



Description of Aerosol Dynamics by the Quadrature Method of Moments

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ABSTRACT. The method of moments (MOM) may be used to determine the evolution of the lower-order moments of an unknown aerosol distribution. Previous applications of the method have been limited by the requirement that the equations governing the evolution of the lower-order moments be in closed form. Here a new approach, the quadrature method of moments (QMOM), is described. The dynamical equations for moment evolution are replaced by a quadrature-based approximate set that satisfies closure under a much broader range of conditions without requiring that the size distribution or growth law maintain any special mathematical form. The conventional MOM is recovered as a special case of the QMOM under those conditions, e.g., free-molecular growth, for which conventional closure is satisfied. The QMOM is illustrated for the growth of sulfuric acid-water aerosols and simulations of diffusion-controlled cloud droplet growth are presented. AEROSOL SCIENCE AND TECHNOLOGY 27:255-265 (1997) © 1997 American Association for Aerosol Research

INTRODUCTION

The method of moments (MOM) is a powerful approach with unique advantages for describing aerosol dynamics under conditions that can include new particle formation, evaporation, growth by condensation and coagulation, and complex mixing flows (Hulburt and Katz, 1964; McGraw and Saunders, 1984; Pratsinis, 1988; Jurcik and Brock, 1993; LaViolette et al., 1996). The MOM solves the problem by tracking the time dependence of just the lower-order radial moments of the size distribution. These lower-order moments are often sufficient for estimating the physical properties (Friedlander, 1977) and optical properties (McGraw et al., 1995) of the simulated aerosol, whereas the full aerosol size distribution

generally contains much more information than is required for many applications. Conversely, the lower-order moments of an aerosol distribution may be inferred from measurement, e.g., from multiwavelength particulate extinction measurements (Livingston and Russell, 1989) and compared with the moments obtained from simulations using the MOM.

The k th radial moment of an aerosol size distribution is defined as

$$\mu_k = \int r^k f(r) dr, \quad (1)$$

where $f(r)$ is the distribution function for the number density of particles having radii in the range r to $r + dr$. The key to the

MOM is that the lower-order moments can be tracked directly without requiring additional knowledge of the distribution. The conventional MOM accomplishes this feat by formulating equations for evolution of the moments in closed form, i.e., involving only functions of the moments themselves. However, the closure requirement is a severe restriction on the method, which is probably the main reason for the method not having received more widespread application. Necessary and sufficient conditions for closure are described in the following text.

In this paper, a new approach, the quadrature method of moments (QMOM), is introduced through which the moments of f are tracked in time directly, just as in the conventional case, but for which the requirement of exact closure is replaced by an approximate closure condition that allows the method to be applied under a much broader range of conditions. In addition, the QMOM is shown to include the ordinary MOM as a special case that is recovered for those special growth-law conditions (Eq. 3) for which exact closure is obtained.

MOMENT EVOLUTION EQUATIONS

A coupled set of general dynamic equations (GDEs) suitable for describing aerosol formation in complex flow fields may be written as (Hulburt and Katz, 1964)

$$\begin{aligned} \frac{\partial}{\partial t} f_1 &= R - S - \nabla \cdot D \nabla f_1 \\ &\quad - \nabla \cdot (f_1 \mathbf{v}) + \left(\frac{\partial}{\partial t} f_1 \right)_{\text{nucl}} \\ &\quad - \frac{4\pi}{v_1} \int r^2 \phi(r) f(r) dr, \end{aligned} \tag{2a}$$

$$\frac{\partial}{\partial t} \mu_0 = -\nabla \cdot D \nabla \mu_0 - \nabla \cdot (\mu_0 \mathbf{v}) + \int J(r) dr, \tag{2b}$$

and

$$\begin{aligned} \frac{\partial}{\partial t} \mu_k &= -\nabla \cdot D \nabla \mu_k - \nabla \cdot (\mu_k \mathbf{v}) + \int r^k J(r) dr \\ &\quad + k \int r^{k-1} \phi(r) f(r) dr \end{aligned} \tag{2c}$$

for moment index $k \geq 1$. Here R and S are source and sink rates respectively, for monomer (f_1) production, D is the eddy diffusion constant for turbulent mixing, \mathbf{v} is the local flow velocity, $J(r)$ is the nucleation rate, and $\phi(r)$ is the particle growth law, $\phi(r) \equiv dr/dt$. The last two terms in Eq. 2a represent monomer loss to new particle formation and to growth where, in the growth term, v_1 is the molecular volume.

