



COMPUTER SCIENCE  
TECHNICAL REPORT

A Framework for the Numerical  
Treatment of Aerosol Dynamics

Adrian Sandu and Christian Borden

CSTR-01-03  
September 2001

***MichiganTech***

Michigan Technological University, Houghton, MI 49931

# A Framework for the Numerical Treatment of Aerosol Dynamics

September 23, 2001

Adrian Sandu<sup>a</sup> and Christian Borden

Department of Computer Science, Michigan Technological University, 1400  
Townsend Drive, Houghton, MI 49931. E-mail: {asandu, ctborden}@mtu.edu.

---

<sup>a</sup>Corresponding author.

## Abstract

---

This paper presents a general framework for the discretization of particle dynamics equation by projection methods, which include Galerkin and collocation techniques. Based on this framework a discretization over piecewise polynomial spaces is discussed. Numerical examples are given using both linear and logarithmic coordinates; the results show that this discretization approach is able to accurately solve aerosol size distribution using a relatively small number of “size bins”.

**Keywords:** Aerosol dynamics, projection methods, piecewise polynomials.

---

## 1 Introduction

As our understanding expands, new processes are incorporated into air quality computer models. One example is the particulate matter (aerosol) processes, the importance of which is now widely recognized. Aerosols are now a priority focus area in environmental science due to the leading role they play as a cause of adverse human health, and their ability to scatter and absorb incoming solar radiation and thus modify warming due to greenhouse gases and reduce visibility. To accurately study the effects of aerosols it is necessary to resolve aerosol number and mass distributions as a function of chemical composition and size.

In this paper we develop a framework for solving the aerosol dynamics equation, which determines the size distribution of atmospheric particles. Approximations of the size distribution are considered in a suitable finite dimensional space. The discrete equation is obtained by projecting the dynamics equation onto the discrete space (using a Galerkin or a collocation approach). This approach leads to a bilinear system of coupled ordinary differential equations, which can be solved by a time-stepping method of choice. For simplicity the framework is developed for number densities of single-component particles, but it can be directly extended to mass or volume densities and multiple component particles.

To illustrate the general approach, we consider discretizations over piecewise-polynomial spaces. A linearly-implicit second order time-stepping method is proposed for the time integration. Numerical examples show that good accuracy is obtained with a small number of grid points.

The paper is organized as follows. Section 2 presents the particle dynamics equations and Section 3 surveys previous efforts to solve these equations numerically. The discretization framework is introduced in Section 4. Numerical results are presented in Section 6 and Section 7 draws conclusions and pinpoints future work.

## 2 The continuous particle dynamics equation

In this paper the continuous particle size distributions are considered functions of particle volume ( $v$ ) and time ( $t$ ). For simplicity we consider single component particles, but the discretization techniques can be directly generalized to multiple components.

The size distribution function (number density) of a family of particles will be denoted by  $n(v, t)$ ; the number of particles per unit volume of air with the volume between  $v$  and  $v + dv$  is  $n(v, t)dv$ .

Similar formulations can be given in terms of volume, surface, or mass densities [16]. However, recovering mass from a volume formulation is difficult in practice, as the densities are only approximatively known and are a function of composition and size.

The aerosol population undergoes a series of physical and chemical transformations. *Growth* processes include condensation, evaporation, deposition and sublimation (of gases to/from the particle surface). The growth of each component's volume takes place at a rate that depends on the particle's dimension and composition,  $dv/dt = I_v(v, t)$ . *Coagulation* forms new particles of volume  $v + w$  from the collision of two smaller particles of volumes  $v$  and  $w$ ; the collision rate  $\beta_{v,w}n(v)n(w)$  is proportional to the number of available small particles. *Nucleation* of gases creates small particles. *Emissions* increase the number of particles of a specific composition and size, while *deposition* processes remove particles from the atmosphere. In addition, the constituents interact chemically inside each particle, changing the chemical composition (but not the number) of particles.

Under the above physical transformations the number density changes according to [4]

$$\begin{aligned} \frac{\partial n(v, t)}{\partial t} &= -\partial [I_v(v) n(v, t)] / \partial v \\ &+ \frac{1}{2} \int_0^v \beta_{v-w, w} n(v-w, t) n(w, t) dw - n(v, t) \int_0^\infty \beta_{v, w} n(w, t) dw \\ &+ S(v, t), \\ n(v, 0) &= n^0(v), \quad n(0, t) = 0. \end{aligned} \tag{1}$$

The different terms in equation (1) describe, in order, the modification in the number of particles due to growth, creation of particles of volume  $v$  by coagulation, loss of volume  $v$  particles due to coagulation, increase in particle number due to nucleation, emissions and depositions (sources and sinks). Each of the terms will be explained in detail below. The equation is subject to a specified initial condition  $n^0(v)$ , and the boundary condition of no zero volume particles.

## 3 Previous work

Three major approaches are used to represent the size distribution of aerosols: continuous, discrete and parameterized. In this paper we focus on continuous models (i.e. continuous size distributions and the general dynamic equations in continuous form).

For computational purposes one needs to use finite-dimensional approximations of the continuous size distributions. In the *sectional approach* the size domain  $v \in [0, \infty]$  is divided into size bins  $v \in [V_i^{\text{low}}, V_i^{\text{high}}]$ . In each size bin  $i$  there are  $n_i$  particles per unit volume, all of them having the same mean volume  $V_i$ . Variations of this approach include the *full-moving* structure, the *quasi-stationary approach*, as well as the *moving-center* structure [9].

A nice survey of several popular numerical methods for particle dynamics equations is given in Zhang et al. [22].

