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Study and modification of a size-discrete semi-implicit simulation model for polydisperse aerosol coagulation

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ABSTRACT

Aerosols in many natural and industrial processes are affected by coagulation, the merging of aerosol particles. The present work focuses on a semi-implicit size-discrete method to simulate the evolution of the particle size distribution by coagulation. The derivation of the method is retraced and generalized in terms of the size definition and discretization schemes. Based on this, new forms for the partition coefficients, that implement the redistribution of coagulated particles into the discrete size intervals, are proposed. For this, either constant or linear particle size distribution functions in terms of number and volume concentrations are assumed in each size interval. These assumptions also impact the calculation of the mean volume of the particles in each discretization interval. The inclusion of the coagulation kernel, which describes the physical conditions and the collision mechanism, in the partition coefficients is also tested. The modifications are compared against existing methods in the reproduction of analytical solutions for certain coagulation problems. It was found that certain combinations of modifications were able to reduce the numerical error caused by particle size discretization by more than 80% in the tested cases.

1. Introduction

Aerosols consist of very small solid or liquid particles that are carried by a gas. In the form of clouds, fog and haze they are a crucial part of atmospheric physics. In chemical and process engineering, they are created during combustion and subsequent flue gas cleaning, produced in the form of technical nanoparticles, and they can also inadvertently form during absorption (Brachert, Kochenburger, & Schaber, 2013) and filmwise condensation (Fernández & Prieto, 2016).

Aerosol science is present in multiple fields. They have a great impact on climate so they are very present in climate models (Wang et al., 2013). Aerosols also appear in multiple industrial processes where heat and mass transport phenomena occur (Schaber, Körber, Ofenloch, Ehrig, & Deuflhard, 2002; Wix, Schaber, Ofenloch, Ehrig, & Deuflhard, 2007). They can be formed in cooling towers (Ishimatsu, Miyamoto, Hori, Tanaka, & Yoshida, 2001), in chemical industrial processes, biomass combustion (Martinsson et al., 2015), etc. To improve the understanding of atmospheric systems involving aerosols as well as the design of technical processes, accurate aerosol simulation tools are needed.

One of the principal mechanisms that determine the evolution of aerosol particle populations is coagulation. Under conditions of high particle concentration, like new particle formation in coastal regions (Dal Maso et al., 2002), or vehicle exhaust systems (Kim, Gautam, & Gera, 2002), coagulation can be an important phenomenon to understand the behaviour of aerosols. During coagulation,

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particles collide and subsequently merge together. This lowers the number of particles and increases their size, but the total volume of all particles remains constant. The phenomenon is particularly strong in systems with high particle concentrations or with consideration of long time intervals.

The evolution of the aerosol particle size distribution (PSD) due to coagulation can be described by an integro-differential equation. Many aerosol simulation tools include a coagulation model in which this equation is numerically solved. One particularly efficient method was presented by Turco, Hamill, Toon, Whitten, and Kiang (1979) for the application in atmospheric models. It uses a discretization of the particle size and a semi-implicitly mixed Euler integration scheme in time. The advantages include rapid computation of the solution because of the lack of iterations, inherent and unconditional stability, and exact conservation of the total particle volume. It has since been adapted and extended by Toon, Turco, Westphal, Malone, and Liu (1988), Jacobson, Turco, Jensen, and Toon (1994), and Fernández Díaz, González-Pola Muñiz, Rodríguez Braña, Arganza García, and García Nieto (2000), Fernández Díaz, Rodríguez Braña, Argüelles Díaz, Gómez García, and García Nieto (2003). Debry and Sportisse (2007) give comparative observations about different partition coefficients.