Equations 2 cannot be solved in their present form because the growth-law terms appearing in Eqs. 2a and 2c each involve integration over a distribution function $f(r)$ that is unknown. Much of the success of the conventional MOM has been due to the fact that there are special cases of practical importance where the growth function $\phi(r)$ is such that a closed set of equations in terms of the moments of $f(r)$ is obtained. Hulburt and Katz (1964) have shown that the necessary and sufficient conditions for exact closure of Eq. 2 is a growth law of the form

$$\phi(r) = a + br, \tag{3}$$

where a and b are independent of r . For $b = 0$, Eq. 3 describes particle growth in the free-molecular size regime, a well known result treated in previous applications of the MOM (e.g., McGraw and Saunders, 1984) that will not be discussed further here. The case $a = 0$ applies under certain quasi-equilibrium conditions to describe the growth of solution droplets as subsequently discussed for sulfuric acid-water droplets. When Eq. 3 pertains, the growth term in Eq. 2c becomes

$$\begin{aligned} k \int r^{k-1} \phi(r) f(r) dr \\ &= ak \int r^{k-1} f(r) dr + bk \int r^k f(r) dr \\ &= ak \mu_{k-1} + bk \mu_k. \end{aligned} \tag{4}$$

The same substitution solves the integral in Eq. 2a, which describes monomer loss from the vapor due to particle growth. Reduction of the growth-law integrals in this fashion results in a closed set of five coupled nonlinear equations for the evolution of f_1 and moments μ_0 – μ_3 and is the basis for the conventional MOM. (For $b=0$ only the four equations through μ_2 are required for closure.) As noted previously, Eq. 3 is a severe restriction on the types of growth laws that can be handled by the method. For example, the important cases of diffusion-controlled growth (subsequently discussed and size-dependent evaporation rates of small particles (Kelvin effect) are described by growth–evaporation laws that are not of the form of Eq. 3. To circumvent this limitation, the QMOM obtains closure through approximation of the growth integrals, using a much less restrictive quadrature method that will now be described.

QUADRATURE-BASED CLOSURE

The focus of the quadrature approximation is again the growth-law integrals of Eqs. 2 because these involve the unknown particle size distribution. Approximating these integrals by means of n -point Gaussian quadrature (Lanczos, 1988) we obtain

$$k \int r^{k-1} \phi(r) f(r) dr \cong k \sum_{i=1}^n r_i^{k-1} \phi(r_i) w_i \quad (5)$$

for $k \geq 1$. The essence of quadrature-based closure lies in the fact that the abscissas r_i and weights w_i may be completely specified in terms of the lower-order moments of the unknown distribution function $f(r)$. Specifically, the abscissas and weights are independent of $\phi(r)$ and $f(r)$ [beyond the dependence on $f(r)$ through its lower-order moments]. For example, the moments themselves may be written in the form of Eq. 5. For n -point quadrature these are

$$\mu_k = \int r^k f(r) dr = \sum_{i=1}^n r_i^k w_i \quad (6)$$

for $k = 0$ through $2n - 1$. Inspection of Eq. 6 shows that the first $2n$ moments (μ_k , for

$k = 0$ through $2n - 1$) determine the n abscissas and n weights. However, the direct solution of Eq. 6 for these quantities would require a nonlinear search and is not recommended. A much better approach is to use the moment sequence to construct a tridiagonal Jacobi matrix from which the quadrature abscissas and weights can be obtained. An algorithm that contains both these steps is described in Appendix A.

Substitution of the right-hand side of Eq. 5 into Eqs. 2, in place of the integrals, results in an approximate closure condition (because the equality of Eq. 5 is approximate) that does not contain the unknown distribution function $f(r)$. The transformed equations are in closed form because evolution of the μ_k is given in terms of the abscissas and weights, which, as previously described, are themselves given in terms of the lower-order moments of $f(r)$. This closure is the basis for the QMOM. In summary, the basic idea of the QMOM is to evaluate the growth contribution to the moment derivatives in Eqs. 2 using the quadrature approximation of Eq. 5. This gives a prescription for updating the moment sequence, which can then be inverted to obtain new abscissas and weights, and so on, until the time evolution of the system (Eqs. 2) has been obtained. Inspection of Eqs. 4–6 reveals that the QMOM reduces to the conventional MOM when the growth law of Eq. 3 pertains. Thus any simulation of moment evolution that can be performed using the conventional MOM can be performed using the QMOM, whereas the latter can also handle more general cases that as we will shown, are beyond the capability of the conventional approach. All that remains to implement the QMOM is a numerical means for rapid conversion from moments to quadrature abscissas and weights and this is provided in Appendix A.

