Atmospheric aerosols, suspensions of solid or liquid particles, are an important multi-phase system. Aerosols scatter and absorb shortwave (solar) radiation, affecting climate (Charlson et al., 1992; Schwartz, 1996) and visibility; nucleate cloud droplet formation, modifying the reflectivity of clouds (Twomey et al., 1984; Schwartz and Slingo, 1996) as well as contributing to composition of cloudwater and to wet deposition (Seinfeld and Pandis, 1998); and affect human health through inhalation (NRC, 1998). Existing and prospective air quality regulations impose standards on concentrations of atmospheric aerosols to protect human health and welfare (EPA, 1998). Chemical transport and transformation models representing the loading and geographical distribution of aerosols and precursor gases are needed to permit development of effective and efficient strategies for meeting air quality standards, and for examining aerosol effects on climate retrospectively and prospectively for different emissions scenarios.

Important aerosol properties and processes depend on their size distribution: light scattering, cloud nucleating properties, dry deposition, and penetration into airways of lungs. The evolution of the mass loading itself depends on particle size because of the size dependence of growth and removal processes. For these reasons it is increasingly recognized that chemical transport and transformation models must represent not just the mass loading of atmospheric particulate matter but also the aerosol microphysical
properties and the evolution of these properties if aerosols are to be accurately represented in these models. If the size distribution of the aerosol is known, a given property can be evaluated as the integral of the appropriate kernel function over the size distribution. This has motivated the approach of determining aerosol size distribution, and of explicitly representing this distribution and its evolution in chemical transport models.

Atmospheric chemical transport models to date mainly represent only mass of aerosol constituents. Part of the reason for this is inadequate understanding of the key processes that govern the aerosol size distribution:

- Gas-to-particle conversion--conversion of volatile gaseous species by atmospheric reaction to nonvolatile material that adds to existing particles and can result in new particle formation.
- Nucleation (new particle formation).
- Activation of aerosol particles to form cloud droplets.
- Reactions in cloud droplets giving rise to additional aerosol material upon cloud droplet evaporation.
- Removal of aerosol material from the atmosphere through clouds and precipitation.
- Dry deposition of aerosol particles.

Much research is required to gain understanding of these processes sufficient to confidently represent them in chemical transport and transformation models.

Additionally a major hurdle facing the aerosol modeling community is representation of the aerosol size distribution and its evolution. Representation of aerosol dynamics is difficult in box models, and virtually prohibitive in atmospheric transport models (which may be thought of as a set of box models in parallel with coupling between adjacent boxes due to transport), because of the large numbers of variables (several hundred or more), namely the number of particles within a given size range for a set of size ranges, which must be tracked in each grid cell of the model in order to accurately represent the size distribution and to avoid errors associated with numerical diffusion within the distribution.

An alternative and potentially attractive approach is to represent the aerosol dynamics by the low-order moments of the size distribution, where the \( k \)th moment of an aerosol size distribution is defined as

\[
\mu_k = \int r^k n(r) dr
\]

where \( n(r) \) is the radial size distribution (number of particles per unit volume and unit radius interval); \( r \) is the radius variable. The method is attractive in that it offers the
hope that key aerosol properties can be represented by a few variables; in our work we use six moments, \( k = 0 \cdots 5 \).

It is a premise of the moment approach that the key information about the aerosol is contained in the low-order moments and that quantities of interest can be reconstructed from them. For example the number concentration is equal to the zeroth moment. The mean radius is equal to the first moment divided by the zeroth moment. The surface-area concentration is equal to \( 4\pi \) times the second moment. The volume concentration is equal to \( 4\pi/3 \) times the third moment. The effective radius, which serves as the basis of highly accurate parametrizations of aerosol optical properties and atmospheric radiative effects (Hansen and Travis 1974) is the ratio of the third to second moments, \( r_e = \mu_3/\mu_2 \). More generally a given aerosol property \( \langle \sigma \rangle \), which would conventionally be evaluated as the integral of the appropriate kernel function over the size distribution, can be evaluated with fair accuracy by quadrature techniques (McGraw, 1997),

\[
\langle \sigma \rangle \equiv \int_0^{\infty} \sigma(r) n(r) dr \equiv \sum_{i=1}^{N} \sigma(r_i) w_i
\]

where the quadrature abscissas \( r_i \) and weights \( w_i \) are obtained from the low order moments by inversion of:

\[
\mu_k = \sum_{i=1}^{N} r_i^k w_i
\]

For \( N \) quadrature points \( 2N \) moments are required. New approximate techniques developed by our group have led to even greater accuracy, typically a few percent for a six-moment representation.