This work endeavors to expand the applicability of the method to arbitrary particle size definitions and discretization schemes. To this aim the derivation of the method will be retraced with generalized particle size notation. Based on the derivation, several variants for the calculation of the mean volumes and of the partition coefficients, that redistribute the coagulation product into the discrete size intervals, will be presented. Special cases of the coagulation problem, in which analytical solutions are known, will be used to evaluate the options with the aim of minimizing the numerical error of the method. The accuracy of new variants are compared to previous ones considering the needed computational effort. Brownian coagulation will also be evaluated in the case of atmospheric aerosols. The results of this work were already successfully applied by Brachert et al. (2013) in the simulation of sulfuric acid aerosols that are created during flue gas desulfurization in fossile power plants.

2. Derivation of the method

2.1. Continuous coagulation equation

Since the size of an aerosol particle is often defined by different characteristics, e.g., radius, diameter, or volume, in linear or logarithmic scales, a generalized notation will be used in this article. Thus, the variables x, y, z represent an arbitrary particle size parameter and $v(x)$, which has to be a strictly increasing function, denotes the volume of a particle with size x. To facilitate the description of coagulation with these variables, volume-conserving addition and subtraction operators $\oplus$ and $\ominus$ are defined, so that $z = x \oplus y$ represents the size of the coagulation product of two particles with sizes x and y. Because the total volume of all particles remains constant during coagulation, $v(z) = v(x) + v(y)$. The reverse operator $\ominus$ works correspondingly ($z \ominus y = x$).

Following this definition, the evolution of the particle size distribution of a single-component aerosol undergoing coagulation can be described by the following integro-differential equation, modified from (Williams and Loyalka, 1991):

$$\frac{\partial n(x,\,t)}{\partial t} = \frac{1}{2}\int_0^x \beta(y,\,x\ominus y)\,n(y,\,t)\,n(x\ominus y,\,t)\,\mathrm{d}y - \int_0^\infty \beta(x,\,y)\,n(x,\,t)\,n(y,\,t)\,\mathrm{d}y\,. \tag{1}$$

The first term of the right-hand side represents the formation frequency of particles with size x through coagulation of particles with sizes y and $x\ominus y$. The second term describes the reduction of the number of particles with size x as they coagulate with other particles. The number density distribution $n(x,\,t)$ is defined so that $n(x,\,t)\,\mathrm{d}x$ is the number of particles with a size between x and $x + \mathrm{d}x$ per unit volume at a time t. $\beta(x,\,y)$, the coagulation kernel, is a parameter for the coagulation frequency between particles with sizes x and y. Its form depends on the mechanism that causes the particle collisions, e.g., Brownian motion or turbulence.

The volume density distribution $q(x,\,t)$ is obtained by multiplying $n(x,\,t)$ by the particle volume $v(x)$. Fernández Díaz et al. (2000) give an adaptation of the coagulation Eq. (1), that accounts for the change in the volume PSD:

$$\frac{\partial q(x,\,t)}{\partial t} = \int_0^x \beta(y,\,x\ominus y)\,q(y,\,t)\,n(x\ominus y,\,t)\,\mathrm{d}y - \int_0^\infty \beta(x,\,y)\,q(x,\,t)\,n(y,\,t)\,\mathrm{d}y\,. \tag{2}$$

2.2. Particle size discretization

To approach the numerical solution of the coagulation equation, the discretization scheme distributes the particle population into a number of adjacent size intervals called bins. The bins are referred to by integer numbers i, j, k that assume values between 1 and NB, the total number of bins. Each bin k contains the particles with sizes between k^- and k^+ and $k^+ = (k+1)^-$.

The discrete PSD is obtained by integrating the continuous distribution between the boundaries of each bin k, which returns the total number concentration N_k and the total volume concentration Q_k in that bin:

$$N_k(t) = \int_{k^-}^{k^+} n(x,\,t)\,\mathrm{d}x, \tag{3}$$

$$Q_k(t) = \int_{k^-}^{k^+} q(x,\,t)\,\mathrm{d}x. \tag{4}$$

Both values are related to each other through the mean volume $\bar{v}_k$ of the particles in bin k: