Aerosol Science and Technology 34: 353–362 (2001)
© 2001 American Association for Aerosol Research
Published by Taylor and Francis
0278-6826/01/$12.00 + .00

Method of Moments Solutions for a Laminar Flow Aerosol Reactor Model

D. A. Terry,[1] R. McGraw,[2] and R. H. Rangel[1]
[1]*Department of Mechanical and Aerospace Engineering, University of California, Irvine, California*
[2]*Department of Applied Science, Brookhaven National Laboratory, Upton, New York*

This study compares the capabilities of the conventional and quadrature methods of moments to describe condensational growth in a well-known laminar flow aerosol reactor model. Governing equations for energy, lower order radial moments of the particle size distribution, and vapor transport are written for a two-dimensional model using both approaches.

Conventional and quadrature techniques to obtain closure of the moment equations are applied and compared. The conventional method requires certain approximations to the growth law. These include assumptions that the Kelvin effect is negligible and that the particle growth rate function is linear in particle size. In contrast, restrictive constraints of the conventional method of moments to obtain closure are avoided by applying the quadrature method. Numerical results for the laminar flow aerosol reactor model are obtained for a six moment formulation of the seed particle distribution to illustrate application of the quadrature method to continuous polydisperse distributions. We present the first calculations with the new quadrature method of moments to a two-dimensional aerosol transport and growth model.

INTRODUCTION

In this work we apply the method of moments (Hulburt and Katz 1964; Randolph and Larson 1988; Friedlander 1983; McGraw and Saunders 1984) to condensational growth in a laminar flow aerosol reactor model using conventional and quadrature techniques. We begin with a brief description of the method as used for an aerosol dynamic equation to model growth and particle transport (Hulburt and Katz 1964; Randolph and Larson 1988). Conventional (Hulburt and Katz 1964) and quadrature (McGraw 1997) methods to obtain closure for a set of moment equations are described. Finally, we present numerical solutions for continuous, or polydisperse, seed particle distributions using the quadrature method of moments without restrictions on particle growth. This is the first application of the quadrature method to a two-dimensional laminar flow reactor model with condensation.

The reactor modeled in this study, the modified Sinclair-LaMer aerosol generator (Sinclair and LaMer 1949; Nicolaon et al. 1970; Nicolaon et al. 1971), consists of evaporation and condensation sections and is similar to reactors that have been used to investigate competition among kinetic processes during aerosol formation (Pesthy et al. 1983; Pratsinis 1988). Early models of laminar flow reactor models have been solved analytically for delta function particle size distributions using a Green's function approach (Liao 1974; Davis and Liao 1975) and numerically with orthogonal collocation (Barrett and Fissan 1989). Pesthy et al. (1983) extended the model to include homogeneous nucleation and vapor wall losses. Pratsinis (1988) and Phanse and Pratsinis (1989) provided a more complete formulation coupling the aerosol dynamic equation to the heat and mass transport equations. Phanse and Pratsinis (1989) discuss the advantages of describing the aerosol dynamics using the moments of the size distribution, but also point to the difficulty of obtaining closure for the resulting moment equations. All studies, except recent papers on multicomponent modal models (Stratmann and Whitby 1989; Wilck and Stratmann 1997), assume axially constant laminar flow velocity profiles and neglect thermophoresis and axially diffusion. Therefore the latter two papers are more general and more accurate with respect to representation of the flow field and transport equations.

In this paper we use a fully two-dimensional model with an aerosol dynamic equation including a Brownian diffusion term. Since the focus of this paper is application of the quadrature method of moments, the hydrodynamic model is not as complete as those found in Phanse and Pratsinis (1989), Stratmann and Whitby (1989), or Wilck and Stratmann (1997).

The emphasis of this and previous modeling studies cited above is the condensation section. For this study the flow is modeled as laminar with sufficiently low particle concentrations to avoid coagulation, and supersaturations are kept sufficiently low to prevent homogeneous nucleation. The modified Sinclair-LaMer aerosol generator, widely used in the laboratory to produce liquid droplets of narrow size distribution (Davis and

Received 20 November 1997; accepted 20 March 2000.
Address correspondence to Roger H. Rangel, Department of Mechanical and Aerospace Engineering, University of California at Irvine, Irvine, CA 92697-3975. E-mail: rhrangel@uci.edu

Liao 1975), is of interest because by carefully controlling conditions in the evaporation section, the size distribution of seed particles present in the flow at the entrance to the condensation section can be controlled. In addition, the conditions of the modified Sinclair-LaMer aerosol generator are very similar to those found in various condensation nuclei counters. The modified Sinclair-LaMer aerosol generator is thus a simple but practical means for comparing different modeling approaches.

METHOD OF MOMENTS

The methodology for using lower order moments of the size distribution to describe droplet or particulate growth was first developed to address the difficulties of modeling growth in nonhomogeneous flow environments (Hulburt and Katz 1964; Randolph and Larson 1988). Despite its unique advantages for complex flow, the method of moments has not received widespread application due to restrictions on the kinds of particle growth laws that could be handled previously by the method (McGraw 1997). Early applications (Friedlander 1983; McGraw and Saunders 1984) involved growth processes that were easily modeled within the restrictions of the conventional method of moments (Hulburt and Katz 1964). Later applications (Pratsinis 1983; Phanse and Pratsinis 1989; Stratmann and Whitby 1989; Wilck and Stratmann 1997) typically obtained closure of the moment equations by specifying the shape of the aerosol size distribution as a log normal function requiring a priori assumptions regarding the shape of the distribution. By differing approaches, Frenklach and Harris (1987) and McGraw (1997) attempt to overcome restrictive a priori assumptions on either the growth processes or the aerosol size distributions. With either of these methods, a priori assumptions concerning the functional form of the size distribution are unnecessary. Here we adopt the quadrature approach.

Description of Moment Equations

Similar to Hulburt and Katz (1964) and Phanse and Pratsinis (1989), we construct a set of moment equations that describes the behavior of the number density distribution function $f(\vec{x}, a, t)$, where $\vec{x}$ represents the position vector, a particle radius, and t time. The set of equations is written as follows:

$$\frac{\partial \mu_n}{\partial t} + \nabla \cdot (\mu_n \vec{v}) = D_{diff} \nabla^2 \mu_n + n \int_0^\infty a^{n-1} G(T, C, a) f(\vec{x}, a, t)\, da, \quad [1]$$

where we have included terms on the right-hand side for Brownian diffusion, and heterogeneous (G) nucleation, but have not included homogeneous nucleation, coagulation, and wall losses. In this set of equations, $\vec{v}$ is the velocity vector of the flow, with T the absolute temperature, C the dispersed vapor phase mass concentration, and D_{diff} the Brownian particle diffusivity.