CALCULATIONS

The QMOM is best demonstrated by choosing conditions that focus on handling the growth law and that are also amenable

to exact calculation by other methods for comparison. Both these features are present in spatially homogeneous box models for which complex flow field considerations can be set aside, whereas all of the essential difficulties associated with closure of the moment evolution equations remain. Returning to the moment evolution equations, we note that in the absence of new particle formation the right-hand side of Eq. 2b vanishes for the box model (μ_0 is constant), but the full growth-law term remains in Eq. 2c for the higher-order moments. Two cases with widely different growth laws will now be considered. The first of these (growth of sulfuric acid-water aerosols) we only mention as an example of a situation, which differs from the well-known case of free-molecular growth, for which the growth law follows Eq. 3 and closure in both the QMOM and the usual MOM is exact. The second example (diffusion-controlled growth) can only be handled by the QMOM and the more detailed calculations presented in subsequent text will be limited to this case.

Growth of Sulfuric Acid-Water Aerosols

In this example we consider the size change in sulfuric acid-water droplets as the relative humidity of water is changed. Exchange of water vapor is fast compared with exchange of sulfuric acid, which is generally present in the vapor in only trace amounts. As a result, droplets are in local equilibrium with respect to evaporation and condensation of water vapor. To further simplify the example, we will assume that the amount of sulfuric acid present in the droplets is unchanged over the time interval of interest and that water equilibration is sufficiently rapid that the size of each droplet is dependent only on its initial size and on the relative humidity (RH) of water. This dependence has been studied for laboratory-generated sulfuric acid aerosols (McMurry and Stolzenburg, 1989). At any given RH, each particle has a radius proportional

to its initial radius. The corresponding growth law is therefore

$$\phi(r) = k_1 r, \quad (7)$$

where k_1 is a function of time that depends on the rate of change in RH, but is independent of r . This growth law is of the type described by Eq. 3. Consequently, results from both the QMOM and the conventional MOM satisfy closure for this case.

Diffusion-Controlled Growth

Diffusion-controlled growth applies under conditions that the particle size is greater than the mean-free path of the gas, about $0.1 \mu\text{m}$ in the lower atmosphere. As a result, most processes in clouds, where particle radii typically exceed $1 \mu\text{m}$, involve diffusion-controlled growth. The growth law is (Pruppacher and Klett, 1980)

$$\phi(r) = k_2/r, \quad (8)$$

which, as this is not of the form of Eq. 3, implies that the conventional MOM equations cannot be closed. Here k_2 is a function that depends on a number of variables, including vapor concentration f_1 , but like k_1 is independent of r .

Failure of the conventional MOM for the growth law of Eq. 8 can be seen by inspecting the moment evolution equations, which in this simplified box model are

$$\begin{aligned} \frac{d}{dt} \mu_0 &= 0, \\ \frac{d}{dt} \mu_1 &= k_2 \int r^{-1} f(r) dr = k_2 \mu_{-1}, \\ \frac{d}{dt} \mu_2 &= 2k_2 \int f(r) dr = 2k_2 \mu_0. \end{aligned} \quad (9)$$

This system of equations cannot be closed off at any value of the moment index k because the equation for μ_1 involves μ_{-1} , which depends on the entire particle size distribution. Until now, the only way of applying the MOM to this problem has been to assume that the size distribution $f(r)$ maintains a special mathematical form

parameterized by its moments. For example, to obtain closure of Eqs. 9, Hulburt and Katz (1964) use the leading term of a Laguerre series expansion based on the gamma distribution to represent $f(r)$. This function is parameterized by its first three moments, in terms of which all of the other moments can be expressed. For functions in this Laguerre class (Hulburt and Katz, 1964),

$$\mu_{-1} = \frac{\mu_0^2 \mu_1}{2\mu_1^2 - \mu_0 \mu_2} \quad (10)$$

Upon substitution of this expression into Eqs. 9, a closed system of equations is obtained.

