

# Simulation of Multicomponent Aerosol Condensation by the Moving Sectional Method

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The moving sectional method is extended to simulate multicomponent aerosol dynamics resulting from condensation/evaporation processes. This method uses a Lagrangian approach in which section boundaries and component masses in a section vary according to the characteristics of condensation/evaporation rates while conserving number concentration in a section throughout the simulation. Simulation of model problems for which new analytical solutions have been obtained shows excellent agreement with the analytical solutions. Limitations and applicability of the sectional method are discussed.

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## 1. INTRODUCTION

The aerosol general dynamic equation (GDE) presents unique computational challenges because of the inherently different nature of coagulation and condensation/evaporation processes. In addition, the aerosol may be of a single component or may contain several components. The simplest problems to address computationally are those involving a single component and either, but not both, coagulation or condensation/evaporation; the most demanding involve several components and simultaneous coagulation and condensation/evaporation.

Considerable effort has been devoted to the development of numerical techniques for solving the aerosol GDE (13). These efforts can be classified according to whether they address single component or multicomponent aerosols and which processes, notably coagulation and/or condensation/evaporation, are occurring. Table I summarizes many of the previous approaches organized by these two criteria.

Most of the numerical methods that have been proposed are based on dividing the size

distribution into elements or sections and assuming the form of the size distribution within these elements. The assumption of a constant distribution leads to the so-called "sectional method" (1, 4, 5). Additionally, the elements or sections may remain fixed throughout the calculation or move. Fixed elements are desirable for representing coagulation; moving elements are preferable for condensation/evaporation. The original sectional method, fixed elements with a constant distribution within elements, is ideally suited for simulating coagulation but suffers from numerical diffusion errors when condensation/evaporation occur (20, 22).

The aerosol size distribution function during condensation/evaporation is described by a first-order hyperbolic partial differential equation for which the occurrence of numerical dispersion and diffusion errors in numerical solutions is well known (7, 12). Discrete approximation of spatial derivatives of a hyperbolic differential equation may lead to significant numerical dispersion and diffusion errors. Numerical *dispersion* causes the modes of the numerical solution to propagate at speeds different from those of the exact solution, resulting in spurious oscillations around the true solution peak and/or negative values.

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TABLE I  
Approaches to the Numerical Simulation of Aerosol Dynamics

Process	Single component	Multicomponent
Condensation/evaporation only	Lagrangian calculation (9) Finite element method incorporating characteristics and combined Eulerian-Lagrangian scheme (17) Galerkin finite element method and Smolarkiewicz repeated upwinding scheme (19)	Moving sectional method ((6) and this work)
Coagulation only	J-space transformation with cubic spline method (16) Sectional method (4)	Sectional method (5)
Condensation/evaporation and coagulation	J-space transformation with cubic spline method (8) Orthogonal collocation and cubic spline on finite element method (2) Methods of (16) and (17) (18) Sectional method (22, 23)	Sectional method (1)

Furthermore, numerical dispersion errors often cause instabilities since these errors generally increase with time. Numerical *diffusion* errors cause the amplitude of the numerical solution to propagate in such a way that peaks in the solution dissipate. Note that this diffusion arises from numerical errors, and is not a physical phenomenon. Also note that when the amplitude of the numerical solution is larger than that of the exact solution, the numerical solution may become unstable.

Since aerosol condensation/evaporation is particle number conserving, a numerical scheme addressed to condensation/evaporation should not lose or gain particles from the computational domain during the simulation. (For the evaporation process, number conservation may be applied to particles larger than the critical size.)

Numerous schemes have been developed to solve hyperbolic partial differential equations numerically. Highly accurate schemes such as the Galerkin finite element method and the Lax-Wendroff method have little numerical diffusion errors but exhibit numerical dispersion errors for steep concentration profiles. Conversely, first-order upwind differencing

schemes are free of numerical dispersion errors but have numerical diffusion errors. Some numerical schemes such as the flux-corrected transport (FCT) (24) and the Smolarkiewicz's repeated upwinding scheme (15) have been developed to eliminate numerical dispersion and reduce numerical diffusion. These schemes eliminate numerical dispersion by introducing diffusion terms and then subtracting the introduced diffusion contribution from the calculated quantities. Because all the schemes mentioned above calculate the transport of conserved quantities at fixed grids, i.e., using the Eulerian approach, numerical errors arising from the discrete approximation of spatial derivatives cannot be eliminated, only reduced.

In Lagrangian methods, grids and computational cells move with the conserved quantities along the characteristics thus eliminating numerical errors due to the discrete approximation of spatial derivatives. The most significant difficulty of applying a Lagrangian approach to aerosol condensation/evaporation is the distortion of the grids and cells that occurs as the different particle sizes grow or shrink at different rates. Some numerical

schemes use the idea of remapping the functional values from the distorted Lagrangian grid onto a regular grid by interpolation at each time step. During interpolation, however, new numerical errors are introduced. Tsang and Brock (17) used a finite element method incorporating characteristics (11, 21) for representing aerosol condensation and add an Eulerian-Lagrangian scheme (10) for evaporation for single component aerosols. Seigneur *et al.* (13) also used a finite element method with characteristics. However, even though its global accuracy is good this method does not conserve particle number concentration as a function of time.

Gelbard (6) combined the sectional approach with the Lagrangian concept of a movable grid and developed a moving sectional method for simulating aerosol condensation/evaporation of a single condensable species. In that method the boundaries of each particle size section move with time in accordance with the growth or shrinkage of particles at those boundaries. In that way numerical diffusion errors resulting from the need to apportion the particle size distribution over a set of fixed sections are avoided. Also, numerical dispersion errors resulting from the convection term are avoided since the convection term is automatically handled by solving the characteristic equations.

Single component aerosol dynamics involving either coagulation or condensation/evaporation can be simulated quite accurately with methods in the literature. Single component aerosol dynamics involving both coagulation and condensation/evaporation is considerably more difficult but can, nonetheless, be treated with available methods. The vast majority of paper in the literature address single-component problems and many are limited to condensation/evaporation alone. In this latter case, an approach involving moving elements or sections is the preferred one. For multicomponent aerosol dynamics the only technique that has been developed and applied is the sectional method, with fixed sections. The representation of condensation/evapo-

ration is of course prone to numerical diffusion just as in the single-component case.

There are several numerical schemes available to calculate multidimensional convection problems which have the same form as those of multicomponent aerosol condensation/evaporation (7, 12), but the schemes require significant computer memory and computing time. For an  $s$  component system, a finite difference method with  $n$  grids for each component needs, for example, an  $n^s$  grid point and a finite element method with  $n$  cells for each component requires an  $n^s \times n^s$  matrix system although the matrix is usually banded and/or sparse. Also one has to calculate  $s$  condensation rate equations at each grid point ( $s \times n^s$  calculations for a finite difference method). Furthermore, to calculate the spatial and temporal derivatives with a reasonable accuracy, rather time-consuming schemes should be used, also requiring large computing time and memory. To calculate multicomponent aerosol dynamics due to condensation/evaporation processes by conventional multidimensional convection schemes does not appear to be attractive. For this reason we adopt the sectional concept together with that of a Lagrangian grid.

Our goal in this work is to extend the concept of the moving sectional method to multicomponent aerosol condensation/evaporation processes. To demonstrate the performance of the method we obtain analytical solutions to model problems of multicomponent condensation/evaporation to which the numerical solutions may be compared. Accuracy and computational efficiency are further compared between the moving sectional method and the original, fixed grid sectional method.

Finally, limitations of the sectional method due to its inherent assumptions are discussed.

## 2. GENERAL DYNAMIC EQUATION FOR CONDENSATION/EVAPORATION

The general dynamic equation for multicomponent condensation/evaporation processes is (3)

$$\begin{aligned} & \frac{\partial n(m, M, t)}{\partial t} \\ & + \sum_{i=1}^{s-1} \frac{\partial}{\partial m_i} [I_i(m, M, t)n(m, M, t)] \\ & + \frac{\partial}{\partial M} [I(m, M, t)n(m, M, t)] = 0, \quad [1] \end{aligned}$$

where  $M$  denotes the total mass of a particle and  $m_i$  is the mass of the  $i$ th component in the particle  $m = (m_1, \dots, m_{s-1})$ , where  $s$  is the number of components.  $n(m, M, t)dmdM$  is the number of particles having total mass in the range  $[M, M + dM]$ , mass of component  $i$  in the range  $[m_i, m_i + dm_i]$ ,  $i = 1, \dots, s - 1$ , at the time  $t$ .  $I_i$  is the time rate change of mass concentration of the  $i$ th component and  $I$  is the time rate change of the total mass concentration. The initial condition is the size-composition distribution at time zero,

$$n(m, M, 0) = n_0(m, M). \quad [2]$$

The boundary condition is given by

$$n(m, M^*, t) = 0, \quad [3]$$

where  $M^*$  is the critical total mass below which no aerosol particles exist.

### 3. SECTIONAL METHOD

#### 3.1. Sectional Method Development

The essential idea of the sectional method is to divide the aerosol size spectrum into  $N$  sections and to assume:

1. In a section, the total particle mass size distribution is uniform.
2. All particles in a section have identical composition.

The total mass concentration of the  $l$ th section is defined as

$$\begin{aligned} Q_l(t) &= \int_{M_{l,l}}^{M_{l,u}} \int_0^M \cdots \int_0^M Mn(m, M, t) \\ &\quad \times dmdM, \quad [4] \end{aligned}$$

where  $M_{l,l}$  is the lower bound and  $M_{l,u}$  is the upper bound of total mass in the  $l$ th section.

By applying the first assumption, we get

$$\begin{aligned} \int_0^M \cdots \int_0^M Mn(m, M, t) dm \\ = q_l(t)f'(M), \quad [5] \end{aligned}$$

where  $q_l(t)$  is a constant in the section and  $f'(M) = df/dM$ , where  $f(M)$  is the size variable of interest. An example of  $f(M)$  is  $\ln(M/M_0)$ .

Combining Eqs. [4] and [5] gives

$$\begin{aligned} Q_l(t) &= \int_{M_{l,l}}^{M_{l,u}} q_l(t)f'(M)dM \\ &= q_l(t)[f(M_{l,u}) - f(M_{l,l})]. \quad [6] \end{aligned}$$

The total number concentration of particles in the  $l$ th section is given by

$$\begin{aligned} N_l(t) \\ = \int_{M_{l,l}}^{M_{l,u}} \int_0^M \cdots \int_0^M n(m, M, t)dmdM. \quad [7] \end{aligned}$$

If one chooses  $\ln M$  as the size variable, i.e.,  $f(M)$ , then  $N_l(t)$  is given by

$$N_l(t) = \frac{Q_l(t) \left[ \frac{1}{M_{l,l}} - \frac{1}{M_{l,u}} \right]}{\ln M_{l,u} - \ln M_{l,l}}. \quad [8]$$

By applying the second assumption,

$$\begin{aligned} \frac{m_i}{M} &= \text{constant} \quad M_{l,l} \leq m_i \leq M_{l,u} \\ i &= 1, \dots, s - 1. \quad [9] \end{aligned}$$

The mass concentration of the  $k$ th component in the  $l$ th section,  $Q_{l,k}(t)$ , is given by

$$\begin{aligned} Q_{l,k}(t) &= \int_{M_{l,l}}^{M_{l,u}} \int_0^M \cdots \int_0^M m_k n(m, M, t) \\ &\quad \times dmdM \\ &= \int_{M_{l,l}}^{M_{l,u}} M \int_0^M \cdots \int_0^M \left( \frac{m_k}{M} \right) \\ &\quad \times n(m, M, t)dmdM \end{aligned}$$

$$= \left( \frac{m_k}{M} \right) Q_l(t) \\ k = 1, \dots, s-1. \quad [10]$$

### 3.2. Moving Sectional Method

During the condensation/evaporation processes for a fixed volume, if the section boundaries move without loss or gain of particles in that section, the particle number concentration in a section must be constant with time. Thus  $N_l(t)$  of Eq. [8] is constant, or  $dN_l(t)/dt = 0$ ,  $l = 1, \dots, N$ . Thus if the temporal variation of the section boundaries and each component mass in a section are known, one can calculate the evolution of total mass concentration of aerosol and mass concentration of each component in a section. If section boundaries follow the characteristics of condensation/evaporation rates, particle number will be conserved.

In the moving sectional method all particles initially within a section remain there and the section boundaries move according to the particle growth equation, i.e., the characteristics. Therefore, all particles in a section cannot escape the section boundaries. The section boundaries obey the characteristic equations shown below,

$$\frac{dM_j}{dt} = I(m, M, t) \quad j = 1, \dots, 2N, \quad [11]$$

where the index  $j$  denotes the  $2N$  section boundaries.

The problem now is how to calculate component mass changes within each section as a result of the condensation/evaporation processes. Each component in a section also varies according to the condensation rate equation,

$$\frac{dm_i}{dt} = I_i(m, M, t) \quad i = 1, \dots, s-1. \quad [12]$$

Because of the second assumption of the sectional method, the ratio  $(m_i/M)$  is constant in a section; therefore it is immaterial which value of  $M$  is used to calculate  $(m_i/M)$ . Thus at each time step, temporal component mass

variations in a section are calculated by solving Eqs. [11] and [12] with a fixed value of  $M$  in the section, and one can then calculate the multicomponent condensation/evaporation processes from Eqs. [8] and [10].

One important advantage of this method is that the moving sectional method can simulate particles of the same total mass but different compositions. Since compositions of adjacent sections are discontinuous at the section boundary, condensation/evaporation rates of each section at the boundary are different. As a result, sections can in principle overlap each other or holes in the calculation domain may develop between sections. In the examples to be considered here, we have assumed linear total mass condensation/evaporation rates. In these cases no overlapping of sections or holes between sections develop, but with other growth rate equations, in the same total mass range, overlapping sections with different compositions could exist.

## 4. ANALYTICAL SOLUTIONS FOR MULTICOMPONENT AEROSOL CONDENSATION/EVAPORATION

In this section, we will derive new analytical solutions for multicomponent aerosol condensation/evaporation. These solutions will be used as a reference to evaluate the performance of the numerical scheme developed above.

Two problems of multicomponent aerosol condensation/evaporation will be considered. Both problems are characterized by linear condensation/evaporation rates. Although such rates describe the processes of volume reaction controlled gas-to-particle conversion (14), the main reason for their selection is to enable analytical solution of the GDE. The two problems differ in their initial aerosol size distributions; one problem has an exponential initial size distribution and the other has a log-normal initial size distribution.

Condensation/evaporation rate equations are taken to be linear in total mass and in each component,

$$I = \alpha M \quad [13]$$

$$I_i = \alpha_i m_i \quad i = 1, \dots, s-1. \quad [14]$$

Note that if the value of  $\alpha_i$  is negative, the  $i$ th component evaporates. A three-component system will be considered in the numerical examples to follow.

The derivation of the analytical solutions for mass concentration together with definitions of the notation are given in the Appendix.

For the exponential initial size distribution case, the total mass concentration of the  $l$ th section is

$$\begin{aligned} Q_l(t) &= \int_{M_{l,l}}^{M_{l,u}} \int_0^M \dots \int_0^M \frac{N_0}{M_0} M \\ &\times \exp \left[ -\frac{M}{M_0} e^{-\alpha t} - \alpha t \right] \\ &\times \prod_{i=1}^{s-1} \frac{1}{m_{i0}} \exp \left[ -\frac{m_i}{m_{i0}} e^{-\alpha_i t} - \alpha_i t \right] dm dM \\ &= \int_{M_{l,l}}^{M_{l,u}} \frac{N_0}{M_0} M \exp \left[ -\frac{M}{M_0} e^{-\alpha t} - \alpha t \right] \\ &\times \prod_{i=1}^{s-1} \left( 1 - \exp \left[ -\frac{M}{m_{i0}} e^{-\alpha_i t} \right] \right) dM. \quad [15] \end{aligned}$$

The mass concentration of  $j$ th component in the  $l$ th section is

$$\begin{aligned} Q_{l,j}(t) &= \int_{M_{l,l}}^{M_{l,u}} \frac{N_0}{M_0} \exp \left[ -\frac{M}{M_0} e^{-\alpha t} - \alpha t \right] \\ &\times \left[ \prod_{i=1, i \neq j}^{s-1} \left( 1 - \exp \left[ -\frac{M}{m_{i0}} e^{-\alpha_i t} \right] \right) \right] \\ &\times \left[ \left( 1 - \exp \left[ -\frac{M}{m_{j0}} e^{-\alpha_j t} \right] \right) m_{j0} e^{\alpha_j t} \right. \\ &\quad \left. - M \exp \left[ -\frac{M}{m_{j0}} e^{-\alpha_j t} \right] \right] dM. \quad [16] \end{aligned}$$

For the log-normal initial distribution problem, the total mass concentration of the  $l$ th section is

$$\begin{aligned} Q_l(t) &= \int_{M_{l,l}}^{M_{l,u}} \frac{N_0}{(2\pi)^{1/2} \ln \sigma} \\ &\times \exp \left[ -\frac{(\ln M - \ln \bar{M} - \alpha t)^2}{2 \ln^2 \sigma} \right] \\ &\times \prod_{i=1}^{s-1} \frac{1}{2} \left[ 1 \pm \operatorname{erf} \left( \frac{(\ln M - \ln \bar{m}_i - \alpha_i t)}{\sqrt{2} \ln \sigma_i} \right) \right] \\ &\times dM. \quad [17] \end{aligned}$$

The mass concentration of the  $j$ th component in the  $l$ th section is

$$\begin{aligned} Q_{l,j}(t) &= \int_{M_{l,l}}^{M_{l,u}} \frac{N_0}{(2\pi)^{1/2} M \ln \sigma} \\ &\times \exp \left[ -\frac{(\ln M - \ln \bar{M} - \alpha t)^2}{2 \ln^2 \sigma} \right] \\ &\times \prod_{i=1, i \neq j}^{s-1} \frac{1}{2} \left[ 1 \pm \operatorname{erf} \left( \frac{(\ln M - \ln \bar{m}_i - \alpha_i t)}{\sqrt{2} \ln \sigma_i} \right) \right] \\ &\times \frac{1}{2} \exp \left[ \ln \bar{m}_j + \alpha_j t + \frac{\ln^2 \sigma_j}{2} \right] \\ &\times \left[ 1 \pm \operatorname{erf} \left( \frac{(\ln M - \ln \bar{m}_j - \alpha_j t - \ln^2 \sigma_j)}{\sqrt{2} \ln \sigma_j} \right) \right] \\ &\times dM. \quad [18] \end{aligned}$$

By using any appropriate numerical integration method, such as Gaussian quadrature, one can evaluate  $Q_l(t)$  and  $Q_{l,j}(t)$ .

## 5. NUMERICAL SOLUTIONS OF MULTICOMPONENT AEROSOL CONDENSATION/EVAPORATION

For generality and ease of calculation, non-dimensionalization will be used henceforth. Also, because of the wide range of magnitudes of the independent variables, a logarithmic transformation similar to that used by Gelbard and Seinfeld (2) is used:

$$X = \ln \left( \frac{M}{M_0} \right) \quad [19]$$

$$x_i = \ln \left( \frac{m_i}{M_0} \right) \quad i = 1, \dots, s-1. \quad [20]$$

The parameters chosen for the numerical simulations for the exponential initial distribution are  $m_{10} = 0.3$ ,  $m_{20} = 0.2$ . For the log-normal initial distribution, the parameters employed are, for total number concentration, number median dimensionless mass of 3.0 and geometric standard deviation of 3.0. For components 1 and 2, number median dimensionless masses are 0.1 and 0.3, and geometric standard deviations are 5.0 and 5.0, respectively.

Two cases of condensation/evaporation rates were solved for the model problems. Case 1 is a pure condensation process with condensation rate coefficients chosen as  $\alpha = 1.0 \text{ s}^{-1}$ ,  $\alpha_1 = 0.9 \text{ s}^{-1}$ , and  $\alpha_2 = 1.1 \text{ s}^{-1}$ . The second case is one of mixed condensation/evaporation. The evaporation rate coefficient of the first component is chosen as  $\alpha_1 = -0.1 \text{ s}^{-1}$  and the condensation rates of total mass and component 2 are  $\alpha = 1.0 \text{ s}^{-1}$  and  $\alpha_2 = 1.7$

$\text{s}^{-1}$ . The dimensionless time  $\tau$  is defined as  $\tau = \alpha t$  for these linear cases.

Ten sections were used for both the moving and the fixed sectional methods. The dimensionless calculation domain in terms of  $M/M_0$  is  $[10^{-2}, 10^2]$  for the exponential initial distribution and  $[10^{-2}, 10^5]$  for the log-normal initial distribution.

The fixed sectional results were obtained according to the algorithm of Warren and Seinfeld (22). The average density of particles was  $10^6 \text{ g m}^{-3}$  and the reference total mass was that of a particle of size  $1 \mu\text{m}$ .

To assess the numerical errors due to the moving sectional method and to compare the results of the moving sectional method to those of the fixed sectional method, error sources from the time integration and the sectional method were carefully reduced or eliminated as much as possible. When linear condensation/evaporation rates are assumed, the log-

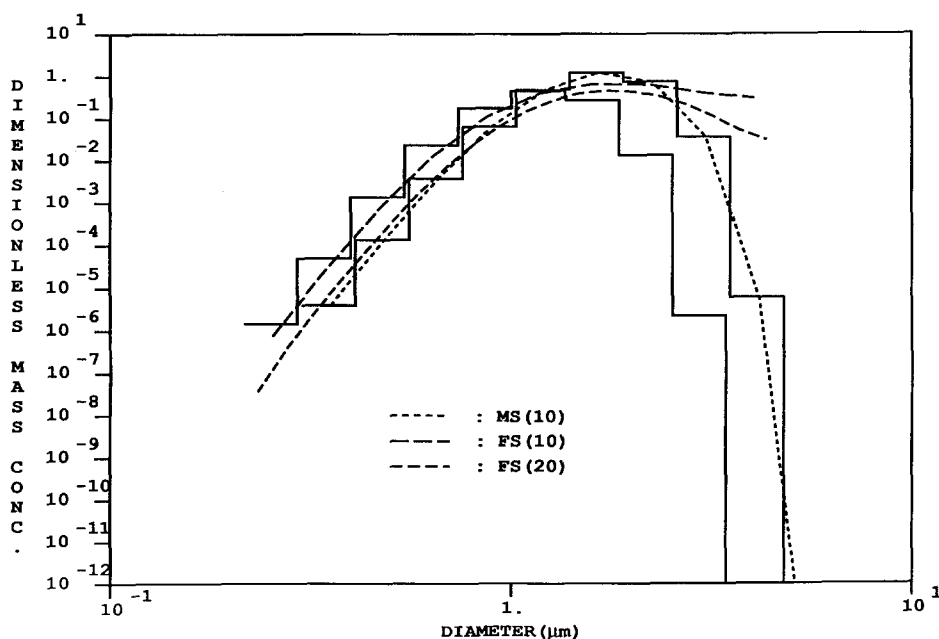


FIG. 1. Comparison of analytical solution (solid line; the one on the left is the initial size distribution) and sectional solutions of dimensionless total mass concentration for case 1 with exponential initial size distribution at dimensionless time 1. MS(10), moving sectional method with 10 sections; FS(10), fixed sectional method with 10 sections; FS(20), fixed sectional method with 20 sections.

arithmically transformed rate equations become zeroth-order ordinary differential equations. In such a case, the Euler method produces no numerical error due to time integration. Initial conditions calculated from the analytical expression were used for both analytical and numerical calculations to reduce numerical errors due to the assumptions of the sectional model.

Figure 1 shows a comparison between the analytical solution and the numerical solutions using sectional methods of total mass concentration for case 1 (all species condense) with exponential initial size distribution at dimensionless time 1. Excellent agreement between the moving sectional solution and the exact solution is obtained. The fixed sectional method with 10 sections gives numerical solutions that exhibit the effect of numerical diffusion errors. Increasing the number of sections by a factor of 2 increased somewhat the accuracy of numerical solution. Note the underpredictions at the peak and overpredictions

at the larger particle end of the spectrum that are characteristics of numerical diffusion errors. Figures 2 and 3 show the results for case 1 and the exponential initial size distribution for dimensionless mass concentrations of components 1 and 2, respectively. Again, the moving sectional method shows excellent results, and the fixed sectional method exhibits numerical diffusion errors.

Figures 4, 5, and 6 show the results for case 2 (component 1 evaporates, but others condense and the overall effect is condensation) with the exponential initial size distribution. As above, for the moving sectional method, good agreement with the exact solution is obtained although the numerical solutions underpredict at the smaller particle end of the spectrum. Doubling the number of sections did not appreciably increase the accuracy of the numerical solution using fixed sections.

To compare the results of the various approaches quantitatively, the median relative error,

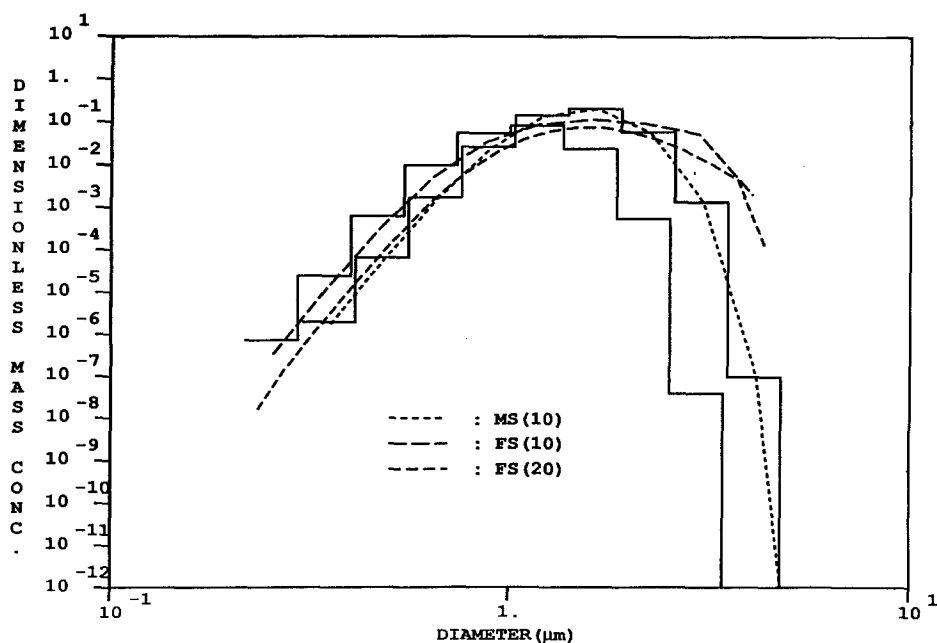


FIG. 2. Comparison of analytical solution (solid line; the one on the left is the initial size distribution) and sectional solutions of dimensionless mass concentration of component 1 for case 1 with exponential initial size distribution at dimensionless time 1. MS(10), moving sectional method with 10 sections; FS(10), fixed sectional method with 10 sections; FS(20), fixed sectional method with 20 sections.



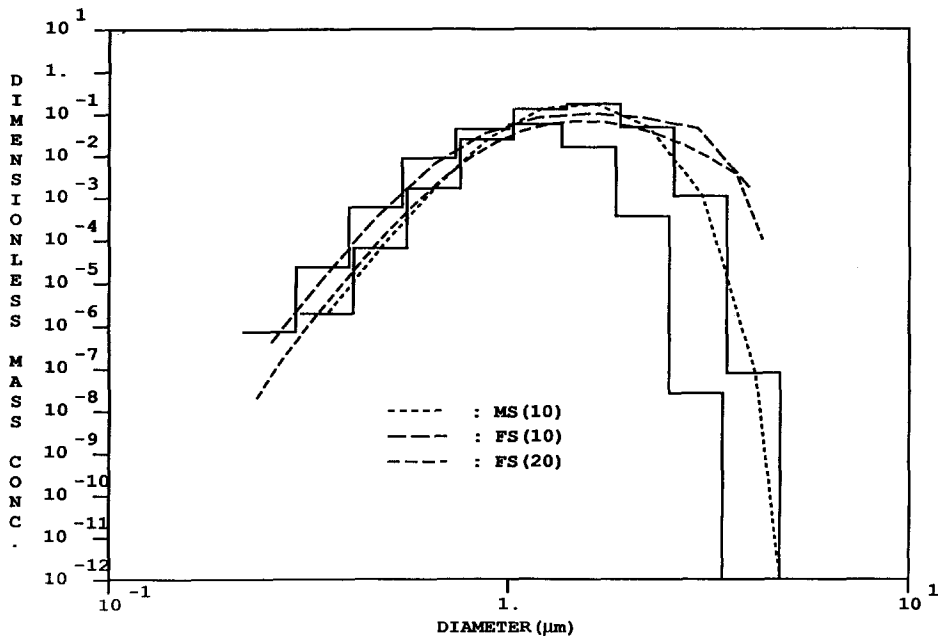


FIG. 3. Comparison of analytical solution (solid line; the one on the left is the initial size distribution) and sectional solutions of dimensionless mass concentration of component 2 for case 1 with exponential initial size distribution at dimensionless time 1. MS(10), moving sectional method with 10 sections; FS(10), fixed sectional method with 10 sections; FS(20), fixed sectional method with 20 sections.

median relative error

$$= \text{median} \left[ \frac{\text{abs}(Q_{i|\text{analytical}} - Q_{i|\text{numerical}})}{Q_{i|\text{analytical}}} \times 100 \right], \quad [21]$$

the value of relative error below which one-half of values of relative errors lies, was computed. Since the dimensionless mass concentration varies widely (more than an order of 10 in the computational domain), to use maximum absolute and therefore maximum relative errors among sections is not appropriate because a small shift of the numerical solution profile at the far right side of the domain results in a large error. Table II gives the median relative errors of the different methods for both cases.

In Table III, the ratio of computing times between the fixed and moving sectional methods is shown. The moving sectional method

requires slightly more computing time than the fixed sectional method for the same number of sections, but less computing time than the fixed sectional method with twice the number of sections.

Since the moving sectional method incorporates a number-conserving condition and uses condensation/evaporation rates as the characteristics of the section boundaries movement, it retains the advantages of both the fixed sectional method and the finite element method incorporating characteristics, being free from numerical dispersion error while conserving number concentration. This method also has the advantage that it can be developed to treat multicomponent aerosols and both condensation and evaporation.

The sectional method (irrespective of whether the sections are moving or fixed) is not without limitations. The first assumption inherent in the method that total mass size distribution is uniform in a section is the sim-

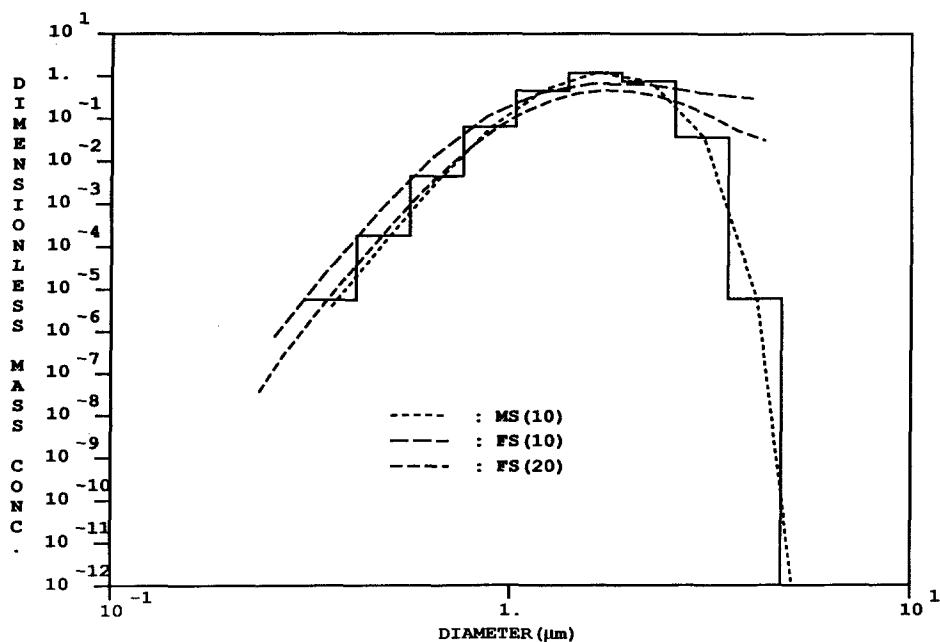


FIG. 4. Comparison of analytical solution (solid line) and sectional solutions of dimensionless total mass concentration for case 2 with exponential initial size distribution at dimensionless time 1. MS(10), moving sectional method with 10 sections; FS(10), fixed sectional method with 10 sections; FS(20), fixed sectional method with 20 sections.

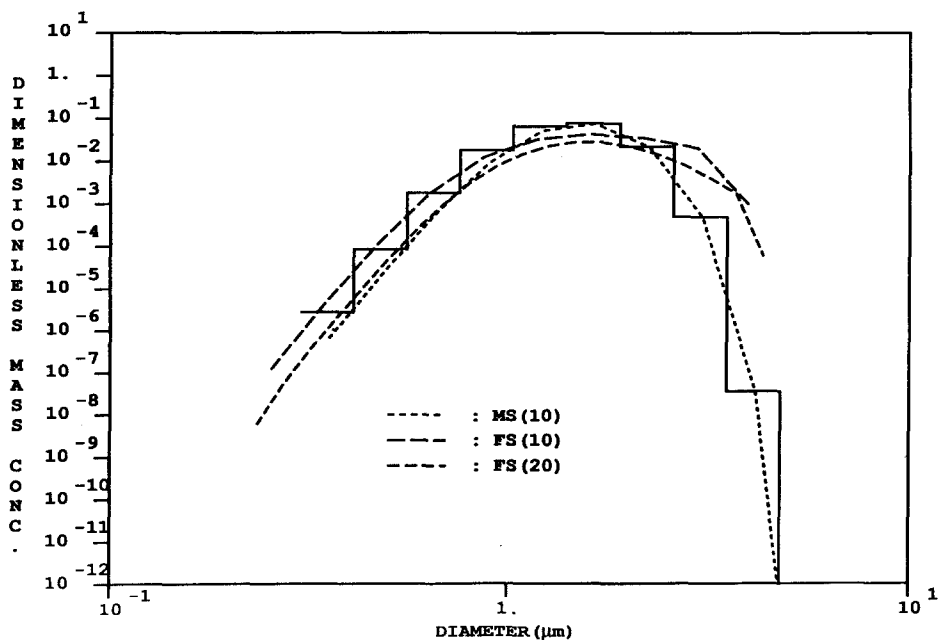


FIG. 5. Comparison of analytical solution (solid line) and sectional solutions of dimensionless mass concentration of component 1 for case 2 with exponential initial size distribution at dimensionless time 1. MS(10), moving sectional method with 10 sections; FS(10), fixed sectional method with 10 sections; FS(20), fixed sectional method with 20 sections.

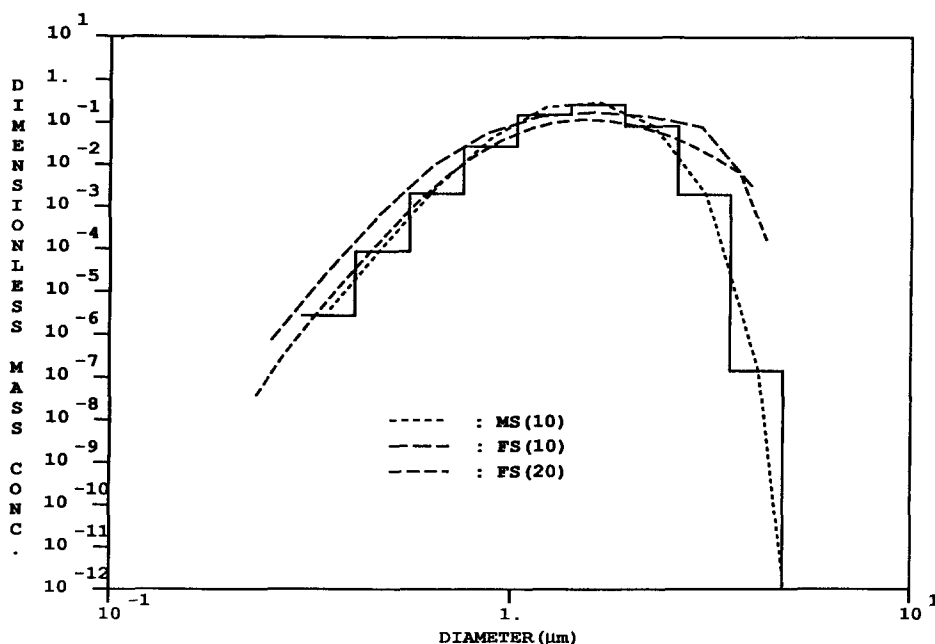


FIG. 6. Comparison of analytical solution (solid line) and sectional solutions of dimensionless mass concentration of component 2 for case 2 with exponential initial size distribution at dimensionless time 1. MS(10), moving sectional method with 10 sections; FS(10), fixed sectional method with 10 sections; FS(20), fixed sectional method with 20 sections.

plest form of approximation of the size distribution and introduces numerical diffusion errors. Of course, such errors are inevitable in a fixed grid approach, even with higher order functional forms of approximation. Thus the first assumption in the sectional method can

be viewed as a useful mathematical approximation.

The second assumption of the sectional method is that the compositions of each component are constant in a section; that is, particles of the same mass all have the same com-

TABLE II

Comparison of Median Relative Error, Eq. [21]<sup>a</sup> of Total Dimensionless Mass Concentration, at  $\tau = 1$

Case	Method	Exponential distribution	Log-normal distribution
Case 1 <sup>b</sup>	Moving sectional method	$0.214 \times 10^0$	$0.166 \times 10^0$
	Fixed sectional method	$0.640 \times 10^3$	$0.354 \times 10^4$
	Fixed sectional method (20 sections)	$0.305 \times 10^3$	$0.203 \times 10^4$
Case 2 <sup>c</sup>	Moving sectional method	$0.560 \times 10^0$	$0.195 \times 10^1$
	Fixed sectional method	$0.445 \times 10^3$	$0.425 \times 10^4$
	Fixed sectional method (20 sections)	$0.200 \times 10^3$	$0.215 \times 10^4$

<sup>a</sup> Median relative error = median  $[\text{abs}(Q_i|_{\text{analytical}} - Q_i|_{\text{numerical}})/Q_i|_{\text{analytical}} \times 100]$ .

<sup>b</sup> All components condense.

<sup>c</sup> Component one evaporates, but others condense.

TABLE III  
Comparison of Ratio of Computing Times at  $\tau = 1$

Case	Method	Exponential distribution	Log-normal distribution
Case 1 <sup>a</sup>	Moving sectional method	1	1
	Fixed sectional method	0.852	0.897
	Fixed sectional method (20 sections)	1.356	1.402
Case 2 <sup>b</sup>	Moving sectional method	1	1
	Fixed sectional method	0.916	0.923
	Fixed sectional method (20 sections)	1.583	1.769

<sup>a</sup> All components condense.

<sup>b</sup> One component evaporates, but others condense.

position. This approximation introduces not only numerical errors but also may not be physically realistic. The moving sectional method can produce particles of the same size but different composition since sections can overlap each other and/or holes in the calculation domain may develop between sections. This property of the moving sectional method alleviates the limitations arising from the second assumption of the method.