The standard discrete version of the coagulation equation uses a monomer size distribution (the volume of the particles in bin each  $i$  is a multiple of the smallest volume,  $V_i = i V_1$ ,  $i = 1, 2, \dots$ ). The semi-implicit scheme to solve the discrete coagulation equation [20] is discussed by Jacobson in [9, Section 16]. The differential equation is discretized in time using backward Euler formula, and the quadratic terms  $n_{j-\ell}(t)n_\ell(t)$  are replaced by the "linearized" version  $n_{j-\ell}(t)n_\ell(t - \Delta t)$ , where  $\Delta t$  is the numerical time step size. The scheme can be adapted to general size distributions, and admits a volume-conserving formulation. A combination of cubic splines (coagulation) and moving finite element techniques (growth part) was used by Tsang and Hippe [21]. Meng, Dabdub and Seinfeld [14] present a size-resolved and chemically-resolved

model for aerosol dynamics in a mass density formulation. Gelbard and Seinfeld [4, 5, 6] solve the coupled dynamic equations using orthogonal collocation over finite elements. Lushnikov [13] uses generating functions to solve analytically the coagulation equation for particles consisting of monomers of two kinds, under the assumptions of a constant coagulation kernel  $\beta$  and particular initial distributions.

The integro-differential coagulation equation is difficult to solve accurately, due to the fact that the limit of integration of the first term depends on the variable  $v$  and the integrands are quadratic (the first term is of nonlinear Volterra type in the terminology of integral equations). The algorithms proposed in the literature for the coagulation equation include semi-implicit solutions, finite element method, orthogonal collocation over finite elements, J-space transformations, analytical solutions, etc.

The growth equation in number densities has the form of an advection equation, with the “flow speed” equal to the time derivative of the volume [17, Section 12]. A nice survey of several popular numerical methods for the growth equations is given in Zhang et. al. [22]. Different solution of the growth equations were proposed in [2, 8, 11, 12].

Many models include different processes successively, using a time splitting scheme. This enables the use of numerical methods tuned to each subprocess but introduces hard-to-quantify splitting errors. Simultaneous solutions of all dynamic processes are given in [4, 5, 6, 7, 14, 16, 21].

## 4 A general framework for discretization

We solve equation (1) by a semi-discretization in particle size ( $v$ ), followed by a time integration of the resulting system of ordinary differential equations. The semi-discretization in size is done by projecting the solution on a finite-dimensional subspace  $\text{span}\{\phi_1(v), \dots, \phi_s(v)\}$ ; this generalizes the sectional approach. The dynamic equation is imposed to hold exactly in a certain subspace (Galerkin approach) or at a certain set of nodes (collocation approach).

### 4.1 Discretization of the particle size distribution

The continuous number distribution is given a finite-dimensional approximation. Let  $\{\phi_i\}_{1 \leq i \leq s}$  be a set of continuous basis functions; then

$$n(v, t) = \sum_{i=1}^s n_i(t) \phi_i(v) , \quad \phi_i(x) = \text{basis function} . \quad (2)$$

The set of time-dependent expansion coefficients

$$n(t) = [n_1(t), \dots, n_s(t)]^T , \quad (3)$$

will be determined from the dynamics equation.

In logarithmic scaled coordinates the basis function arguments are changed accordingly,  $\phi_i(\log[v/V_m])$ .

The representation (2) places the problem in the general framework of *projection methods* [1]. For example  $\phi_i$  can be piecewise polynomials or can be orthogonal polynomials. The result is a continuous distribution  $n(v, t)$ . Higher order approximations can be obtained by increasing the order of the basis functions without changing the number of bins  $s$ .

We note in passing that the full-stationary sectional approach can be formally cast into the form (2) by using Dirac\* basis functions  $\phi_i(v) = \delta(v - V_i)$ . For this reason we extend the sectional interpretation and call  $\text{span}\{\phi_i\}$  the *size bin*  $i$ , and refer to  $n_i$  as the number of particles in *bin*  $i$ .

### 4.2 Coagulation

The theoretical coagulation equation for single-component particles is [9, Section 16]

$$\frac{\partial n(v, t)}{\partial t} = \frac{1}{2} \int_0^v \beta_{v-w, w} n(v-w, t) n(w, t) dw - n(v, t) \int_0^\infty \beta_{v, w} n(w, t) dw . \quad (4)$$

---

\*Recall that  $\delta(x) = 0$  for  $x \neq 0$ ,  $\delta(0) = \infty$ , and  $\int_{V_i-\epsilon}^{V_i+\epsilon} f(x) \delta(x - V_i) dx = f(V_i)$ .

**The Galerkin approach.** To obtain a discrete form of the coagulation equation one inserts (2) into (4):

$$\begin{aligned} \sum_{i=1}^s n'_i(t) \phi_i(v) &= \frac{1}{2} \sum_{k=1}^s \sum_{m=1}^s n_k(t) n_m(t) \int_0^v \beta_{v-w,w} \phi_k(v-w) \phi_m(w) dw \\ &\quad - \sum_{k=1}^s \sum_{m=1}^s n_k(t) n_m(t) \int_0^\infty \beta_{v,w} \phi_k(v) \phi_m(w) dw . \end{aligned}$$

The resulting equation is multiplied by the test function  $\xi_j(v)$  ( $j = 1 \dots s$ ) and integrated from  $v = 0$  to  $\infty$  to obtain a system of  $s$  ordinary differential equations

$$\begin{aligned} \sum_{i=1}^s n'_i(t) \int_0^\infty \phi_i(v) \xi_j(v) dv &= \frac{1}{2} \sum_{k=1}^s \sum_{m=1}^s n_k(t) n_m(t) \int_0^\infty \left( \int_0^v \beta_{v-w,w} \phi_k(v-w) \phi_m(w) dw \right) \xi_j(v) dv \\ &\quad - \sum_{k=1}^s \sum_{m=1}^s n_k(t) n_m(t) \int_0^\infty \left( \int_0^\infty \beta_{v,w} \phi_m(w) dw \right) \phi_k(v) \xi_j(v) dv . \end{aligned} \quad (5)$$