To demonstrate the new approach of the QMOM, we consider an initial distribution having the Khrgian-Mazin (KM) cloud drop size distribution form (Pruppacher and Klett, 1980),

$$f(r) = ar^2 \exp(-br), \quad (11)$$

where r is particle radius (μm) and a and b are parameters in the distribution. In this example we set $a = 0.108 \mu\text{m}^{-3} \text{cm}^{-3}$ and $b = 0.6 \mu\text{m}^{-1}$ in order to obtain a normalized distribution with an initial mean particle radius of $5 \mu\text{m}$. The lead term in the Laguerre series is satisfied by the KM distribution as a special case. Thus, in this example the initial distribution function ($t = 0$) satisfies Eq. 10 exactly and follows the Laguerre form. It is important to emphasize, however, that this special initial distribution is not a requirement for the QMOM. For r in units of μm , $k_2 = 0.78 \mu\text{m}^2/\text{s}$ in air at $T = 278 \text{K}$ and fixed water supersaturation of 101% ($S = 1.01$). The numerical value of k_2 follows the equations of Pruppacher and Klett (1980) and includes both heat and mass transfer to the water drop, but neglects departure from Eq. 8 to include corrections to the r dependence of the growth law for the transition size regime. Such corrections are readily handled in the QMOM, but are negligible for particle radius exceeding $1 \mu\text{m}$ and

neglected here for ease of illustrating the method.

Figure 1 shows the initial distribution (dotted curve) and two final distributions at time $t = 20 \text{s}$. The solid curve results from analytical propagation of the initial distribution under the growth law to obtain the exact final distribution (Eq. B3 of Appendix B). The dashed-dotted curve describes the Laguerre distribution parameterized in terms of the moments at $t = 20 \text{s}$. The latter were obtained by integrating Eqs. 9, with μ_{-1} from Eq. 10, using the moments of the $t = 0$ distribution (Eq. 11) as initial conditions. The Laguerre model fails to adequately represent the exact distribution at the later time, even though the initial distribution was selected to have the Laguerre form.

Figure 2 show the time evolution of the first five moments through 20 s for the exact distribution, for the Laguerre model, and for a QMOM simulation using three-point quadrature. Moment integrals over the exact distribution of Eq. B3 (cf. Eq. 1) were evaluated as a function of t by numerical integration (Wolfram, 1991). [Note that since the first and third of Eqs. 9 are themselves closed in this simplified model, all three approaches (QMOM, MOM, and Laguerre closure) trivially give the correct second moment, which varies linearly with time.] The QMOM simulation results are in excellent agreement with the numerical integrations over the exact evolved distribution. However, the moments obtained from Eqs. 9 and 10 for the parameterized Laguerre distribution, particularly the higher-order moments ($\mu_3 - \mu_5$), depart substantially from the exact distribution results.

Figure 3 amplifies the differences between the QMOM and exact calculations, which are unresolved in Fig. 2. The comparison is given only for the odd moments because, as previously noted, the QMOM and exact calculations are in agreement for the second moment. That they are also in agreement for the fourth moment, again a consequence of the simplified equations of

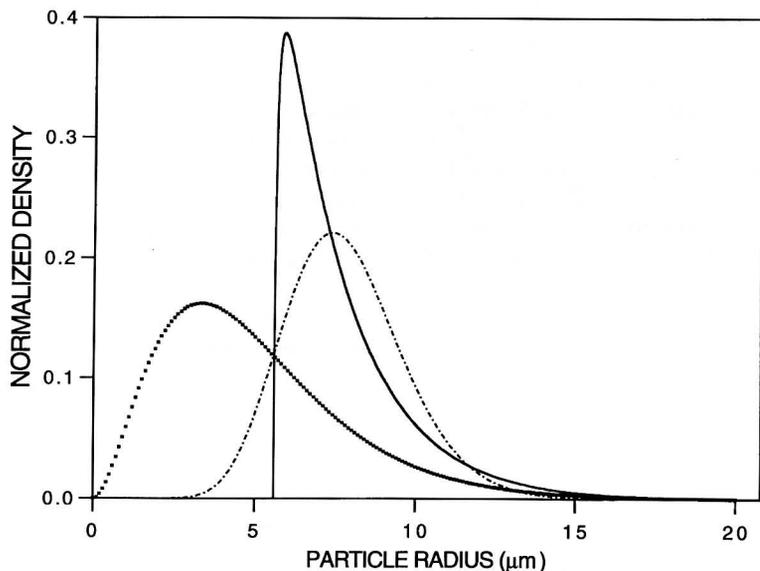


FIGURE 1. Particle size distributions. Diffusion-controlled growth of water drops at $T=278$ K and fixed supersaturation of 101% ($S=1.01$). Dotted curve, initial normalized KM distribution (Eq. 11) with mean particle radius of $5 \mu\text{m}$. Solid curve, exact distribution after 20 s from Eq. B3. Dashed-dotted curve, Laguerre distribution parameterized by the moments 0–2 after propagation to $t=20$ s using the Laguerre closure method (Eqs. 9 and 10).