As a final point, while moving elements offer many advantages for simulating condensation/evaporation, they are not desirable for coagulation. It is therefore unlikely that a multicomponent coagulation and condensation/evaporation simulation method will involve moving sections. If one seeks to reduce potential numerical diffusion and at the same time to relax the assumption that all particles of the same size have the same composition, it may be necessary to attack the multicomponent GDE as a true multivariable equation.

## 6. CONCLUSION

The moving sectional method has been extended to multicomponent aerosol condensation/evaporation processes with multiple condensable species using a Lagrangian approach and a mass conservation condition. This method accurately simulates the model multicomponent condensation/evaporation

processes for which analytical solutions are obtained. Limitations and applicability of the sectional method are discussed.

## APPENDIX

### ANALYTICAL SOLUTION FOR MULTICOMPONENT AEROSOL CONDENSATION/EVAPORATION

The general multicomponent condensation equation is given by Eq. [1]. The analytical solution of Eq. [1] can be expressed as

$$n(m, M, t) = \exp\left[-\sum_{i=1}^{s-1} \alpha_i t - \alpha t\right] \times n_0(m_1 e^{-\alpha_1 t}, \dots, m_{s-1} e^{-\alpha_{s-1} t}, M e^{-\alpha t}). \quad [\text{A1}]$$

The exponential initial distribution is given by

$$n_0(m, M) = \frac{N_0}{M_0} \exp\left[-\frac{M}{M_0}\right] \prod_{i=1}^{s-1} \frac{1}{m_{i0}} \exp\left[-\frac{m_i}{m_{i0}}\right] \quad [\text{A2}]$$

and the analytical solution for the exponential initial distribution is

$$n(m, M, t) = \frac{N_0}{M_0} \exp\left[-\frac{M}{M_0} e^{-\alpha t} - \alpha t\right] \times \prod_{i=1}^{s-1} \frac{1}{m_{i0}} \exp\left[-\frac{m_i}{m_{i0}} e^{-\alpha_i t} - \alpha_i t\right]. \quad [\text{A3}]$$

Then the total mass concentration of the  $l$ th section is

$$Q_l(t) = \int_{M_{l1}}^{M_{l2}} \int_0^M \dots \int_0^M \frac{N_0}{M_0} M \times \exp\left[-\frac{M}{M_0} e^{-\alpha t} - \alpha t\right] \times \prod_{i=1}^{s-1} \frac{1}{m_{i0}} \exp\left[-\frac{m_i}{m_{i0}} e^{-\alpha_i t} - \alpha_i t\right] dm dM$$

$$= \int_{M_{li}}^{M_{lu}} \frac{N_0}{M_0} M \exp \left[ -\frac{M}{M_0} e^{-\alpha t} - \alpha t \right] \\ \times \prod_{i=1}^{s-1} \left( 1 - \exp \left[ -\frac{M}{m_{i0}} e^{-\alpha_i t} \right] \right) dM. \quad [\text{A4}]$$

The mass concentration of  $j$ th component in the  $l$ th section is

$$Q_{l,j}(t) = \int_{M_{li}}^{M_{lu}} \frac{N_0}{M_0} \exp \left[ -\frac{M}{M_0} e^{-\alpha t} - \alpha t \right] \\ \times \left[ \int_0^M \cdots \int_0^M \prod_{i=1}^{s-1} \frac{m_j}{m_{i0}} \right. \\ \left. \times \exp \left[ -\frac{m_i}{m_{i0}} e^{-\alpha_i t} - \alpha_i t \right] dm \right] dM. \quad [\text{A5}]$$

If  $i \neq j$ , the integration is the same as that of Eq. [A4]. If  $i = j$ , using integration by parts,

$$\int_0^M \frac{m_j}{m_{j0}} \exp \left[ -\frac{m_j}{m_{j0}} e^{-\alpha_j t} - \alpha_j t \right] dm_j \\ = - \left[ m_j \exp \left[ -\frac{m_j}{m_{j0}} e^{-\alpha_j t} \right] \right]_0^M \\ + \int_0^M \exp \left[ -\frac{m_j}{m_{j0}} e^{-\alpha_j t} \right] dm_j \\ = -M \exp \left[ -\frac{M}{m_{j0}} e^{-\alpha_j t} \right] \\ + m_{j0} e^{\alpha_j t} \left( 1 - \exp \left[ -\frac{M}{m_{j0}} e^{-\alpha_j t} \right] \right). \quad [\text{A6}]$$

Therefore,

$$Q_{l,j}(t) = \int_{M_{li}}^{M_{lu}} \frac{N_0}{M_0} \exp \left[ -\frac{M}{M_0} e^{-\alpha t} - \alpha t \right] \\ \times \left[ \prod_{i=1, i \neq j}^{s-1} \left( 1 - \exp \left[ -\frac{M}{m_{i0}} e^{-\alpha_i t} \right] \right) \right] \\ \times \left[ \left( 1 - \exp \left[ -\frac{M}{m_{j0}} e^{-\alpha_j t} \right] \right) m_{j0} e^{\alpha_j t} \right. \\ \left. - M \exp \left[ -\frac{M}{m_{j0}} e^{-\alpha_j t} \right] \right] dM. \quad [\text{A7}]$$

The log-normal initial distribution is given by

$$n_0(m, M) \\ = \frac{N_0}{(2\pi)^{1/2} M \ln \sigma} \exp \left[ -\frac{(\ln M - \ln \bar{M})^2}{2 \ln^2 \sigma} \right] \\ \times \prod_{i=1}^{s-1} \frac{1}{(2\pi)^{1/2} m_i \ln \sigma_i} \\ \times \exp \left[ -\frac{(\ln m_i - \ln \bar{m}_i)^2}{2 \ln^2 \sigma_i} \right], \quad [\text{A8}]$$

where  $\bar{M}$  and  $\bar{m}_i$  are number median masses and  $\sigma$  and  $\sigma_i$  are geometric standard deviations.