We build the following matrices of integral coefficients

$$\begin{aligned} A &= \left[ \int_0^\infty \phi_j(v) \xi_i(v) dv \right]_{1 \leq i, j \leq s} \\ B^j &= \left[ (1/2) \int_0^\infty \left( \int_0^v \beta_{v-w,w} \phi_k(v-w) \phi_m(w) dw \right) \xi_j(v) dv \right]_{1 \leq k, m \leq s} , \quad 1 \leq j \leq s , \\ C^j &= \left[ \int_0^\infty \left( \int_0^\infty \beta_{v,w} \phi_m(w) dw \right) \phi_k(v) \xi_j(v) dv \right]_{1 \leq k, m \leq s} , \quad 1 \leq j \leq s . \end{aligned} \quad (6)$$

If  $n(t)$  is the vector of number concentrations (3), the equation (5) becomes

$$A n'(t) = \begin{bmatrix} n^T(t) (B^1 - C^1) n(t) \\ \vdots \\ n^T(t) (B^s - C^s) n(t) \end{bmatrix} . \quad (7)$$

One can regard  $B$  and  $C$  as 3-tensors, in which case the semi-discrete coagulation equation reads

$$A n'(t) = [(B - C) \times n(t)] \cdot n(t) . \quad (8)$$

In the pure *Galerkin* approach [1]  $\{\phi_i(v)\} = \{\xi_i(v)\}$  are (piecewise) continuous basis functions. The equation (7) is relatively expensive to solve, since one has to evaluate a large number of double integrals for building the tensors  $B$  and  $C$ . If  $\beta(v, w)$  does not change in time one can compute the tensors once and reuse them throughout integration, thus making the whole process computationally feasible.

**The collocation approach.** In the *collocation* approach  $\{\phi_i(v)\}$  are continuous basis functions, but the test functions are deltas,  $\{\xi_i(v) = \delta(v - V_i^c)\}$ , with  $V_i^c$  the collocation points. The resulting equation is also of form (7), but the integral coefficients to be computed simplify to

$$\begin{aligned} A &= [\phi_j(V_i^c)]_{1 \leq i, j \leq s} \\ B^j &= \left[ (1/2) \int_0^v \beta_{V_j^c-w,w} \phi_k(V_j^c-w) \phi_m(w) dw \right]_{1 \leq k, m \leq s} , \quad 1 \leq j \leq s , \\ C^j &= \left[ \phi_k(V_j^c) \int_0^\infty \beta_{V_j^c,w} \phi_m(w) dw \right]_{1 \leq k, m \leq s} , \quad 1 \leq j \leq s . \end{aligned} \quad (9)$$

This approach is computationally less expensive since all coefficients involve only simple integrals.

### 4.3 Growth

Growth processes include condensation, evaporation, deposition and sublimation (of gases to/from the particle surface). Consider again the case of single component particles. The growth equation in number densities

$$\frac{\partial n(v, t)}{\partial t} = -\frac{\partial}{\partial v} [I(v) n(v, t)] , \quad I(v) = \frac{dv(t)}{dt} , \quad n(0, t) = 0 , \quad n(v, 0) = n^0(v) , \quad (10)$$

has the form of an advection equation, with the “flow speed” provided by the time derivative of the volume. This equation is to be solved subject to an initial distribution  $n^0(v)$  and the boundary condition of no zero-sized particles [17, Section 12],

A similar derivation process (as presented for coagulation) leads to the semi-discrete formulation of the growth equations

$$A n'(t) = G n(t) , \quad (11)$$

where

$$A = \left[ \int_0^\infty \phi_j(v) \xi_i(v) dv \right]_{1 \leq i, j \leq s} , \quad G = \left[ \int_0^\infty I(v) \phi_j(v) \xi_i'(v) dv \right]_{1 \leq i, j \leq s} . \quad (12)$$

For  $G$  we have used one integration by parts and homogeneous boundary conditions at  $v = 0$  and  $v = \infty$ . For the Galerkin approach use  $\xi_i = \phi_i$ . For the collocation approach one obtains

$$A = [\phi_j(V_i^c)]_{1 \leq i, j \leq s} , \quad G = [-[I\phi_j]'(V_i^c)]_{1 \leq i, j \leq s} . \quad (13)$$

### 4.4 Sources and sinks

Sources and sinks (i.e. emissions, nucleation and deposition processes) have a simple mathematical formulation,

$$\frac{\partial n(v, t)}{\partial t} = S(v, t) - R(v, t) n(v, t) . \quad (14)$$

The simplicity comes from the fact that  $S$  and  $R$  terms are not coupled across different volumes; finite-dimensional approximations of these terms can be given:  $S(t) = \sum_{i=1}^s S_i(t) \phi_i(v)$  and  $R(t) = \sum_{i=1}^s R_i(t) \phi_i(v)$ . The discrete evolution equation can be written as

$$n'(t) = S(t) - R(t)n(t) \quad \text{where} \quad R(t) = \text{diag}(R_1(t), \dots, R_s(t)) . \quad (15)$$

### 4.5 Simultaneous discretization of the dynamic equations

Of particular interest is the coupled solution of coagulation, growth, nucleation, emissions and deposition. The coupled approach will, for example, better capture the competition between nucleation of new particles and condensation on existing particles for gas-to-particle conversion [22].

For single component particles combining (7), (11) and (15) gives the semi-discrete aerosol dynamics equation

$$A n'(t) = \underbrace{G n(t)}_{\text{growth}} + \underbrace{[(B - C) \times n(t)] n(t)}_{\text{coagulation}} + \underbrace{A S(t)}_{\text{sources}} - \underbrace{A R(t) n(t)}_{\text{deposition}} . \quad (16)$$

This is a system of  $s$  coupled ordinary differential equations. The discrete initial conditions

$$n(0) = n^0 \quad (17)$$

are derived by projecting the continuous initial distribution  $n_0(v)$  onto the finite-dimensional solution space,

$$n^0(v) = \sum_{i=1}^s n_i^0 \phi_i(v) , \quad n^0 = [n_1^0, \dots, n_s^0]^T .$$

## 4.6 Time integration

The system (16)-(17) can be solved by any appropriate time-stepping method. The system has a particular form: the growth term is linear, while the coagulation term is bilinear. This makes it easy to express the Jacobian for implicit methods.