the model, can be seen as follows. From Eq. 6,

$$\mu_2 = \sum_{i=1}^3 r_i^2 w_i \quad (12a)$$

and

$$\mu_4 = \sum_{i=1}^3 r_i^4 w_i. \quad (12b)$$

Differentiating gives

$$\frac{d\mu_2}{dt} = 2 \sum_{i=1}^3 w_i r_i \frac{dr_i}{dt} = 2k_2 \sum_{i=1}^3 w_i = 2k_2 \mu_0 \quad (13a)$$

and

$$\begin{aligned} \frac{d\mu_4}{dt} &= 4 \sum_{i=1}^3 w_i r_i^3 \frac{dr_i}{dt} = 4k_2 \sum_{i=1}^3 w_i r_i^2 \\ &= 4k_2 \mu_2, \end{aligned} \quad (13b)$$

where Eq. 8 has been used and the weights are unchanged because no particle loss mechanisms or sources of new particles are present in the model. Thus the evolution of the fourth moment is given by the quadratic equation in time,

$$\mu_4(t) = 4k_2^2 t^2 + 4k_2 \mu_2(0)t + \mu_4(0), \quad (14)$$

in both the exact and QMOM descriptions. Figure 3 shows that the remaining moments obtained by the QMOM (μ_1 , μ_3 , and μ_5) do differ, on a small percentage error basis, from the exact result. The differences, which are much smaller than the errors encountered with the Laguerre closure method, evident in Fig. 2, are seen to not necessarily increase with time. For example, in the case of μ_1 a maximum error of just two parts per thousand is reached within the first 5 s of growth, followed by decreasing error with time for the remainder of the calculation period—a possible result of quadrature accuracy being favored