A further requirement of the moment approach is that the aerosol evolution be expressible as a closed set of differential equations in the moments themselves. Consider aerosol growth resulting from accretion of monomer. The rate of increase of aerosol volume \( (V, \text{ third moment}) \) is proportional to the surface area \( (A, \text{ second moment}) \) and the monomer concentration \([m]\) and thus monomer accretion may be thought of as a "reaction" between second moment and monomer. The rate of increase of volume of a particle of radius \( r \) is

\[
\frac{dV}{dt} = \beta [m] A
\]

for free molecular growth, for which the rate coefficient \( \beta \) is independent of \( r \), there is a closed set of rate equations in the moments:
\[
\frac{d\mu_0}{dt} = 0; \quad \frac{d\mu_k}{dt} = \beta[m]k\mu_{k-1}, \quad k > 0.
\]

More general rate expressions can be evaluated by quadrature techniques, as above. An example is shown in Figure 1; on the scale shown the moments evaluated by quadrature are indistinguishable from those obtained by the exact calculation.

The moment equations are readily incorporated in chemical reaction codes that may be applied to box models or to multi-box chemical transport and reaction models. In the latter case transport of moments is described according to the continuity equation under gradient transport assumptions (Bott, 1989; Easter, 1993). Figure 2 shows an example of the results obtained with this approach for the six lowest-order radial moments for each of several externally-mixed aerosol populations in a subhemispheric model driven by observation-derived meteorological data (Wright et al., 2000). These populations include sulfate, sea salt, and continental background aerosol (2 modes). Collectively they are represented by 24 moments, plus sulfate mass per particle for each of the three non-sulfate populations, for a total of 27 variables. The module produces new particles by binary sulfuric acid-water nucleation; no 'tuning' factor was used, as none was needed to obtain copious new particle formation. Other aerosol evolution processes represented were condensation, coagulation, dry deposition, wet removal, and size-resolved cloud activation. The Global Chemistry Model driven by Observation-derived meteorological data (GChM-O; Benkovitz et al., 1994; Benkovitz and Schwartz, 1997) served as the platform into which we implemented the new aerosol module. The horizontal resolution is 1.125° and there are 15 vertical levels extending to about 100 hPa. For aerosol transport and mixing, the model employs a fourth-order, positive definite advection scheme driven by archived meteorological data from the European Centre for Medium-Range Weather Forecasts (ECMWF). The results are from a simulation period that began October 1, 1986, with no aerosol initially present in the model domain. No aerosol boundary fluxes were included, so the aerosol present was emitted or generated entirely within the model domain. This study demonstrates the capabilities of these moment-based techniques to simultaneously represent aerosol evolution and transport in a large scale model and to calculate size-dependent optical properties and radiative influence of the modeled aerosol.

Additional results and comparisons with conventional approaches to evaluation of aerosol evolution and properties will be presented. Results so far indicate that the moment based approach will be a powerful and useful means of treating atmospheric aerosols and, more broadly, description of aerosol evolution in other environments such as flames.

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REFERENCES


**Figure 1.** a) Initial size distribution (normalized Khrgian-Mazin distribution with mean particle radius of 5 µm) and exact solution for final size distribution under conditions of diffusion controlled growth by accretion of water vapor for 20 s at T = 278 K and fixed saturation of 101% that served as basis for comparison of exact and quadrature methods. b) Time dependence of low order moments (µkm cm⁻³) evaluated from exact solution (solid lines) and by quadrature method of moments (dashed lines). From McGraw (1997).
Figure 2. Modeled sulfate aerosol on October 22, 1986, 0600 UT. (a) log_{10} number concentration (µ_0) at z = 32 meters, (b) number mean radius (r = µ_1/µ_0) at z = 32 meters, (c) effective radius (r_{eff} = µ_3/µ_2) at z = 32 meters, (d) column integral of condensed aerosol volume for the dry aerosol, (e) column integral of condensed aerosol volume for the ambient aerosol, and (f) magnitude of the clear-sky forcing at a wavelength of 550 nm. White regions indicate values below the plotting scale. From Wright et al. (2000).