The radial moments are defined by

$$\mu_n(\vec{x}, t) = \int_0^\infty a^n f(\vec{x}, t, a)\, da \quad [2]$$

for $n = 0, 1, 2, 3, \ldots$. Generally a small number of lower order moments are sufficient for estimating the important physical properties of an aerosol (Hulburt and Katz 1964; Friedlander 1977; McGraw et al. 1995). For particles in a carrier gas, the physical significance of the lower order moments (Sherwin et al. 1967) is as follows. The zero moment μ_0 is the total number of particles per unit volume or number density. The first moment μ_1 is the total particle radii per unit volume. Similarly, $3K\mu_2$, proportional to the second moment, is the total particle surface area per unit volume, and $K\mu_3$, proportional to the third moment, is the total volume fraction. The parameter K is the geometric shape factor, which for spherical particles is $K = 4\pi/3$.

Note that the fourth term of Equation (1) depends on the unknown number density $f(\vec{x}, a, t)$ and seemingly cannot be evaluated without full knowledge of the distribution. However, the main focus below is a discussion of techniques for evaluating this integral in terms of the lower order moments (Equation (2)).

Closure of Moment Equations

Two techniques to implement the method of moments in complex flow environments are outlined below. To obtain closure of Equation (1) with the conventional method of moments, it is necessary that the growth law be both separable and linear in particle radius (Hulburt and Katz 1964; Randolph and Larson 1988). Assuming separability of the growth function into exterior flow coordinates and internal particle coordinates, we write

$$G(C, T, a) = g(C, T)\phi(a). \quad [3]$$

To obtain closure $\phi(a)$ must take the restrictive linear form

$$\phi(a) = \beta_0 + \beta_1 a. \quad [4]$$

For this approach, the set of moment equations above, coupled with the transport equations, can be closed using a small number (either three or four) of moment equations. If a growth law is not of the quadrature form, it must be approximated as such or assumed distribution functions such as log normal are required to obtain closure (Hulburt and Katz 1964).

Recently, McGraw (1997) introduced a new approach, the quadrature method of moments, through which exact moment equations are replaced by a quadrature based set that satisfies closure under a broad range of conditions. Thus the quadrature method of moments and the related quadrature inversion virtually eliminate these restrictions on particle growth. Furthermore, the quadrature method is a generalization of the conventional method to which it reduces for the special form of Equation (4). The method can be summarized as follows. Consider an integral aerosol property I as given by Equation (5) below, where $f(a)$ is the unknown aerosol size distribution and $\sigma(a)$ is a known kernel function of particle size. Generally for physical properties and dynamics we require evaluation of integrals of unknown size distributions; for example, the growth integral in Equation (1) is in just this form. In the quadrature method we write the integral

property I using the quadrature sum

$$I = \int \sigma(a) f(a)\, da \cong \sum_{i=1}^{N} \sigma(a_i)\, w_i, \qquad [5]$$

where N is the number of quadrature points. Similarly, if we rewrite the kernel as $\sigma(a) = a^k P(a)$, we can write

$$I = \int \sigma(a) f(a)\, da = \int a^k P(a) f(a)\, da \cong \sum_{i=1}^{N} a_i^k P(a_i)\, w_i. \qquad [6]$$

Both σ and P may be functions of parameters in addition to the radius. For each quadrature abscissa a_i there is a corresponding weight w_i. Notice that the moments themselves can be approximated by this quadrature form for $P(a) = 1$:

$$\mu_k = \int_0^\infty a^k f(a)\, da = \sum_{i=1}^{N} a_i^k w_i. \qquad [7]$$

Equation (7), which is exact for $k = 0$ through $k = 2N - 1$, shows that the first $2N$ moments uniquely determine the N abscissas and N weights. However, the direct solution of Equation (7) for these quantities would require a nonlinear search and is not recommended. A superior quadrature inversion algorithm for computing the abscissas and weights directly from the moments is described in McGraw (1997). Briefly summarized, computation of quadrature abscissas and weights from the moments may be accomplished in two steps, which include (1) construction of the *Jacobi matrix* (Press et al. (1992), whose elements are given in terms of the moments), and (2) solution of the eigenvalue problem associated with this matrix. The net result is that we have evaluated the integral I in terms of the moments of $f(a)$ even though this function itself is unknown.

MODIFIED SINCLAIR-LAMER MODEL

Efforts to quantitatively model laminar flow reactors began with the modified Sinclair-LaMer aerosol generator model (Davis and Liao 1975). The model includes steady state laminar flow of an aerosol-vapor-carrier gas mixture in a cooled pipe with condensation described using a Green's function approach. Pesthy et al. (1983) extended the model to include homogeneous nucleation. The addition of the aerosol dynamic equation to track evolution of particle growth was first seen in Hulburt and Katz (1964) and more recently in Pratsinis (1988). Pratsinis (1988) and Phanse and Pratsinis (1989) included homogeneous nucleation and coagulation and obtained closure of the aerosol dynamic equation using a method of moments with an assumed log normal particle size distribution. Recently, studies on single and multicomponent modal models (Stratmann and Whitby 1989; Wilck and Stratmann 1997) also obtain closure using an a priori assumption that the aerosol size distribution consists of log normal modes. In this section, we outline a two-dimensional laminar flow reactor model consisting of coupled partial differential equations for energy, vapor, and particle transport without a priori assumptions of the aerosol size distribution.