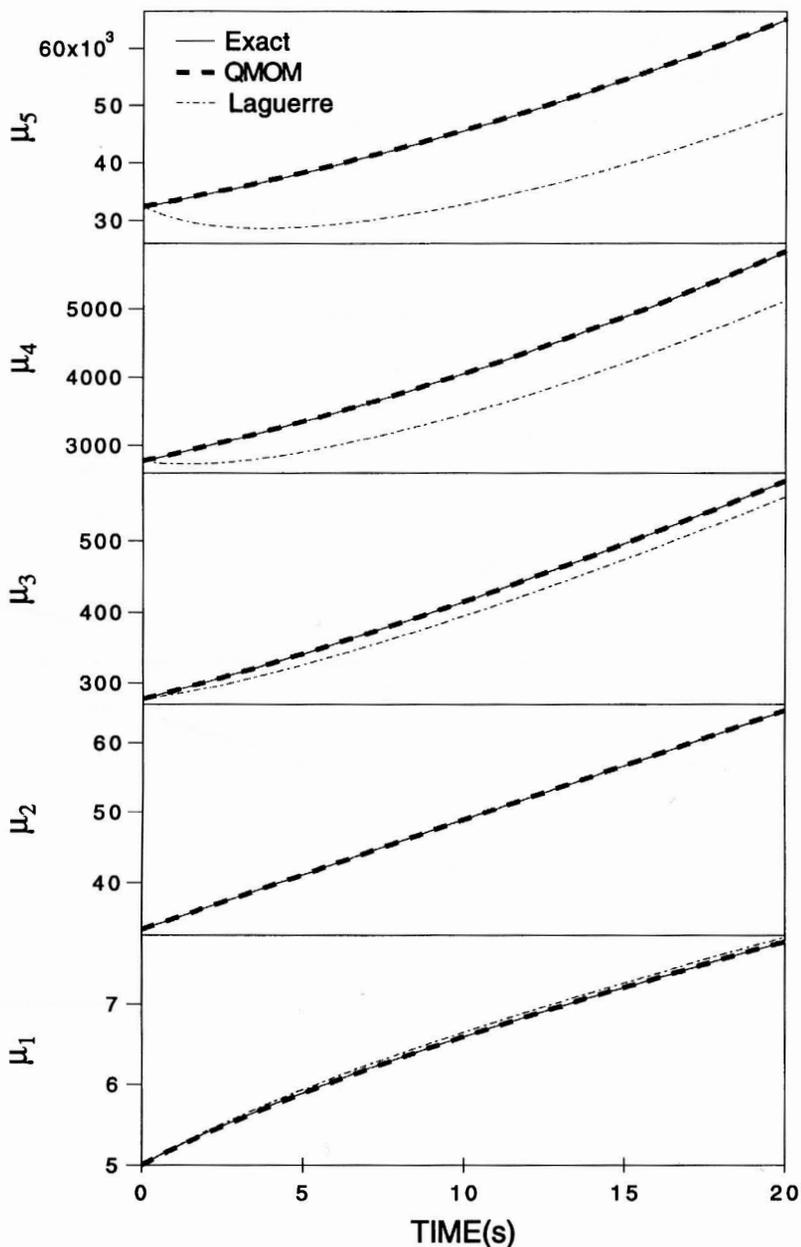


FIGURE 2. Moment evolution for diffusion-controlled growth. Conditions are the same as in Fig. 1. The k th moment has units of $(\mu\text{m})^k$. Solid curves denote moments obtained from exact propagation of the initial test distribution shown in Fig. 1. Heavy dashed curves denote moments obtained using the QMOM. Dashed-dotted curves denote moments obtained from integration of Eqs. 9 and 10 using the Laguerre closure method. Note that both the QMOM and Laguerre closure methods are exact for the second moment, which varies linearly with time as a consequence of the simplified equations of the box model (Eqs. 9).

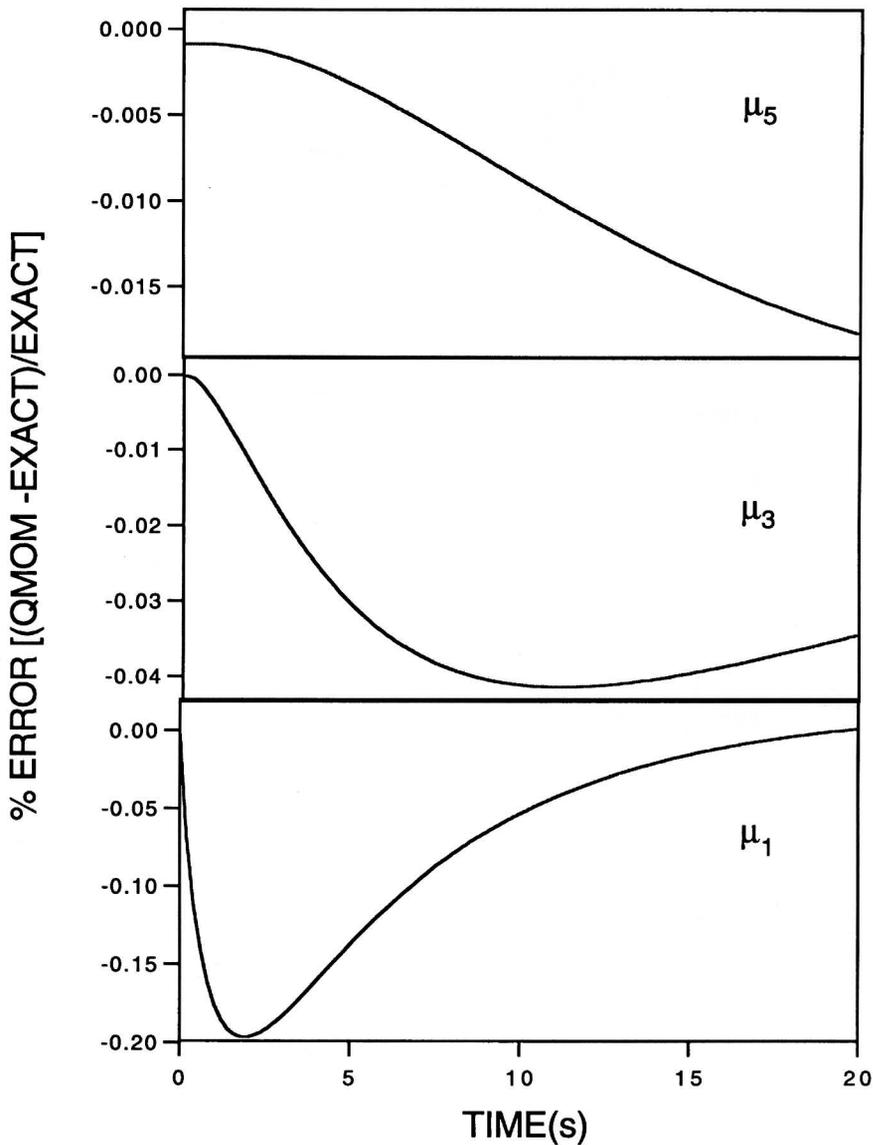


FIGURE 3. Percent error in the QMOM calculations of Fig. 2 for diffusion-controlled growth. Results for only the odd order moments are shown. The QMOM and exact calculations are in full agreement for the zeroth, second, and fourth order moments as a consequence of the simplified equations of the box model (Eqs. 9).

by the narrowing of the particle size distribution with time seen in Fig. 1.