Particularly attractive are linearized versions of the implicit numerical methods which avoid iterative solutions. The following linearized backward Euler time discretization has second order time accuracy for the coagulation term:

$$A n(t^{k+1}) = A n(t^k) + \frac{\Delta t}{2} J^{\text{coag}}(n(t^k)) n(t^{k+1}), \quad (18)$$

where the Jacobian of the coagulation term is

$$J^{\text{coag}}(n) = \begin{bmatrix} n^T D^1 \\ \vdots \\ n^T D^s \end{bmatrix}, \quad D^i = (B^i - C^i) + (B^i - C^i)^T, \quad 1 \leq i \leq s. \quad (19)$$

This can be coupled with the Crank-Nicholson method for the growth and source terms to obtain the second order, non-iterative scheme

$$\left( A - \frac{\Delta t}{2} [J^{\text{coag}}(n(t^k)) + G + A R(t^{k+1})] \right) n(t^{k+1}) = \left( A + \frac{\Delta t}{2} [G + A R(t^k)] \right) n(t^k) + \frac{\Delta t}{2} [S(t^{k+1}) + S(t^k)]. \quad (20)$$

## 4.7 Extension to volume densities

The framework can be easily extended to treat volume, surface, or mass densities. For example the volume density  $\mathcal{V}(v, t) = v n(v, t)$  can be discretized as

$$\mathcal{V}(v, t) = \sum_{i=1}^s \mathcal{V}_i(t) \phi_i(v).$$

The aerosol dynamics equation for volume densities is

$$\begin{aligned} \frac{\partial \mathcal{V}(v, t)}{\partial t} &= \\ \text{(growth)} & - \partial [\mathcal{V}(v, t) I(v, t)] / \partial v + \mathcal{V}(v, t) I(v, t) / v \\ \text{(coagulation)} & + \int_0^v \frac{\beta_{v-w, w}}{w} \mathcal{V}(v-w, t) \mathcal{V}(w, t) dw - \mathcal{V}(v, t) \int_0^\infty \frac{\beta_{v, w}}{w} \mathcal{V}(w, t) dw \\ \text{(sources, deposition)} & + S(v, t) - R(v, t) \mathcal{V}(v, t). \end{aligned} \quad (21)$$

A discretization of equation (21) can be obtained following the framework approach; one obtains a discrete system of the form (20) with B, C and G redefined accordingly.

## 4.8 Extension to multiple components

Complex models treat particles composed of multiple chemical constituents. Let  $v_q(v, t)$ ,  $q = 1, \dots, m$  be the volume of the  $q$ -th chemical component in particles of volume  $v$ ; the multi-component aerosol population is described by the individual volume densities of each constituent  $\mathcal{V}^q(v, t) = v_q(v, t) n(v, t)$ ; the total volume of component  $q$  (per unit volume of air) contained in all particles having individual volumes between  $v$  and  $v + dv$  is  $\mathcal{V}^q(v, t) dv$ . Under these transformations the volume densities of each constituent  $\mathcal{V}^q(v, t)$ ,  $q = 1, \dots, m$  change according to [4, 16]

$$\begin{aligned} \frac{\partial \mathcal{V}^q(v, t)}{\partial t} &= \\ \text{(growth)} & - \partial [\mathcal{V}^q(v, t) \sum_{k=1}^m I_k(v, t)] / \partial v + \mathcal{V}^q(v, t) I_q(v, t) / v \\ \text{(coagulation)} & + \int_0^v \frac{\beta_{v-w, w}}{w} \mathcal{V}^q(v-w, t) \mathcal{V}^q(w, t) dw - \mathcal{V}^q(v, t) \int_0^\infty \frac{\beta_{v, w}}{w} \mathcal{V}^q(w, t) dw \\ \text{(sources)} & + S_q(v, t) \\ \text{(deposition)} & - R_q(v, t) \mathcal{V}^q(v, t) \\ \text{(chemistry)} & + K(\mathcal{V}_1, \dots, \mathcal{V}_m, t), \end{aligned} \quad (22) \quad q = 1, \dots, m,$$

where  $\mathcal{V}(v, t) = \sum_{q=1}^m \mathcal{V}^q(v, t)$  is the total volume distribution; the  $m$  integro-differential equations are coupled through  $\mathcal{V}(v, t)$  and  $K(\mathcal{V}_1, \dots, \mathcal{V}_m, t)$ . The system (22) is subject to the initial and boundary conditions

$$\mathcal{V}^q(v, t=0) = (\mathcal{V}^q)^0(v), \quad \mathcal{V}^q(v=0, t) = 0, \quad q = 1, \dots, m. \quad (23)$$

The equations (22)-(23) can be discretized in size using the same approach. For each component volume one has

$$\mathcal{V}^q(v, t) = \sum_{i=1}^s \mathcal{V}_i^q(t) \phi_i(v), \quad \mathcal{V}_i^q(t) = \mathcal{V}^q(V_i, t), \quad q = 1, \dots, m, \quad (24)$$

and the semidiscrete system reads

$$\begin{aligned} \frac{d}{dt} \mathcal{V}^q(t) = & \underbrace{G \mathcal{V}^q(t) - \text{diag}_i \left( \frac{I_q(V_i)}{V_i} \right) \sum_{k=1}^m \mathcal{V}^k(t)}_{\text{growth}} \\ & + \underbrace{[B \times \mathcal{V}^q(t)] \mathcal{V}^q(t) - \left[ C \times \sum_{k=1}^m \mathcal{V}^k(t) \right] \mathcal{V}^q(t)}_{\text{coagulation}} \\ & + \underbrace{S(t)}_{\text{nucl.+em}} - \underbrace{R_q \mathcal{V}^q(t)}_{\text{dep.}} + \underbrace{K(\mathcal{V}^1, \dots, \mathcal{V}^m, t)}_{\text{chem.}}, \quad q = 1, \dots, m. \end{aligned}$$

The matrix  $G$  and the tensors  $B$  and  $C$  are redefined according to (22). Note that the same  $G, B, C$  are used for all chemical components  $q$ , which makes the method efficient.