Heat Transport, Mass Transport, and Aerosol Dynamic Equations

The modified Sinclair-LaMer generator model, written in nondimensional form and updated with an aerosol dynamic equation, consists of the energy and vapor transport equations (Davis and Liao 1975)

$$(1 - r^{*2})\frac{\partial \theta}{\partial z^*} = Le\left(\frac{\partial^2 \theta}{\partial r^{*2}} + \frac{1}{r^*}\frac{\partial \theta}{\partial r^*}\right), \qquad [8]$$

$$(1 - r^{*2})\frac{\partial X}{\partial z^*} = \frac{\partial^2 X}{\partial r^{*2}} + \frac{1}{r^*}\frac{\partial X}{\partial r^*} - \phi_s \qquad [9]$$

for laminar, parabolic pipe flow, and the aerosol dynamic equation (Hulburt and Katz 1964; Pratsinis 1988)

$$(1 - r^{*2})\frac{\partial \mu_n}{\partial z^*} = \frac{D_{diff}}{D_m}\left(\frac{\partial^2 \mu_n}{\partial r^{*2}} + \frac{1}{r^*}\frac{\partial \mu_n}{\partial r^*}\right) + [\psi_s]_n \qquad [10]$$

for transport of the condensed phase (see Figure 1 for a schematic of the system coordinates).

In Equations (8), (9), and (10), we use a common nondimensional axial coordinate $z^* = z/r_0 Pe_2$ and a nondimensional radial coordinate $r^* = r/r_0$. The Lewis number Le is the ratio of the Peclet numbers for vapor and thermal diffusion. The reduced temperature and the normalized vapor concentration are defined as

$$\theta = \frac{T - T_w}{T_0 - T_w}, \quad X = \frac{C}{C_0}. \qquad [11]$$

We define the Brownian particle diffusivity $D_{diff} = \kappa T/(6\pi a \mu)$ using the Stokes-Einstein equation (Bird et al. 1960),

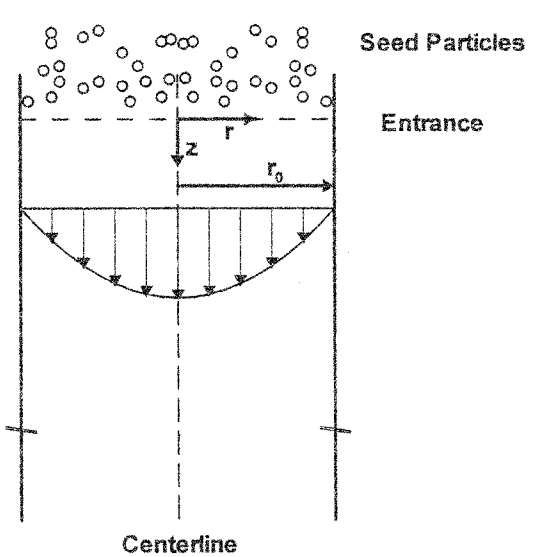

Figure 1. Schematic of the aerosol generator model showing the system coordinates.

Table 1
Fundamental physical parameters (reference case)

Symbol	Parameter	Value	Units
v_0	Centerline velocity	21.0	cm
r_0	Pipe radius	1.0	cm
T_0	Entrance temperature	110	Deg C
T_w	Wall temperature	60	Deg C
C_0	Entrance vapor concentration	1.12×10^{-6}	gm/cm^3
D_m	Vapor mass diffusivity	0.16	cm^2/sec
M	Molecular weight	278.35	gm/mole
R_G	Universal gas constant	8.3143×10^7	ergs/mole-K
α_T	Thermal diffusivity	1.80	cm^2/sec
ρ_L	Liquid DBP material density[a]	1.047	gm/cm^3
a_0	Seed particle radius	1.0×10^{-6}	cm
μ	Absolute viscosity for Helium	2.281×10^{-4}	gm/cm-sec
ρ_G	Helium gas density	0.1875×10^{-3}	gm/cm^3
σ	Surface tension[b]	34.0	dynes/cm

[a]Kemppinen and Gokcen (1956).
[b]Dean (1985).

where κ is Boltzmann's constant. For additional symbols in these equations see Tables 1 and 2.

The sink/source terms in Equations (9) and (10) are defined as

$$\phi_s(a) = \frac{r_0}{C_0 D_m} \int_0^\infty 4\pi a^2 f(a) \rho_L G(\theta, X, a)\, da, \quad [12]$$

$$[\psi_s(a)]_n = \frac{r_0^2}{D_m} n \int_0^\infty a^{n-1} G(\theta, X, a) f(a)\, da. \quad [13]$$

Following the approach in Davis and Liao (1975), we have assumed the axial heat conduction is negligible. Also, the heat release from condensation is considered negligible. We have assumed homogeneous nucleation is not active. Additional details on the development of the model can be found in Davis and Liao (1975) and Phanse and Pratsinis (1989).

In addition, we neglect thermophoresis and axial diffusion in formulating the model equations. Wilck and Stratmann (1997), in using a more complete hydrodynamic model, have shown these processes to be negligible.

Growth Law

The growth law for dibutylphthalate (DBP) in a helium carrier gas (Davis and Liao 1975) is written

$$G(\theta, X, a) = C_0 \frac{D_m}{\rho_L} \left[X - X_e^f \exp(2\sigma M / \rho_L R_G T a) \right] \frac{F}{a}. \quad [14]$$

In this equation, the exponential coefficient $\exp(2\sigma M / \rho_L R_G T a)$ accounts for surface curvature when the particles are very small (i.e., Kelvin effect). In addition, $X_e^f = C_e^f / C_0$ is the nondimensional equilibrium vapor concentration at the local temperature over a flat surface. At the local reduced temperature θ the equilibrium vapor concentration over a flat surface and the equilibrium vapor pressure are

$$C_e^f = \frac{(p_2)_e}{R_G[(T_0 - T_w)\theta + T_w]} M,$$
$$(p_2)_e = 10^{-A/[(T_0 - T_w)\theta + T_w]} 10^B, \quad [15]$$

where the parameters A and B (see Table 2) are experimentally determined vapor pressure constants (Ray et al. 1979).

The factor F, included in Equation (14), is the noncontinuum correction for particle dimensions comparable to the mean free

Table 2
Derivative physical parameters (reference case)

Symbol	Parameter	Value	Units
Pe_1	Thermal Peclet number	11.6	—
Pe_2	Mass diffusion Peclet number	131.25	—
Le	Lewis number	11.25	—
β_0	First growth rate coefficient	2.5×10^4	—
β_1	Second growth rate coefficient	-2.6×10^8	—
C_∞	Vapor concentration at infinity	2.2399×10^{-8}	gm/cm^3
μ_A	Seed particle number density	1.2×10^7	Particles/cm^3
$[\tilde{\mu}_1]_*$	Nominal first reduced moment	27.53518	1/cm
A	First vapor pressure constant[a]	4993.0	Deg K
B	Second vapor pressure constant[a]	12.217	—
v	Kinematic viscosity for Helium	1.27781	cm^2/sec
Re	Reynolds number	16.43358	—
l	Mean free path of vapor molecule	3.015×10^{-5}	cm

[a]Ray et al. (1979).

path of the gas (Fuchs and Sutugin 1970). This is given by

$$F = \{1 + [Kn(1.333Kn + 0.71)/(Kn + 1)]\}^{-1}, \qquad [16]$$

where the Knudsen number Kn is the ratio of the mean free path to particle radius (i.e., $Kn = l/a$).