SUMMARY

The advantages of the QMOM can be summarized as follows: As in the conventional

method of moments, no explicit form for the distribution function $f(r)$, present in the growth-law terms in Eqs. 2, is required to construct a closed set of equations for evolution of the radial moments of $f(r)$. Unlike the conventional method of moments, the QMOM does not require that

the growth law be in the restricted form of Eq. 3 for this closure of the moment evolution equations to be obtained, nor is it necessary for closure to assume that the particle size distribution maintain any special mathematical form. Finally, the conventional MOM is recovered as a special case of the QMOM under those conditions, e.g., free-molecular growth, for which Eq. 3 is satisfied.

The preceding calculations clearly show the drawback of using an assumed particle size distribution to represent $f(r)$ in achieving moment closure. Even if the assumed distribution is known to be accurate at a given time, it may not be accurate at subsequent times during the evolution of the aerosol. Moreover, apart from special cases where exact solution is possible by other methods, the accuracy of such assumed model distributions is not easily assessed. Higher-order functions parameterized using more moments, including higher order Laguerre functions, can be incorporated into a conventional MOM description, but these are subject to the same problem of assuming a distribution form. Furthermore, a different procedure must be employed to achieve closure whenever there is any change in either the model distribution or the growth law (e.g., new analogs of Eq. 10 must be derived). In contrast, the QMOM is easily implemented numerically because the method does not require an assumed model distribution and because the algorithm, governing the conversion between moments and quadrature abscissas and weights, is independent of the mechanism of aerosol growth.

APPENDIX A: INVERSION OF MOMENT SEQUENCES

Inversion of the lower-order radial moment sequence (here applied to the six moments $\mu_0-\mu_5$) to generate the quadrature abscissas and weights for use in Eq. 5 proceeds in two steps. First, a 3×3 symmetric tridiagonal matrix is constructed whose diagonal elements $\{a_1, a_2, a_3\}$ and off-diagonal elements $\{b_1, b_2\}$ are derived from the mo-

ments using the product-difference (PD) algorithm (Gordon, 1968). In the second step the 3×3 symmetric tridiagonal matrix is diagonalized by conventional methods to obtain the three abscissas and the three weights. Each of these steps is now described.

The PD algorithm proceeds in a sequence of steps beginning with setting up a triangular array of elements $P(i, j)$. Elements of the first column are

$$P(i, 1) = \delta_{i,1}, \tag{A1}$$

where $\delta_{i,1} = 0$ for $i \neq 1$ and $\delta_{i,1} = 1$ for $i = 1$. The second column contains the moments with alternating sign:

$$P(i, 2) = (-1)^{i-1} \mu_{i-1}. \tag{A2}$$

Without loss of generality, since the final distributions (or weights) can always be multiplied by the correct value of μ_0 , we will set $P(1,2) = \mu_0 = 1$. Remaining elements of the array are obtained via the recursion

$$P(i, j) = P(1, j-1)P(i+1, j-2) - P(1, j-2)P(i+1, j-1). \tag{A3}$$

Only the table elements along the first row are required for moment inversion. These are given by Eqs. A4, which follow. First row table elements $P(1,1)$ and $P(1,2)$ follows Eqs. A1 and A2. For the remaining elements in the first table row we use Eqs. A1-A3 to obtain

$$\begin{aligned} P(1,3) &= \mu_1 \\ P(1,4) &= \mu_2 - \mu_1^2, \\ P(1,5) &= \mu_1 \mu_3 - \mu_2^2, \\ P(1,6) &= \mu_1(-\mu_2^3 + 2\mu_1 \mu_2 \mu_3 - \mu_3^2 - \mu_1^2 \mu_4 + \mu_2 \mu_4), \\ P(1,7) &= \mu_1(\mu_2 - \mu_1^2)(-\mu_3^3 + 2\mu_2 \mu_3 \mu_4 - \mu_1 \mu_4^2 - \mu_2^2 \mu_5 + \mu_1 \mu_3 \mu_5). \end{aligned} \tag{A4}$$

The equations for $P(1,1)$ through $P(1,6)$ containing the moments μ_0 through μ_4 are as given in Gordon. We include the next term $P(1,7)$ in Eq. A4, which is necessary for inversion of moments μ_0 through μ_5 in the following text. From Eq. A4 a new vector (α) is generated beginning with the assignment of $\alpha(1) = 0$ and continuing with

$$\alpha(n) = P(1, n + 1) / [P(1, n)P(1, n - 2)] \quad (\text{A5})$$

for $n \geq 2$. The first few members of the series are

$$\begin{aligned} \alpha(2) &= \mu_1, \\ \alpha(3) &= (\mu_2 - \mu_1^2) / \mu_1, \\ \alpha(4) &= (\mu_1 \mu_3 - \mu_2^2) / (\mu_1 \mu_2 - \mu_1^3). \end{aligned} \quad (\text{A6})$$