## 5 Piecewise-polynomial discretization

Consider now the discrete space of piecewise-polynomials of order smaller than or equal to  $r$ ,  $P_r(V_{\min}, V_{\max})$ . We assume that  $s - 1$  is a multiple of  $r$ ; the functions are polynomial on each interval  $[V_{1+\ell r}, V_{1+(\ell+1)r}]$ . A basis of  $P_r$  is provided by the Lagrange polynomials in each interval, with the  $r + 1$  nodes given by  $V_{1+\ell r}, V_{2+\ell r}, \dots, V_{1+r+\ell r}$ . Thus, the basis functions  $\phi_i$  have compact support, are piecewise-polynomials of order less than or equal to  $r$  and satisfy the relation

$$\phi_i(V_j) = \begin{cases} 1, & \text{for } i = j, \\ 0, & \text{for } i \neq j. \end{cases}$$

The discrete approximation

$$n(v, t) = \sum_{i=1}^s n(V_i, t) \phi_i(v) = \sum_{i=1}^s n_i(t) \phi_i(v)$$

is the order  $r$  piecewise-polynomial interpolant of  $n(v, t)$  with nodes  $V_i$ . Let  $h = \max_i |V_{i+1} - V_i|$ . Assuming smooth solutions  $n(v, t)$  the interpolation order is

$$\left| n(v, t) - \sum_{i=1}^s n(V_i, t) \phi_i(v) \right| = \mathcal{O}(h^{r+1}).$$

In general one expects that the approximation order is given by the interpolation order.

A direct approach is to consider the system as being discretized first in time then in size. The time integration method (18) applied to the coagulation system leads to the linear second order Fredholm equation (with unknown function  $n(v, t^{k+1})$ )

$$\begin{aligned} n(v, t^{k+1}) = & n(v, t^k) + \frac{\Delta t}{2} \int_0^v \frac{\beta_{v-w, w} + \beta_{w, v-w}}{2} n(v-w, t^k) n(w, t^{k+1}) dw \\ & - \frac{\Delta t}{2} n(v, t^{k+1}) \int_0^\infty \beta_{v, w} n(w, t^k) dw - \frac{\Delta t}{2} n(v, t^k) \int_0^\infty \beta_{w, v} n(w, t^{k+1}) dw. \end{aligned} \quad (25)$$



The Galerkin or collocation discretization over  $P_r(V_{\min}, V_{\max})$  compute a solution which approximates  $n(v, t^{k+1})$  with an accuracy of  $\mathcal{O}(h^{r+1})$ , see for example [1, Section 3.1.3].

In practice the observed convergence orders may be faster than this theoretical bound (“superconvergence”). For example, the collocation method approximates

$$\begin{aligned} n(V_j^c, t) \int_0^\infty \beta_{V_j^c, w} n(w, t) dw &\approx \sum_{k, m=1}^s n_k(t) n_m(t) C_{k, m}^j \\ &= \left( \sum_{k=1}^s n_k(t) \phi_k(V_j^c) \right) \int_0^\infty \beta_{V_j^c, w} \left( \sum_{m=1}^s n_m(t) \phi_m(w) \right) dw . \end{aligned}$$

If the collocation points are the node points  $V_j^c = V_j$  then  $n(V_j, t) = \sum_{k=1}^s n_k(t) \phi_k(V_j)$  by the interpolation requirement. For smooth  $\beta$  the term  $\int_0^\infty \beta_{V_j^c, w} (\sum_{m=1}^s n_m(t) \phi_m(w) dw)$  is a weighted Newton-Cotes approach for integration, therefore if  $r$  is even the negative term is approximated within  $\mathcal{O}(h^{r+2})$ . However, this does not hold for the positive coagulation term (consider for example the case  $j = 1$ ).

The collocation method is less expensive computationally and provides the same order of accuracy, so in principle it is to be preferred. Note at this point that if the collocation points are the node points then the mass matrix  $A$  is the identity matrix, and the tensor  $C$  is 2-dimensional,

$$C^j = \left[ \delta_{k, j} \int_0^\infty \beta_{V_j^c, w} \phi_m(w) dw \right]_{1 \leq k, m \leq s} , \quad 1 \leq j \leq s .$$

The order of accuracy gives a hint on how to carry the numerical evaluation of integral coefficients; if  $B, C$  are approximated within  $\mathcal{O}(h^{r+1})$  the order of accuracy remains unchanged, and if the integral coefficients are approximated within  $\mathcal{O}(h^{r+2})$  the integration errors become negligible when compared to discretization errors for small  $h$ . One can use a Gaussian quadrature with  $(r + 1)/2$  nodes.

The pure Galerkin approximation of the growth term is accurate of order  $\mathcal{O}(h^r)$  [10, Section 9.3]. Note that an upwind Petrov-Galerkin method is more appropriate to ensure stability of the hyperbolic growth term, in which case the order of approximation may be  $\mathcal{O}(h^{r+0.5})$ ; a discussion is outside the scope of this paper. The collocation method for piecewise polynomial basis cannot be directly extended to the growth term due to non-differentiability at node points (which were chosen to be the collocation points also).

This discussion leads to the following idea for an  $\mathcal{O}(h^{r+1})$  discretization of the coagulation-growth equation. Discretize the coagulation term using collocation on  $P_r$  and use a Galerkin approximation on  $P_{r+1}$  for the growth term. Replace the integrals by repeated  $[r/2]$ -point Gaussian quadrature on each  $[V_i, V_{i+1}]$  subinterval. Matrices  $A$  and  $G$  are then defined using the basis polynomials of  $P_{r+1}$ , while the tensors  $B$  and  $C$  are computed from the basis functions of  $P_r$ . Using the timestepping formula (20) the combined numerical method for coagulation-growth can be written as

$$\left( A - \frac{\Delta t}{2} A J^{\text{coag}} (n(t^k)) - \frac{\Delta t}{2} G \right) n(t^{k+1}) = \left( A + \frac{\Delta t}{2} G \right) n(t^k) .$$

## 6 Numerical experiments

**Test problem I.** For the numerical experiments we consider the test problem from [5], which admits an analytical solution. Let  $N_t$  be the total initial number of particles and  $V_m$  the mean initial volume. The initial number distribution is exponential, the coagulation rate is constant, and the growth rate is linear with the volume:

$$N_t(v) = (N_t/V_m) e^{-v/V_m} , \quad \beta(v, w) = \beta_0 , \quad I(v) = \sigma v .$$

This test problem admits an analytical solution, which is given in [5]

$$n(v, t) = \frac{4N_t}{V_m(N_t\beta_0 t + 2)^2} \cdot \exp \left( \frac{-2v \exp(\sigma_o t)}{V_m(N_t\beta_0 t + 2)} - \sigma_o t \right) . \quad (26)$$

We solve the dynamics equation for  $\beta_o = 6.017 * 10^{-10} \frac{\text{cm}^3}{\text{sec} \cdot \text{particles}}$ ,  $\sigma_o = 10^{-1} \frac{1}{\text{sec}}$ ,

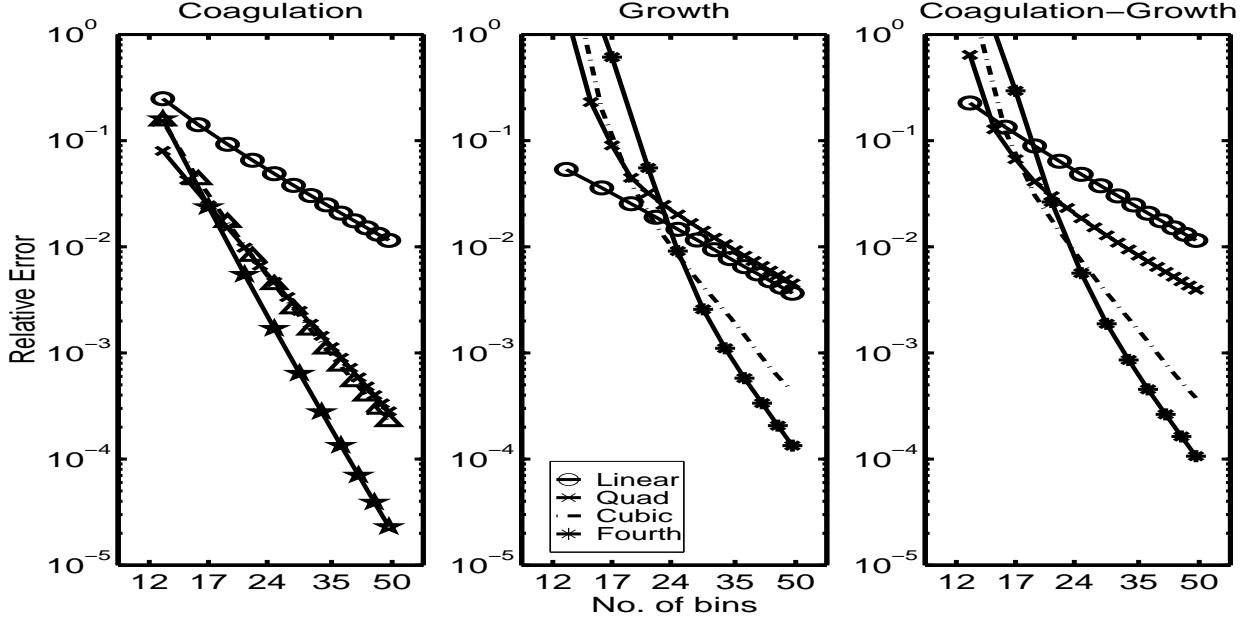


Figure 1: Solution RMS errors at  $t_{final} = 6$  hours for test problem I.

$N_t = 10^4$  particles,  $V_m = 10^{-1} \mu\text{m}^3$ .

Time interval:

$$[t_0 = 0, t_{final} = 12] \text{ hours}, \quad \Delta t = 10^{-2} \text{ hours}.$$

Volume interval:

$$[V_{\min} = 0, V_{\max} = \pi/6] \mu\text{m}^3.$$

The discretization errors are presented in Figure 1. Different slopes show different convergence orders for different element types. The experimental orders (the slopes) are better than our analysis predicted, showing superconvergence at the grid points.

**Test problem II.** We now consider a second test problem that is posed in naturally logarithmic coordinates. Here  $\beta_o = 1.083 \times 10^{-3} \text{ cm}^3 \text{ hour}^{-1} \text{ particles}^{-1}$ ,  $I(v) = 0.02 \mu\text{m}^3 \text{ hour}^{-1} = \text{const}$ , and  $N_t = 10^4$  particles. The volume interval is  $V_{\min} = 10^{-3} \mu\text{m}^3$ ,  $V_{\max} = 1 \mu\text{m}^3$ , the time interval  $[t_0 = 0, t_{final} = 6 \text{ hours}]$ , and the time step  $\Delta t = 1$  second.

The initial concentration is a cosine hill in logarithmic coordinates

$$n_0(v) = \begin{cases} \frac{N_t}{2} \cdot \left[ 1 - \cos \left( 2\pi \frac{\log v - x_{\min}}{x_{\max} - x_{\min}} \right) \right], & \log V_{\min} < x_{\min} < \log v < x_{\max} < \log V_{\max} \\ 0, & \log v \leq x_{\min} \text{ or } \log v \geq x_{\max} \end{cases}.$$

The reference solution was obtained using the standard numerical method for coagulation [9] on the uniform grid  $V_i = i \cdot \Delta v$ ,  $\Delta v = 10^{-3} \mu\text{m}^3$ , such that  $V_1 = V_{\min}$  and  $V_{1000} = V_{\max}$ . The reference growth-coagulation solution is obtained by translating the reference coagulation solution to the right by an appropriate number of bins. Figure 3 shows the initial distribution, as well as the coagulation-growth solution after 1 hour.