Entrance and Boundary Conditions

The entrance conditions for the energy and vapor transport equations, respectively, are $\theta_0 = 1$ and $X_0 = 1$. Initial conditions for the moments of the seed particle distribution are described in Numerical Results. Boundary conditions on the energy equation include $\theta_w = 0$ at the outer pipe boundary and $\partial\theta/\partial r^* = 0$ at the centerline. For the vapor transport equation we have $\partial X/\partial r^* = 0$ at the outer pipe boundary and at the centerline. Similarly, for the particle transport moment equations at the pipe boundary and the centerline we have $\partial\mu_n/\partial r^* = 0$.

As derived above, Equations (9) and (10) for the vapor and particle transport contain singularities at the outer pipe boundary due to the velocity vanishing at the wall for parabolic flows. Severe restrictions on particle growth were imposed by Davis and Liao (1975) on the auxiliary equations used to model the particle transport. Specifically, instantaneous condensation at the pipe boundary was assumed, yet no further growth of boundary particles by diffusive transport was allowed. Recent papers (Pratsinis 1988; Phanse and Pratsinis 1989; Wilck and Stratmann 1997) utilize more realistic boundary conditions, accounting for the high likelihood of at least some condensation on the pipe boundary. However, we have chosen to stay with the simpler approach of no flux through the boundary. While this approach is likely unattainable in practice, with this approach it is easier to interpret the exchange of mass between vapor and condensed phases. Therefore, at the outer boundary where the singularity exists, we do not impose deposition boundary conditions and do not solve for vapor concentration or particle size at the boundary. But there are no restrictions with the quadrature method of moments to prevent the use of a deposition boundary condition.

DEVELOPMENT OF THE MOMENT EQUATIONS

Comparison of the conventional and quadrature methods of moments amounts to comparison of strategies to obtain closure for the set of coupled model equations (Equations (8), (9), and (10)). In this section, we outline the approximations needed to implement the conventional method in contrast to the much less restrictive approach used to obtain closure for the quadrature method of moments.

For numerical convenience, we define a set of reduced moments in the form

$$\tilde{\mu}_n = \int_0^\infty \frac{(a/a_0)^n f(a)}{N_T}\, da, \qquad [17]$$

thus normalizing the distribution function by the total number density N_T and the particle radius by a characteristic length scale a_0. In terms of the original moments, we have

$$\tilde{\mu}_n = \frac{1}{a_0^n N_T}\mu_n. \qquad [18]$$

Implementation of the Conventional Method of Moments

Following the conventional approach described in Hulburt and Katz (1964), the integral in Equation (10) is simplified by assuming the growth law is separable in the form $G(\theta, X, a) = g(\theta, X)\phi(a)$. To achieve this for the growth law of Equation (14) it is necessary to assume the Kelvin effect is negligible with $\exp[2\sigma M/\rho_L R_G T a] \approx 1$. The consequences of ignoring the Kelvin effect are small except at the smallest particle sizes. Also, it is necessary to linearize the particle growth rate function $\phi(a)$. With these approximations, the growth law becomes

$$g(\theta, X) = C_0\frac{D_m}{\rho_L}\left[X - X_e^f\right], \quad \phi(a) = \frac{F}{a} \cong \beta_0 + \beta_1 a. \qquad [19]$$

In Figure 2, nonlinear and approximate forms for the particle growth rate function $\phi(a)$ for DBP are compared. For the linearized growth rate, the coefficients β_0 and β_1, given in Table 2, were obtained by least squares fit over the range 10^{-2} to 5×10^0 and should be considered valid only over this range.

To couple the moment equations to the vapor transport equation we substitute the separable growth law and apply the definition of reduced moments to the sink/source terms of Equations (12) and (13). The resulting expressions are

$$\phi_S = \frac{4\pi r_0^2}{C_0}\frac{\rho_L}{D_m} g(\theta, X)\, a_0^2 N_T[\beta_0\tilde{\mu}_2 + a_0\beta_1\tilde{\mu}_3], \qquad [20]$$

$$[\psi_S]_n = \frac{r_0^2}{D_m} n g(\theta, X)\frac{1}{a_0}(\beta_0\tilde{\mu}_{n-1} + a_0\beta_1\tilde{\mu}_n). \qquad [21]$$

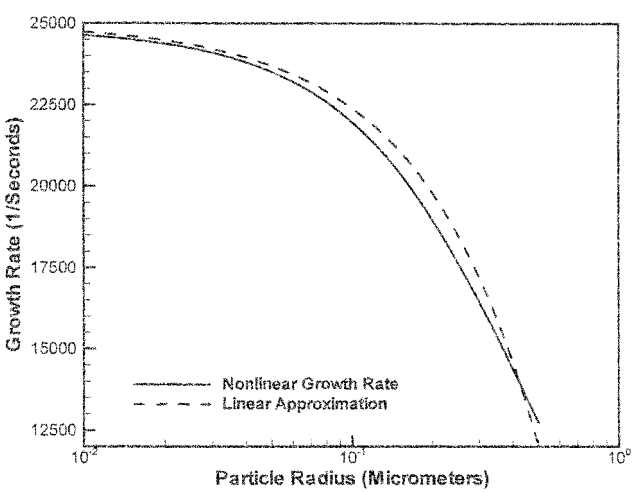

Figure 2. Comparison of the nonlinear growth rate function $\phi(a) = F/a$ and the linear approximations $\phi(a) = \beta_0 + \beta_1 a$ for $\beta_0 = 2.5 \times 10^4$ and $\beta_1 = -2.6 \times 10^8$ (see discussion above Equation (19)).

With these substitutions it is evident a closed set of equations results for the vapor and moments $n = 0$ through $n = 3$.

