics, formed in the atmosphere by atmospheric oxidation processes. The forcing depends *inter alia* on mass loading and mass-scattering efficiency and single scattering albedo, all of which depend on the size distribution and composition of the aerosol. The indirect forcing depends on the change in number concentration of cloud drops, which also depends on aerosol size distribution and composition.

Improving estimates of aerosol forcing requires global atmospheric transport models to estimate the loading and pertinent microphysical properties as a function of location and time. Representing aerosol forcing in global-scale models is necessary first to take into account covariances among the several controlling variables. This approach is necessary also to permit evaluation of model performance by comparison of modeled and observed loadings and properties. Models are essential to permit confident evaluation of aerosol forcing, as opposed to evaluating forcing empirically, for example from measured optical depths. Even if the instantaneous aerosol influence could be determined empirically, models would be required to evaluate the secular evolution of this forcing and to determine future forcing in response to prospective changes in emissions. Ultimately it will be necessary to represent aerosol processes and forcing "on-line" in climate models in order to capture the feedbacks of aerosols on the climate system. These requirements place strong demands on the aerosol research community, first to develop process-level understanding necessary for accurate representation of aerosol evolution and second to develop means of representing these processes with sufficient economy to permit their inclusion in climate models.

Aerosol models must accurately represent the key components of the aerosol life cycle: emissions of aerosols and precursors, chemical transformations responsible for gas-to-particle conversion, new

particle formation, aerosol microphysical evolution, three-dimensional transport, and wet and dry removal processes of aerosols and precursors. The performance of these models must be evaluated, by comparison with measurements, so the models certainly do not supplant measurements. However once the models have been evaluated over a wide range of conditions, they can be used with known confidence to evaluate aerosol loading, and in turn forcing, at present, and for past times and for future emissions scenarios.

In addition to the requirements for climate models there are a variety of other requirements for models describing evolution of atmospheric aerosols. Key among these is the need for models to devise strategies to achieve particle loadings that are protective of human health. The U.S. has adopted a standards for maximum loadings of suspended particulate matter of aerodynamic diameter less than 2.5 μm ($\text{PM}_{2.5}$) on daily and annual time scales. Demonstrating the understanding necessary to develop strategies to achieve such standards requires models describing aerosol evolution certainly on a distance scale of tens to hundreds of kilometers.

To date models of aerosol transport and transformation, especially global models, have for the most part represented aerosol mass loading and not the size distribution or other representation of aerosol microphysical properties. Some of these models have been rather successful in comparison against measurements. Although most modeling studies to date have used meteorological variables from general circulation models to drive the chemical transport and removal processes, there is no inherent requirement for this. Driving models by observationally derived meteorological data in principle allows for detailed comparison that takes account of the large dynamic range in aerosol loadings that must otherwise be smoothed by averaging.

The key reason for the difficulty in representing aerosol processes in chemical transport and transformation models is that unlike gases, aerosols possess many "intensive" properties that must be accurately represented, in addition to the "extensive" property of mass loading. Key among these properties are size distribution and size-dependent composition. These properties govern aerosol light scattering and activation to cloud droplets. Both size and composition are dynamic, evolving as gaseous substances (including, very importantly, water vapor) condense onto and evaporate from the aerosol particles, as particles coagulate, and as particles are selectively removed from the atmosphere by dry deposition or by becoming activated into cloud drops and removed in precipitation. All of these processes are themselves dependent on the properties of the aerosol and therefore in turn on the sources of the aerosol and prior atmospheric processing. Accurate description of aerosol dynamics requires understanding of the controlling physics and chemistry and knowledge of pertinent input variables, together with much more demanding computational requirements than for representation of chemical processes. All of these are lacking to some extent, but are being actively pursued. For the indefinite future we must be content with various approximate treatments of aerosol evolution, and a number of candidate approaches are being examined in a very active field of research.

Intuitively it is attractive to describe the atmospheric aerosol and its evolution by the so-called "bin" approach. The aerosol radius (or mass) distribution is approximated by a number of bins which may be spaced arithmetically, or more commonly, to cover a greater dynamic range, logarithmically. In principle a sufficiently narrowly spaced set of bins should be able to represent the aerosol. In practice a compromise is made that treats, typically, some eight or so bins, each of whose composition is evolved. Despite its attractiveness the approach can suffer, among other problems, from numerical diffusion of particle size. The potential for error here can be quite large, as a factor of 2 in radius, which is the size resolution characteristic of such models, represents a factor of 8 in mass. Of course the error can be reduced by decreasing the bin size. The method has been used recently with some success in describing transport and evolution of aerosols on urban scales leading to rather accurate representation of size and size-dependent composition.

A second approach is the "modal" approach, which considers the aerosol to consist of several modes of distinct size, and perhaps composition. Each mode can evolve chemically and physically by gas-particle exchange. Aerosol growth due to gas-to-particle conversion is represented by

increasing the mode radius of each of the several modes; the width parameter of each mode can also be allowed to evolve. This approach is attractive because fewer variables need to be tracked during a calculation than with the bin method, but there is the disadvantage that the modes are required to evolve according to a pre-specified and fixed functional form (e.g. lognormal, modified gamma, etc.) whereas the true evolution of the size distribution can proceed along a quite different path.

Another promising approach to representation of aerosol microphysical properties in transport-transformation codes is by evolving the moments of the size distribution, the "method of moments." The k^{th} moment of an aerosol radial size distribution is

$$\mu_k = \int_0^\infty r^k f(r) dr$$

where $f(r)$ is the radial size distribution. An advantage of moment methods is freedom from numerical diffusion of particle size. Another advantage is that the moments evolve as chemical species. Note that the area density of an aerosol is $[A] = \int_0^\infty r^2 f(r) dr$ so that condensation of a monomer, which can be viewed as a "reaction" between monomer and surface area, has a rate that is given to good approximation as

$$\text{Rate} = k[A][m] = k \mu_2 [m]$$

where $[m]$ is the monomer concentration. Moments are conserved on transport and mixing, just like chemical substances. Thus the evolution of the zeroth moment is

$$\frac{d\mu_0}{dt} = -D \mu_0 - (\mu_0 \mathbf{v}) + \int_0^\infty J(r) dr$$

where $J(r)$ is the rate of nucleation of new particles having radius r . The moment equations are not closed, so in principle an infinite set of moment equations is required. However approximations have been developed that allow the moments to be evolved as a closed set of only the low-order moments. What comes out of the treatment is not the size distribution of the aerosol, as one has hitherto been accustomed to, but the moments of the evolved distribution. While this may initially seem dissatisfying, it turns out that most aerosol properties of interest, if not all, can be evaluated in terms of the low-order moments. For example, the effective radius, which leads to successful parametrizations of aerosol optical properties, is equal to the quotient of the third and second moments: $r_e = \mu_3 / \mu_2$. Calculations of aerosol direct forcing using only the lowest six moments agree with the forcing computed exactly for known (measured or modeled) test aerosol size distributions within 1%. Similar levels of accuracy are obtained for the extinction coefficient and several other important aerosol optical properties. We have begun to incorporate a moment representation of aerosol size distribution into a chemical transport and transformation model, and initial results will be presented.

It is essential to carry out a major effort in modeling aerosol loading and properties and in evaluating model performance by comparison with observation. This places high demands on the aerosol research community. However the stakes are high and we must rise to the challenge.