Figure 2 displays the numerical errors. A meaningful solution for coagulation can be obtained with as few as 7 bins. For growth one needs at least 50 points to obtain a good numerical solution. The results for all elements show a second order convergence, a fact that we cannot explain at this time.

To solve the coagulation-growth problem the obvious approach is to increase the number of bins to 50, such that each subproblem is treated correctly; the cost of building 50-dimensional tensors for coagulation is however significant.

In order to avoid these extra costs we propose the following approach, based on different grids for coagulation and for growth. Let  $\{x_i\}$ ,  $1 \leq i \leq s$  and  $\{x'_j\}$ ,  $1 \leq j \leq s'$  be two sets of gridpoints points on

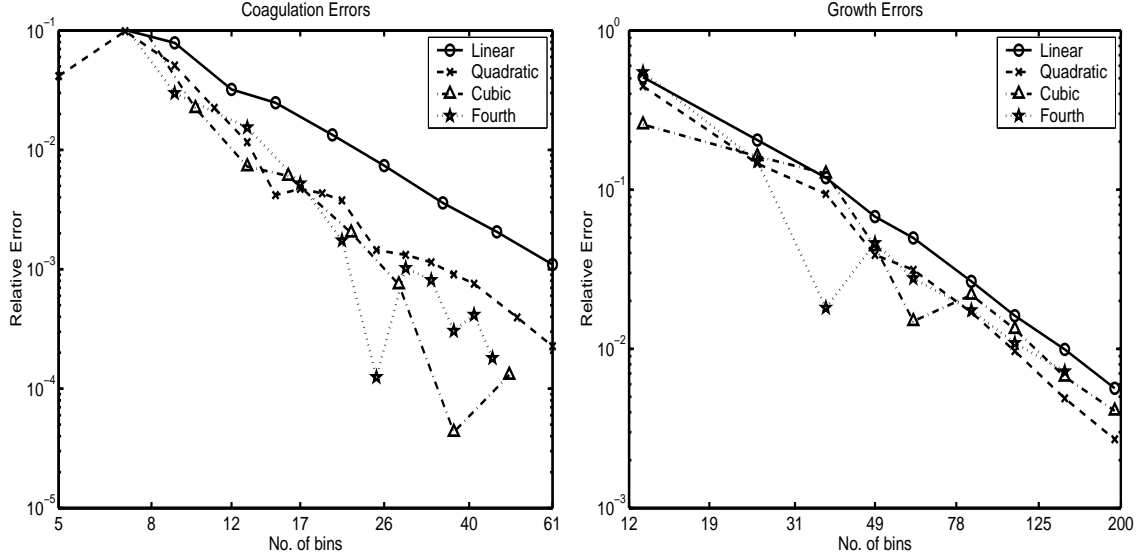


Figure 2: Solution RMS errors at  $t_{final} = 1$  hour for test problem II.

$[V_{\min}, V_{\max}]$ . If the function  $N$  is represented on grid  $x$  as  $[N_1, \dots, N_s]^T$  then a representation on the grid  $x'$  is given by piecewise-polynomial interpolation:

$$\begin{bmatrix} N'_1 \\ \vdots \\ N'_{s'} \end{bmatrix} = [\phi_i(x'_j)]_{1 \leq i \leq s, 1 \leq j \leq s'}^T \cdot \begin{bmatrix} N_1 \\ \vdots \\ N_s \end{bmatrix} = T^{x, x'} \cdot \begin{bmatrix} N_1 \\ \vdots \\ N_s \end{bmatrix},$$

where  $\phi_i$  are the basis piecewise-polynomials associated with the points  $\{x_i\}$ .

This allows us to use a finer grid for growth and a coarser grid for coagulation. This also allows to combine a logarithmic grid for coagulation and a linear grid for growth. The method can be formulated on the fine grid as

$$\left( A - \frac{\Delta t}{2} A \cdot T^{\text{up}} \cdot J^{\text{coag}} (T^{\text{down}} n^k) \cdot T^{\text{down}} - \frac{\Delta t}{2} G \right) n^{k+1} = \left( A + \frac{\Delta t}{2} G \right) n^k. \quad (27)$$

where  $T^{\text{down}}$  and  $T^{\text{up}}$  are the transformation matrices from fine to coarse and from coarse to fine (it is easy to see that coagulation is solved on the coarse grid, and the solution extended to the fine grid).

The results displayed in Figure 3 are obtained using cubic elements (in logarithmic coordinate) with 145 gridpoints for growth and 13 gridpoints for coagulation. The reference solution is reproduced quite well, with a rms error norm of about  $1.4\text{e-}2$ .

## 7 Conclusions

Aerosols are becoming an important topic in air pollution modeling. For a correct representation of particles in the atmosphere one needs to accurately solve for the size distribution of particle populations.

In this work we develop a general framework for the discretization of aerosol dynamics equation using projection methods, which include Galerkin and collocation approaches. The resulting semidiscrete system is bilinear and is solved by a second order order linearly-implicit time stepping method. Although the formulation is given in terms of number densities and single-component particles, the ideas extend directly to volume, surface and mass densities, as well as to multiple-component aerosols.