Implementation of the Quadrature Method of Moments

In applying the conventional method of moments to the modified Sinclair-LaMer aerosol generator model, approximations to the growth law were made to obtain a closed set of equations. McGraw (1997) proposed the quadrature method of moments as a means to obtain an approximate set of equations to obtain closure, but with the important advantage that the growth law is not restricted in its form. In this subsection, we outline application of the quadrature method to obtain the equations needed for numerical solution.

Without restrictions on the form of the growth law of Equation (14), we use quadrature (McGraw 1997) to rewrite the sink term of Equation (12) as

$$\phi_S = \frac{r_0^2}{C_0}\frac{\rho_L}{D_m}4\pi \sum_{i=1}^{N} a_i^2 G(\theta, X, a_i)\, w_i. \qquad [22]$$

To rewrite this equation in a form comparable to the reduced moments used in the conventional method, we define normalized abscissas and weights using

$$b_i = \frac{a_i}{a_0}, \quad \gamma_{1i}^2 = \frac{w_i}{\mu_0}, \qquad [23]$$

where a_i and w_i are the quadrature points as in Equation (6). After substitution and minor manipulation, the sink term has the final form

$$\phi_S = \frac{r_0^2}{C_0}\frac{\rho_L}{D_m}a_0^2 N_T 4\pi \sum_{i=1}^{N} b_i^2 G(\theta, X, a_0 b_i)\gamma_{1i}^2, \qquad [24]$$

where we have also used $\mu_0 = N_T$ from Equation (18).

Similarly, the source terms of the moment equations take the form

$$[\psi_S]_n = \frac{r_0^2}{D_m} n \frac{1}{a_0} \sum_{i=1}^{N} b_i^{n-1} G(\theta, X, a_0 b_i)\gamma_{1i}^2, \qquad [25]$$

where Equations (10), (18), and (23) have been used. The growth law appearing in Equation (25) is obtained, without restriction, directly from Equation (14). Given the definitions of normalized abscissas and weights from Equation (23), we obtain

$$G(\theta, X, a_0 b_i) = C_0 \frac{D_m}{\rho_L}\left[X - X_e^f \exp(2\sigma M/\rho_L R_G T a_0 b_i)\right]\frac{F}{a_0 b_i}. \qquad [26]$$

NUMERICAL RESULTS

To simultaneously solve the energy, vapor, and moment equations we use the method of lines and a finite difference formulation of the energy and mass transport equations in the radial coordinate, writing separate equations at each radial location for the energy, vapor, and aerosol moments. To march along the axial coordinate, we use a fourth-order Runge-Kutta algorithm from Gerald and Wheatley (1989). The solution grid is two dimensional with a radial step size of 0.02 and an axial step size of 10^{-6} in nondimensional coordinates. The solution technique simultaneously solves for the vapor and condensate phases, thereby eliminating the need to iterate the vapor transport and aerosol equations. All calculations are performed in double precision.

The method of moments is equally applicable to discrete and continuous distributions. To validate the models, comparisons were made for the discrete monodisperse and bidisperse cases of Davis and Liao (1975). For these discrete cases, the abscissas and weights obtained from inversion of the moments (McGraw 1997) leads directly to particle size.

Several comparisons were made for the conventional and quadrature models. First, we ran the conventional model as formulated above. Second, we ran the quadrature model for the same approximated growth law used in the conventional method. Finally, we ran the quadrature model using the full nonlinear growth law. For the monodisperse case, the average particle radii from the conventional model and the quadrature model with the approximated growth law were virtually identical at 0.273 μm. This value compares favorably with Davis and Liao (1975), reported at 0.26 μm. Calculated results from the quadrature model using the nonlinear growth law gave an average particle radius of 0.2749 μm. Having validated the quadrature approach, we proceed to apply the model to a continuous polydisperse seed particle distribution representing the gamma probability density distribution with six moments. In addition, we discuss special solutions for two and four moments.

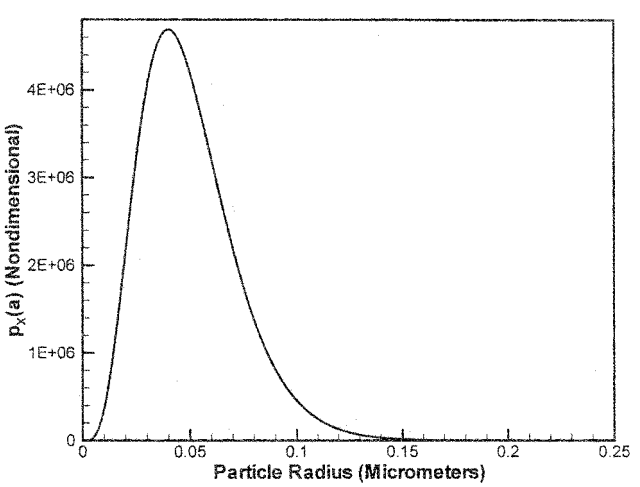

Figure 3. Two parameter gamma probability density distribution function with $\alpha = 5.0$ and $\beta = 1.0 \times 10^{-6}$ for $p_X(a) = a^{\alpha-1}e^{-\alpha/\beta}/[\beta^\alpha \Gamma(\alpha)]$, where $\Gamma(\alpha)$ is the usual gamma function.

Gamma Probability Density Distribution

The value of moment methods to describe particle evolution is best realized when the initial distribution is polydisperse. In these cases, for example with bin or sectional models, the number of equations needed to describe particle evolution becomes large as the particle distribution is partitioned into a large number of bins that must be tracked.