The expressions for $\alpha(5)$ and $\alpha(6)$ are more complicated and will not be given here because they are easily generated from Eqs. A5 and A4. [The recursive algebra associated with evaluations of Eqs. A3 and A5, and generation of Eq. A8 in the following text, is most easily handled using a symbolic computation program such as Mathematica (Wolfram, 1991).] Finally, the matrix elements are obtained as sums and products of the $\alpha(n)$,

$$\begin{aligned} a_n &= \alpha(2n) + \alpha(2n - 1), \\ b_n^2 &= \alpha(2n + 1)\alpha(2n), \end{aligned} \quad (\text{A7})$$

and b_n is obtained as the positive square root of b_n^2 . The first members of the series are

$$\begin{aligned} a_1 &= \mu_1 \\ b_1^2 &= \mu_2 - \mu_1^2, \\ a_2 &= (\mu_1^3 - 2\mu_1\mu_2 + \mu_3) / (\mu_2 - \mu_1^2), \\ b_2^2 &= (-\mu_2^3 + 2\mu_1\mu_2\mu_3 - \mu_3^2 \\ &\quad - \mu_1^2\mu_4 + \mu_2\mu_4) / (\mu_2 - \mu_1^2)^2. \end{aligned} \quad (\text{A8})$$

The expression for a_3 is more complicated, but because it is easily generated from Eqs. A5 and A7, we omit it here. This completes the PD algorithm and construction of the symmetric tridiagonal matrix,

$$\mathbf{J}_3 = \begin{pmatrix} a_1 & b_1 & \\ b_1 & a_2 & b_2 \\ & b_2 & a_3 \end{pmatrix}, \quad (\text{A9})$$

which is also known as the Jacobi matrix (Press and Teukolsky, 1990). We now describe the second step, computation of the quadrature abscissas and weights.

Once the Jacobi matrix has been determined, generation of the abscissas and weights for a quadrature formula such as Eq. 5 is straightforward (Press and Teukolsky, 1990). All that is required is solution of the eigenvalue problem associated with the Jacobi matrix (\mathbf{J}). The abscissas r_j are simply the eigenvalues of \mathbf{J} and the weights are obtained in terms of the corresponding eigenvectors \mathbf{v}_j using the Christoffel-Darboux identity (Press and Teukolsky, 1990),

$$w_j = \mu_0 v_{j1}^2, \quad (\text{A10})$$

where v_{j1} is first component of eigenvector \mathbf{v}_j . For a normalized distribution $\mu_0 = 1$. Table 1 shows the results of applying moment inversion to the moments of the initial distribution of Figs. 1 and 2. As a check, the moments (column 2) are recovered upon substitution of the abscissas and weights (columns 5 and 6, respectively) into

TABLE 1. Inversion of Moments from the Initial ($t=0$) Distribution of Figs. 1 and 2

n	$\mu_n (t=0)$	a_n	b_n	r_n	w_n
0	1.0				
1	5.0	5.0	2.88675	15.2853	0.0283736
2	33.3333	8.33343	4.7139	7.18626	0.452838
3	277.778	11.6673		2.52914	0.518788
4	2777.78				
5	32407.4				

Eq. 6. Columns 3 and 4 contain the a_n and b_n (positive root of b_n^2) calculated using Eqs. A4, A5, and A7.

APPENDIX B: EXACT SIZE DISTRIBUTION EVOLUTION FOR DIFFUSION-CONTROLLED GROWTH

Integration of the diffusion-controlled growth law of Eq. 8 gives

$$(r')^2 = r^2 + 2k_2t, \quad (\text{B1})$$

where r is the initial particle radius and r' is the particle radius after time t . Let $f_0(r)$ be the distribution at $t = 0$ and $f(r)$ denote the corresponding distribution at time t . Conservation of particle number implies

$$f(r') dr' = f_0(r) dr. \quad (\text{B2})$$

Combining Eqs. B1 and B2, and expressing results in terms of the original radius variable r , we obtain

$$f(r) = \frac{r}{\sqrt{r^2 - 2k_2t}} f_0\left(\sqrt{r^2 - 2k_2t}\right), \quad (\text{B3})$$

for the particle size distribution at time t . Note that $f(r)$ is set to zero for negative argument of the square root.

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