To exemplify the use of the framework we consider piecewise-polynomial discretizations with linear, quadratic, cubic and fourth order elements. The collocation approach was used for coagulation and the Galerkin approach for growth. Two test problems are employed: one with analytical solution in the infinite

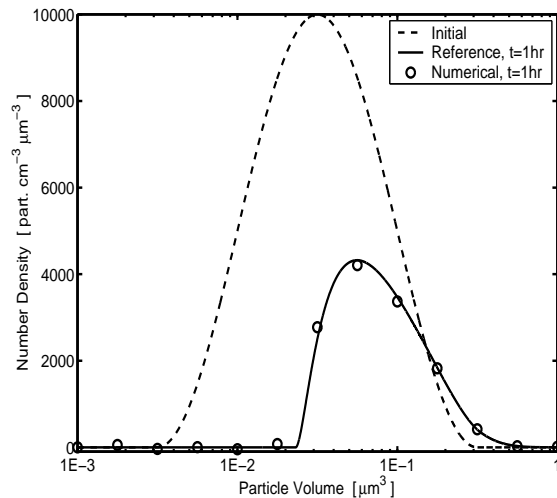


Figure 3: The coupled coagulation-growth solutions at  $t_{final} = 1$  hour for test problem II. The numerical solution is obtained using a combined grid with  $s=145$  for growth and  $s=13$  for coagulation.

volume range  $v \in [0, \infty]$ ; and a second one with the solution “living” in a finite volume interval and formulated in logarithmic coordinates.

In the standard formulation (linear coordinates) the numerical solution displays excellent accuracy – the convergence orders at node points are one order better than those predicted by a direct theory. In logarithmic coordinates (preferred by environmental modelers) good solutions can be obtained for the coagulation equation; but the growth problem is ill-scaled and can be solved accurately only by increasing the number of bins. A mixed solution is proposed to solve growth on a fine grid and coagulation on a coarse grid. Lower accuracy is observed in logarithmic coordinates, although the numerical solutions reproduce quite well the reference solutions.

Future work will focus on testing the discretization framework on multiple component particles and on coupled aerosol dynamics and chemistry models.

## Acknowledgements

This work was supported by NSF CAREER award ACI-0093139.

## References

- [1] K.E. Atkinson. *The Numerical Solution of Integral Equations of the Second Kind*. Cambridge University Press, 1997.
- [2] F.S. Binkowski and U. Shankar. The regional particulate matter model: 1: model description and preliminary results. *Journal of Geophysical research*, 100:26,191–26,209, 1995.
- [3] A. Bott. A positive definite advection scheme obtained by nonlinear renormalization of the advection fluxes. *Monthly Weather Review*, 117:1006–1115, 1989.
- [4] F.M. Gelbard and J.H. Seinfeld. Coagulation and growth of a multicomponent aerosol. *Journal of Colloid and Interface Science*, 63(3):472–479, 1978.
- [5] F.M. Gelbard and J.H. Seinfeld. Numerical solution of the dynamic equation for particulate systems. *Journal of Computational Physics*, 28:357–375, 1978.
- [6] F.M. Gelbard and J.H. Seinfeld. Simulation of multicomponent aerosol dynamics. *Journal of Colloid and Interface Science*, 78(2):485–501, 1980.

- [7] M.Z. Jacobson. Development and application of a new air pollution modeling system - II. Aerosol module structure and design. *Atmospheric Environment*, 31(2):131–144, 1997.
- [8] M.Z. Jacobson. Numerical techniques to solve condensational and dissolutional growth equations when growth is coupled to reversible reactions. *Aerosol Science and Technology*, 27:491–498, 1997.
- [9] M.Z. Jacobson. *Fundamentals of atmospheric modeling*. Cambridge University Press, 1999.
- [10] C. Johnson. *Numerical Solution of Partial Differential Equations by the Finite Element Method*. Cambridge University Press, 1987.
- [11] Y.P. Kim and J.H. Seinfeld. Simulation of multicomponent aerosol condensation by the moving sectional method. *Journal of Colloid and Interface Science*, 135(1):185–199, 1990.
- [12] F.W. Lurmann, A.S. Wexler, S.N. Pandis, S. Musara, N. Kumar, and J.H. Seinfeld. Modeling urban and regional aerosols - ii. application to California’s south coast air basin. *Atmospheric Environment*, 31:2695–2715, 1997.
- [13] A.A. Lushnikov. Evolution of coagulating systems iii. coagulating mixtures. *Journal of Colloid and Interface Science*, 54(1):94–101, 1976.
- [14] Z. Meng, D. Dabdub, and J.H. Seinfeld. Size-resolved and chemically resolved model of atmospheric aerosol dynamics. *Journal of Geophysical Research*, 103(D3):3419–3435, 1998.
- [15] M. Phadnis and G.R. Carmichael. Transport and distribution of primary and secondary non-methane hydrocarbons in east Asia under continental outflow conditions. *J. Geophys. Res.*, in press, 1999.
- [16] C. Pilinis. Derivation and numerical solution of the species mass distribution equation for multicomponent particulate systems. *Atmospheric Environment*, 24A(7):1923–1928, 1990.
- [17] J.H. Seinfeld and S.N. Pandis. *Atmospheric chemistry and physics. From air pollution to climate change*. John Wiley & Sons, Inc., 1997.
- [18] C.H. Song and G.R. Carmichael. Partitioning of hno<sub>3</sub> modulated by alkaline aerosol particles. *J. Geophys. Res.*, in review, 1999.
- [19] C.H. Song, G.R. Carmichael, and S.Y. Cho. An alternative way to couple kinetic (non-equilibrium/condensation/evaporation) process with thermodynamic equilibrium relationships. *Atmospheric Environment*, to be submitted, 1999.
- [20] R.P. Turco, P. Hamil, O.B. Toon, R.C. Whitten, and C.S. Kiang.. The NASA-Ames Research Center stratospheric aerosol model: I. Physical processes and computational analogs. *NASA Technical Publication*, TP 1362, 1979.
- [21] H. Tsang and J.M. Hippe. Asymptotic behavior of aerosol growth in the free molecule regime. *Aerosol Science and Technology*, 8:265–278, 1988.
- [22] Y. Zhang, J.H. Seinfeld, M.Z. Jacobson, and F.S. Binkowski. Simulation of aerosol dynamics: a comparative review of algorithms used in air quality models. *submitted to AS&T*, 1999.