As an example of a polydisperse seed particle distribution, consider a continuous two parameter gamma probability density distribution. A plot of this density distribution with $\alpha = 5$ and $\beta = 1.0 \times 10^{-6}$ cm is shown in Figure 3. These parameters were chosen to yield a narrow initial distribution with high number densities in the same size region as the monodisperse and bidisperse reference cases from Davis and Liao (1975). Specifically, we set a_0 to the peak of the distribution by using $\beta = a_0$. The analytic expression for the reduced moments of the two parameter gamma distribution is

$$(\tilde{\mu}_k)_0 = \frac{\beta^k}{a_0^k} \frac{\Gamma(\alpha + k)}{\Gamma(\alpha)}. \qquad [27]$$

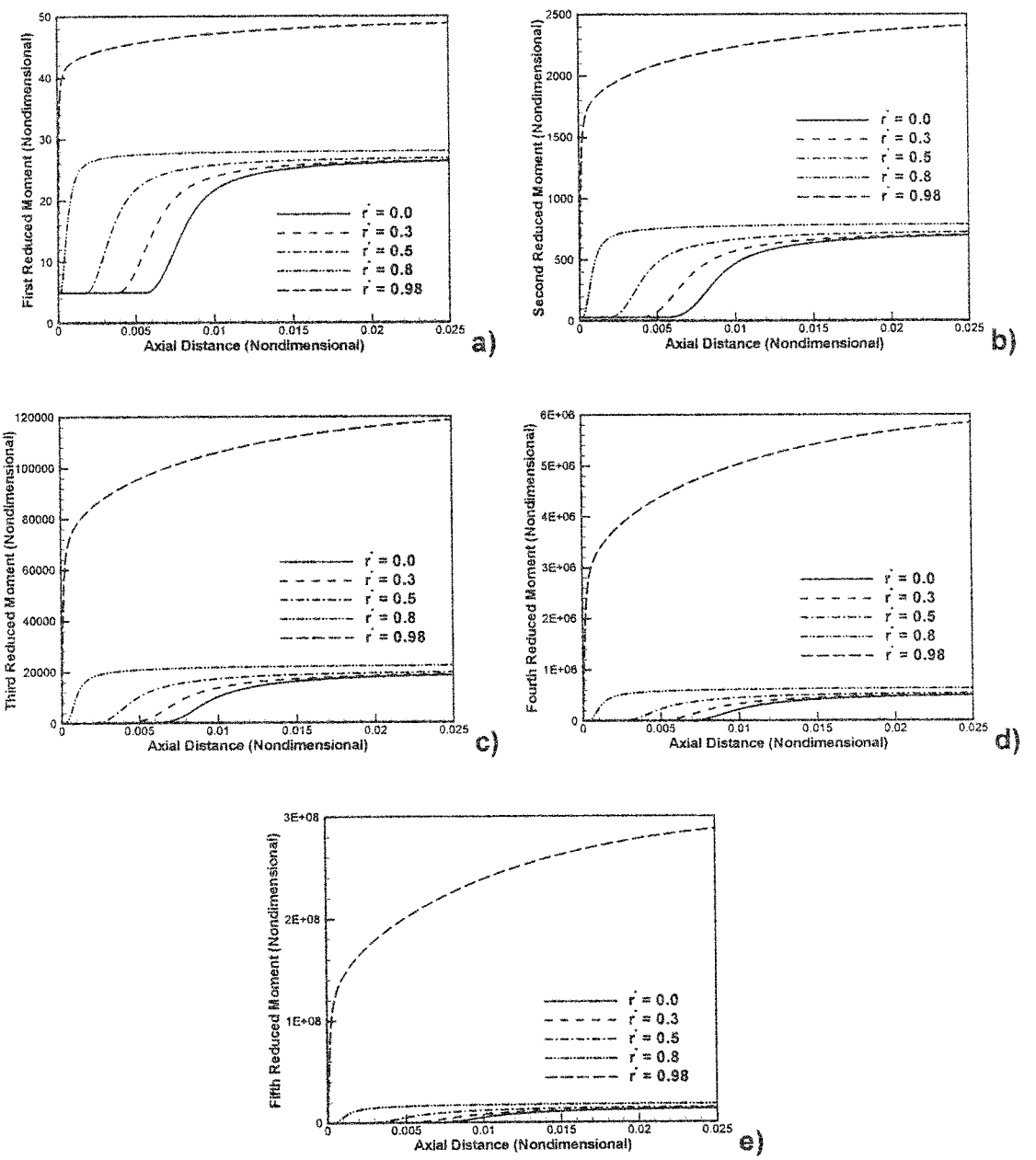

Figure 4. Evolution of (a) first, (b) second, (c) third, (d) fourth, and (e) fifth reduced moments for two parameter gamma probability density distribution represented by six moments ($T_w = 60°$C, $\alpha_T = 0.16$ cm^2/s, $D_m = 1.8$ cm^2/s).

For the parameters shown in Figure 3, Equation (27) gives the initial conditions for the lowest six moments:

$$(\tilde{\mu}_0)_0 = 1, \quad (\tilde{\mu}_1)_0 = 5, \quad (\tilde{\mu}_2)_0 = 30, \quad (\tilde{\mu}_3)_0 = 210,$$
$$(\tilde{\mu}_4)_0 = 1680, \quad (\tilde{\mu}_5)_0 = 15120. \qquad [28]$$

Six Reduced Moment Solution

The expanded set of source terms for the reduced moment equations for six moments follows from Equation (25) as

$$[\psi_S]_n = \frac{r_0^2}{D_m}\frac{n}{a_0}\left[b_1^{n-1}G\gamma_{11}^2 + b_2^{n-1}G\gamma_{12}^2 + b_3^{n-1}G\gamma_{13}^2\right], \qquad [29]$$

where n goes from 0 to 5 and G represents the growth law. With $n = 0$ we have the solution for the zeroeth moment, $\tilde{\mu}_0 = 1$, reflecting the fact that no particles are nucleated or lost in the model. The sink function for vapor loss, which is proportional to surface area, is obtained from Equation (24):

$$\phi_S = \frac{r_0^2}{C_0}\frac{\rho_L}{D_m}a_0^2 N_T 4\pi\left[b_1^2 G\gamma_{11}^2 + b_2^2 G\gamma_{12}^2 + b_3^2 G\gamma_{13}^2\right]. \qquad [30]$$

Results for the evolution of the reduced moments are presented in Figure 4. The zeroeth moment, which is constant, is not shown. The results for the first through fifth reduced moments are shown in subplots (a)–(e).

Characteristics of vapor diffusion and particle growth are revealed by examining Figure 4a. Because the flow is cooled from the outside, in the outermost streamline ($r^* = 0.98$) the first reduced moment (proportional to particle radius) rises rapidly. Because the flow is also moving slowly, the particles have sufficient time to grow large for a given axial distance and exceed the average particle radius calculated below as vapor diffuses from the inner to outer regions of the flow. As a consequence, a deficit of vapor develops at the inner streamlines, and as the inner regions of the flow are cooled, particle growth is quenched early by depletion of available vapor.