For the log-normal initial distribution problem, the analytical solution is

$$n(m, M, t) = \frac{N_0}{(2\pi)^{1/2} M \ln \sigma} \\ \times \exp \left[ -\frac{(\ln M - \ln \bar{M} - \alpha t)^2}{2 \ln^2 \sigma} \right] \\ \times \prod_{i=1}^{s-1} \frac{1}{(2\pi)^{1/2} m_i \ln \sigma_i} \\ \times \exp \left[ -\frac{(\ln m_i - \ln \bar{m}_i - \alpha_i t)^2}{2 \ln^2 \sigma_i} \right]. \quad [\text{A9}]$$

Then the total mass concentration of the  $l$ th section is

$$Q_l(t) = \int_{M_{li}}^{M_{lu}} \frac{N_0 M}{(2\pi)^{1/2} M \ln \sigma} \\ \times \exp \left[ -\frac{(\ln M - \ln \bar{M} - \alpha t)^2}{2 \ln^2 \sigma} \right] \\ \times \left[ \int_0^M \cdots \int_0^M \prod_{i=1}^{s-1} \frac{1}{(2\pi)^{1/2} m_i \ln \sigma_i} \right. \\ \left. \times \exp \left[ -\frac{(\ln m_i - \ln \bar{m}_i - \alpha_i t)^2}{2 \ln^2 \sigma_i} \right] dm \right] dM. \quad [\text{A10}]$$

By changing variables,  $u_i = \ln m_i$ , and using the definition of the error function,

$$\begin{aligned}
& \int_0^M \cdots \int_0^M \prod_{i=1}^{s-1} \frac{1}{(2\pi)^{1/2} m_i \ln \sigma_i} \\
& \quad \times \exp \left[ -\frac{(\ln m_i - \ln \bar{m}_i - \alpha_i t)^2}{2 \ln^2 \sigma_i} \right] dm \\
& = \int_{-\infty}^{\ln M} \cdots \int_{-\infty}^{\ln M} \prod_{i=1}^{s-1} \frac{1}{(2\pi)^{1/2} \ln \sigma_i} \\
& \quad \times \exp \left[ -\frac{(u_i - \ln \bar{m}_i - \alpha_i t)^2}{2 \ln^2 \sigma_i} \right] du \\
& = \prod_{i=1}^{s-1} \frac{1}{2} \left[ 1 \pm \operatorname{erf} \left( \frac{(\ln M - \ln \bar{m}_i - \alpha_i t)}{\sqrt{2} \ln \sigma_i} \right) \right], \\
& \hspace{15em} [\text{A11}]
\end{aligned}$$

where the plus sign is used if  $\ln M \geq 0$ , and the minus sign is used if  $\ln M < 0$ . The error function is defined as

$$\operatorname{erf}(z) = \frac{2}{\sqrt{\pi}} \int_0^z \exp[-t^2] dt. \quad [\text{A12}]$$

So  $Q_l(t)$  is

$$\begin{aligned}
Q_l(t) &= \int_{M_{li}}^{M_{lu}} \frac{N_0}{(2\pi)^{1/2} \ln \sigma} \\
& \quad \times \exp \left[ -\frac{(\ln M - \ln \bar{M} - \alpha t)^2}{2 \ln^2 \sigma} \right] \\
& \quad \times \prod_{i=1}^{s-1} \frac{1}{2} \left[ 1 \pm \operatorname{erf} \left( \frac{(\ln M - \ln \bar{m}_i - \alpha_i t)}{\sqrt{2} \ln \sigma_i} \right) \right] \\
& \quad \times dM. \quad [\text{A13}]
\end{aligned}$$

The mass concentration of the  $j$ th component in the  $l$ th section is

$$\begin{aligned}
Q_{l,j}(t) &= \int_{M_{li}}^{M_{lu}} \frac{N_0}{(2\pi)^{1/2} M \ln \sigma} \\
& \quad \times \exp \left[ -\frac{(\ln M - \ln \bar{M} - \alpha t)^2}{2 \ln^2 \sigma} \right] \\
& \quad \times \left[ \int_0^M \cdots \int_0^M \prod_{i=1}^{s-1} \frac{m_j}{(2\pi)^{1/2} m_i \ln \sigma_i} \right. \\
& \quad \times \exp \left[ -\frac{(\ln m_i - \ln \bar{m}_i - \alpha_i t)^2}{2 \ln^2 \sigma_i} \right] dm \Big] dM. \\
& \hspace{15em} [\text{A14}]
\end{aligned}$$

If  $i \neq j$ , the integration is the same as that of Eq. [A11]. If  $i = j$ , expanding the exponential term and completing the square,

$$\begin{aligned}
& \int_0^M \frac{m_j}{(2\pi)^{1/2} m_j \ln \sigma_j} \\
& \quad \times \exp \left[ -\frac{(\ln m_j - \ln \bar{m}_j - \alpha_j t)^2}{2 \ln^2 \sigma_j} \right] dm_j \\
& = \int_{-\infty}^{\ln M} \frac{e^{u_j}}{(2\pi)^{1/2} \ln \sigma_j} \\
& \quad \times \exp \left[ -\frac{(u_j - \ln \bar{m}_j - \alpha_j t)^2}{2 \ln^2 \sigma_j} \right] du_j \\
& = \int_{-\infty}^{\ln M} \frac{1}{(2\pi)^{1/2} \ln \sigma_j} \\
& \quad \times \exp \left[ \ln \bar{m}_j + \alpha_j t + \frac{\ln^2 \sigma_j}{2} \right] \\
& \quad \times \exp \left[ -\frac{(u_j - (\ln \bar{m}_j + \alpha_j t + \ln^2 \sigma_j))^2}{2 \ln^2 \sigma_j} \right] du_j \\
& = \frac{1}{2} \exp \left[ \ln \bar{m}_j + \alpha_j t + \frac{\ln^2 \sigma_j}{2} \right] \\
& \quad \times \left[ 1 \pm \operatorname{erf} \left( \frac{(\ln M - \ln \bar{m}_j - \alpha_j t - \ln^2 \sigma_j)}{\sqrt{2} \ln \sigma_j} \right) \right], \\
& \hspace{15em} [\text{A15}]
\end{aligned}$$

where the plus sign is used if  $\ln M \geq 0$ , and the minus sign is used if  $\ln M < 0$ .

Thus  $Q_{l,j}(t)$  is,

$$\begin{aligned}
Q_{l,j}(t) &= \int_{M_{li}}^{M_{lu}} \frac{N_0}{(2\pi)^{1/2} M \ln \sigma} \\
& \quad \times \exp \left[ -\frac{(\ln M - \ln \bar{M} - \alpha t)^2}{2 \ln^2 \sigma} \right] \\
& \quad \times \prod_{i=1, i \neq j}^{s-1} \frac{1}{2} \left[ 1 \pm \operatorname{erf} \left( \frac{(\ln M - \ln \bar{m}_i - \alpha_i t)}{\sqrt{2} \ln \sigma_i} \right) \right] \\
& \quad \times \frac{1}{2} \exp \left[ \ln \bar{m}_j + \alpha_j t + \frac{\ln^2 \sigma_j}{2} \right] \\
& \quad \times \left[ 1 \pm \operatorname{erf} \left( \frac{(\ln M - \ln \bar{m}_j - \alpha_j t - \ln^2 \sigma_j)}{\sqrt{2} \ln \sigma_j} \right) \right] \\
& \quad \times dM. \quad [\text{A16}]
\end{aligned}$$

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