To test numerical accuracy, calculations to verify conservation of mass have been performed as shown in Figure 5a. As seen in the figure, as the amount of condensate increases, the amount of vapor decreases. The total, as a summation of the vapor and condensate phases, remains constant. We also compute the effective particle radius (Figure 5b) across the radial dimension of the pipe using the ratio of the third and second reduced moments weighted by mass flux as a function of radial position. This quantity ranges from about 7 nondimensional units, or 0.07 μm, at the entrance to 0.2749 μm at mixout. Though a direct comparison is not truly appropriate, these results are in good agreement with the monodisperse and bidisperse cases of Davis and Liao (1975). Specifically, as mentioned previously, Davis and Liao (1975) reported an average particle radius of 0.26 μm for the monodisperse case.

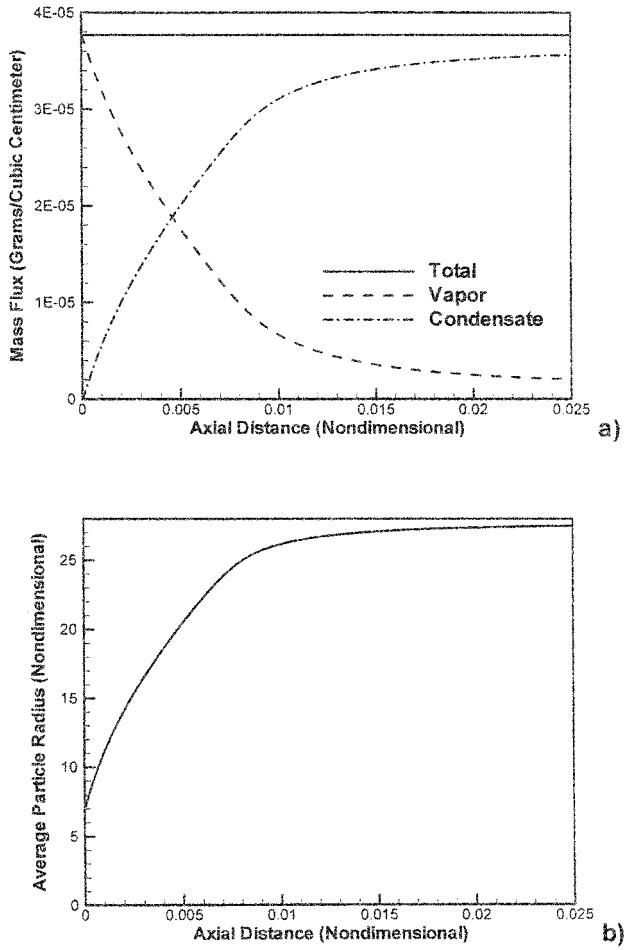

Figure 5. Evolution of (a) mass flux and (b) effective particle radius for two parameter gamma probability density distribution represented by six moments ($T_w = 60°$C, $\alpha_T = 0.16$ cm^2/s, $D_m = 1.8$ cm^2/s).

Special Two and Four Reduced Moment Solutions

Special, or restricted, solutions of Equations (29) and (30) are obtained for 1-point and 2-point quadrature. For 1-point quadrature (one abscissa and one weight), only the zeroeth and first moment are tracked. With 2-point quadrature we expand Equation (30) using two abscissas and weights for $n = 0$ through 3. In each of these solutions, as in the six moment solution, the zeroeth moment is constant. For 1- and 2-point quadrature, in which we calculate μ_1 and μ_1–μ_3, respectively, the evolution of the reduced moments are similar graphically to the results in Figures 4a–c.

Quantitative comparisons among 1-, 2-, and 3-point quadrature are given in Table 3 at five radial positions for the nondimensional axial location $x^* = 0.020$. Of particular interest are the results for the first reduced moments. Results show convergence is achieved with 3-point quadrature for our numerical example. This conclusion is further verified by comparing the second and third reduced moments. For completeness we also list the

Table 3
Comparison of 1-, 2-, and 3-point quadrature at $z^* = 0.020$

Radial position (r^*)	1-point	2-point	3-point
	First reduced moment		
0.0	26.1192153438	26.0537151350	26.0534989772
0.3	26.3239771404	26.2593802022	26.2591668791
0.5	26.8320390945	26.7694823358	26.7692758632
0.8	28.0770486966	28.0192003052	28.0190125384
0.98	48.5751978818	48.5365906121	48.5352864570
	Second reduced moment		
0.0	—	681.5426890448	681.5355798244
0.3	—	692.2859552988	692.2788852220
0.5	—	719.2973987666	719.2904145466
0.8	—	787.6757191112	787.6691136662
0.98	—	2375.0293326 4	2374.9541900 6
	Third reduced moment		
0.0	—	17903.909595 1	17903.909028 8
0.3	—	18326.460223 9	18326.460149 4
0.5	—	19403.307974 4	19403.308212 5
0.8	—	22219.500520 9	22219.508285 0
0.98	—	116142.61245 6	116143.45778 3
	Fourth reduced moment		
0.0	—	—	472420.782867
0.3	—	—	487254.637484
0.5	—	—	525566.930232
0.8	—	—	629056.309086
0.98	—	—	5680900.82782
	Fifth reduced moment		
0.0	—	—	1.252337994×10^7
0.3	—	—	1.301368638×10^7
0.5	—	—	1.429690705×10^7
0.8	—	—	1.787628789×10^7
0.98	—	—	2.780312806×10^8

fourth and fifth reduced moments from the 3-point quadature calculations.

It is tempting to view the abscissas and weights as having a direct physical correlation to the size distribution. However, quadrature abscissas and weights are more appropriate for obtaining integral properties of the distribution rather than providing a representation of the distribution itself. Other retrieval techniques are more appropriate for obtaining continuous distributions for moments (Yue et al. 1997; Wright 2000).

DISCUSSION

The focus of this paper has been to compare the capabilities of the conventional and quadrature methods of moments using a well-known laminar flow aerosol reactor model. We presented formulations of a simple two-dimensional laminar flow reactor model using both of these approaches to obtain closure of the coupled aerosol equations. In writing the aerosol dynamic equation we eliminated a priori assumptions for the particle size distributions of earlier models by applying the quadrature method of moments. Subsequently, numerical results are presented for a six moment solution of a laminar flow reactor model using the quadrature method. Conclusions are drawn regarding the vapor diffusion processes which contribute to excess particle growth in the outer regions of the flow and retard growth in the inner regions of the flow.

Significant advantages have been achieved with the quadrature method of moments. Restrictive constraints to obtain closure with the conventional method of moments have been avoided. In removing these constraints, the quadrature method allows virtually any condensation growth/evaporation law to be represented. Earlier applications of the quadrature method of moments include diffusion (McGraw 1997) and coagulation (Barrett and Webb 1998) for zero-dimensional box models. This

paper presents the first calculations with the new quadrature method of moments for a more realistic aerosol transport and growth model.

REFERENCES

Barrett, J. C., and Fissan, H. (1989). Wall and Aerosol Condensation during Cooled Laminar Tube Flows, *J. Colloid Interface Sci.* 130:498.

Barrett, J. C., and Webb, N. A. (1998). A Comparison of Some Approximate Methods for Solving the Aerosol General Dynamic Equation, *J. Aerosol Sci.* 29:31.

Bird, R. B., Stewart, W. E., and Lightfoot, E. N. (1960). *Transport Phenomena*, John Wiley and Sons, New York, Equation. 16.5-4.

Davis, E. J., and Liao, S. C. (1975). The Growth Kinetics and Polydispersity of Condensational Aerosols, *J. Colloid Interface Sci.* 50:488.

Dean, J. A. (1985). *Lange's Handbook of Chemistry*, 13th ed., McGraw-Hill Book Company, New York, Table 10–35.

Frenklach, M., and Harris, S. J. (1987). Aerosol Dynamics Modeling using the Method of Moments, *J. Colloid Interface Sci.* 118:252.

Friedlander, S. K. (1977). *Smoke, Dust, and Haze; Fundamentals of Aerosol Behavior*, John Wiley and Sons, New York, pp. 14–17.

Friedlander, S. K. (1983). Dynamics of Aerosol Formation by Chemical Reaction, *Ann. N.Y. Acad. Sci.* 404:354.

Fuchs, N. A., and Sutugin, A. G. (1970). *Highly Dispersed Aerosols*, Ann Arbor Science Publishers, Ann Arbor, MI.

Gerald, C. F., and Wheatley, P. O. (1989). *Applied Numerical Analysis*, 4th ed., Addison-Wesley Publishing Company, Menlo Park, CA, pp. 356–363.

Hulburt, H. M., and Katz, S. (1964). Some Problems in Particle Technology; A Statistical Mechanical Formulation, *Chem. Eng. Sci.* 19:555.

Kemppinen, A. I., and Gokcen, N. A. (1956). Density of Dibutylphthalate, *J. Phys. Chem.* 60:126.

Liao, S.-C. (1974). *Condensational Aerosol Formation in Nonuniform Temperature, Concentration and Flow Fields*, Ph.D. Dissertation, Clarkson College of Technology, Chemical Engineering, Potsdam, NY, pp. 127.

McGraw, R. (1997). Description of Aerosol Dynamics by the Quadrature Method of Moments, *Aerosol Sci. Technol.* 27:255.

McGraw, R., Huang, P. I., and Schwartz, S. E. (1995). Optical Properties of Atmospheric Aerosols from Moments of the Particle Size Distribution, *Geophys. Res. Lett.* 22:2929.

McGraw, R., and Saunders, J. H. (1984). A Condensation Feedback Mechanism for Oscillatory Nucleation and Growth, *Aerosol Sci. Technol.* 3: 367.

Nicolaon, G., Cooke, D. D., Davis, E. J., Kerker, M., and Matijevic, E. (1971). A New Liquid Aerosol Generator II. The Effect of Reheating and Studies on the Condensation Zone, *J. Colloid Interface Sci.* 35:490.

Nicolaon, G., Cooke, D. D., Kerker, M., and Matijevic, E. (1970). A New Liquid Aerosol Generator, *J. Colloid Interface Sci.* 34:534.

Pesthy, A. J., Flagan, R. C., and Seinfeld, J. H. (1983). Theory of Aerosol Formation and Growth in Laminar Flow, *J. Colloid Interface Sci.* 91: 525.

Phanse, G. M., and Pratsinis, S. E. (1989). Theory for Aerosol Generation in Laminar Flow Condensers, *Aerosol Sci. Technol.* 11:100.

Pratsinis, S. E. (1988). Simultaneous Nucleation, Condensation, and Coagulation in Aerosol Reactors, *J. Colloid Interface Sci.* 124:416.

Press, W. H., Teukolsky, S. A., Vetterling, W. T., and Flannery, B. P. (1992). *Numerical Recipes in FORTRAN 77, Second Edition, The Art of Scientific Computing*, Cambridge University Press, New York, p. 150.

Randolph, A. D., and Larson, M. A. (1988). *Theory of Particulate Processes: Analysis and Techniques of Continuous Crystallization*, 2nd ed., Academic Press, New York, pp. 62–67.

Ray, A. K., Davis, E. J., and Ravindran, P. (1979). Determination of Ultra-Low Vapor Pressures by Submicron Droplet Evaporation, *Chem. Phys.* 71: 582.

Sherwin, M. B., Shinnar, R., and Katz, S. (1967). Dynamic Behavior of the Well-Mixed Isothermal Crystallizer, *AIChE J.* 13:1141.

Sinclair, D., and LaMer, V. K. (1949). Light Scattering as a Measure of Particle Size in Aerosols, *Chem. Rev.* 44:245.

Stratmann, F., and Whitby, E. (1989). Heterogeneous Condensation in Cooled Laminar Tube Flow: A Comparison of Two Aerosol Modeling Techniques, *J. Aerosol Sci.* 20:999.

Wilck, M., and Stratmann, F. (1997). A 2-d Multicomponent Modal Aerosol Model and its Application to Laminar Flow Reactors, *J. Aerosol Sci.* 28:959.

Wright, D. L. (2000). Retrieval of Optical Properties of Atmospheric Aerosols, *J. Aerosol Sci.*, 31:1.

Yue, G. K., Lu, J., Mohnen, V. A., Wang, P.-H., Saxena, V. K., and Anderson, J. (1997). Retrieving Aerosol Optical Properties from Moments of the Particle Size Distribution, *Geophys. Res. Lett.* 24:651.