Journal of Computational Physics 326 (2016) 499–515

Contents lists available at ScienceDirect

Journal of Computational Physics

www.elsevier.com/locate/jcp

Characteristics-based sectional modeling of aerosol nucleation and condensation

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ARTICLE INFO

Article history:
Received 16 April 2015
Received in revised form 29 April 2016
Accepted 2 September 2016
Available online 14 September 2016

Keywords:
Aerosol
Eulerian
Multi-phase
Characteristics
Droplet size distribution
Sectional

ABSTRACT

A new numerical method for the solution of an internally mixed spatially homogeneous sectional model for aerosol nucleation and condensation is proposed. The characteristics method is used to predict droplet sizes within a discrete time step. The method is designed such that 1) a pre-specified number of moments of the droplet size distribution may be preserved, 2) there exists no time step stability restriction related to the condensation rate and section size, 3) highly skewed *fixed* sectional distributions may be used and 4) it is straightforward to extend to spatially inhomogeneous settings and to incorporate droplet coagulation and break-up. We derive, starting from mass conservation, a consistent internally mixed multi-species aerosol model. For certain condensational growth laws analytical solutions exist, against which the method is validated. Using two-moment and four-moment-preserving schemes, we find first order convergence of the numerical solution to the analytical result, as a function of the number of sections. As the four-moment-preserving scheme does not guarantee positivity of the solution, a hybrid scheme is proposed, which, when needed, locally reverts back to two-moment preservation, to prevent negativity. As an illustration, the method is applied to a complete multi-species homogeneous nucleation and condensation problem.

1. Introduction

An aerosol is defined as a mixture of a particulate phase suspended in a carrier gas. Aerosols are around us everywhere, as clouds, smoke, sprays or dust, and we are exposed to them daily. When the particulate phase consists of liquid droplets, the aerosol is often formed by means of nucleation and condensation, with subsequent coagulation and break-up. These processes must be captured accurately to predict the properties of such an aerosol. We propose a new numerical method for a sectional aerosol method, which is capable of solving the General Dynamic Equation (GDE) for the droplet size distribution of an aerosol undergoing nucleation and condensational growth. In contrast to other methods, the new approach does not have a condensational time step restriction, making it an efficient and robust algorithm for predicting aerosol formation and evolution.

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http://dx.doi.org/10.1016/j.jcp.2016.09.005
0021-9991/

The properties of an aerosol can be characterized in terms of composition of the vapor phase and the size distribution of the particulate phase, quantifying the frequency at which a particle with a specific composition and size occurs in the system. In Computational Fluid Dynamics (CFD), there are two main ways of modeling the aerosol and computing these properties as a function of time and space: one can adopt either a Lagrangian or an Eulerian description of the particulate phase. In a Lagrangian description each particle is tracked individually, and explicit averaging over the particles is used to extract property distributions. In an Eulerian description, one computes a selected number of spatially averaged properties of the size distribution. In general, the Lagrangian approach may be considered as the more fundamental approach, as neither spatial averaging is performed, nor are any assumptions made on the size or composition of individual particles. The change in size, composition and position of a particle follows from simple dynamic models, describing properties such as the position, velocity, temperature, size, composition, etc. In the Eulerian formulation, a distribution, or a number of corresponding moments of this distribution, are computed. The level of detail with which the distribution is described is reduced with respect to the Lagrangian approach, which makes the Eulerian description computationally preferential in complex aerosol systems with many particles.

In this paper, we adopt an Eulerian aerosol description, and set out to develop a robust and stable numerical method for finding a solution to the governing aerosol equations. In the formulation of the aerosol model we select the 'internally mixed' concept, defined in this paper to describe a system in which locally all particles have the same chemical composition, independent of size [1]. This reduces the complexity of the model significantly, compared to a size-dependent composition model. Note that composition may still vary spatially. In the Eulerian formulation there are three main branches of methods which can be used to determine the size distribution. The first, and generally most precise, is the *continuous method*, in which the size distribution is discretized using conventional methods like the finite element method or the finite difference method (e.g., see [2–6]). These methods are usually expensive, and almost exclusively applied to spatially homogeneous settings. Also, schemes may become unstable if the condensation rate is high [7]. This problem may be addressed by adding artificial diffusion, or by increasing the number of distribution discretization points, making the method inaccurate or very expensive, respectively. Complementary to continuous methods are *moment methods*, in which no longer the complete distribution is solved, but only a specific number of moments of the distribution. This greatly reduces the degrees of freedom of the system, making moment methods desirable in terms of computational efficiency. The main problem of moment methods is that they require closure, as the moments of interest may depend on higher order moments. To achieve closure, one can make an assumption on the shape of the distribution (e.g., see [8–11]). However, such an assumption may be too restrictive. A popular alternative to mitigate this problem is the Quadrature Method Of Moments (QMOM) (e.g., see [12–19]), in which higher order moments are related to lower order ones using quadrature integration. The QMOM method may become numerically challenging and suffers from a non-uniqueness problem [20]. Moreover, it may not be sufficiently general in terms of the shape of the distribution.

In between these two methods there is a third one: the *discrete population balance method*, often referred to as *sectional method* (e.g., see [21–23,20,24–26]). In this class of methods, the size domain is split up in a number of sections or bins, in which the size distribution function is assumed to be constant. This allows to solve for a total number concentration in each section, rather than the continuous distribution itself. The effects of for example coagulation on the size distribution can now be treated in a discrete way, where a finite number of collision events between two sections is considered, as in [27]. This sets aside the sectional method from the continuous method. A similar 'discrete' approach can be taken for condensational growth or droplet break-up, e.g., see [28]. On one hand, although less accurate, the sectional method offers a significant reduction in computational effort, in comparison with continuous methods [7]. On the other hand, in comparison with moment methods, the method is not subject to any sort of constraint in terms of an assumed particle distribution shape. Another key method is the Sectional Quadrature Method of Moments (SQMOM) [29], in which one also divides the size domain into sections, each having one 'primary particle' responsible for the size distribution reconstruction and 'secondary particles' used for low-order moment preservation in case of particle interactions. It is shown in [29] that all before-mentioned sectional and quadrature methods are special cases of SQMOM, illustrating the generality of the method and uniting the moment approach with the sectional approach.

We adopt the sectional formulation as presented in [27,28,30], which assigns the particles within a section to a unique 'representative size'. This method allows to exactly preserve a pre-specified number of moments of the distribution, and offers great flexibility on arbitrary grids. In [30] a moving section method was proposed, to solve for condensational growth. Given the hyperbolic character of condensational growth, it is straightforward to incorporate the method of characteristics to convect sections in the size domain along trajectories. This method eliminates numerical diffusion and preserves moments exactly. However, in a spatially inhomogeneous setting, such a method introduces complications when computing spatial gradients [20]. One must either interpolate back to a fixed grid, or one must abandon the idea of using the characteristics, and compute the condensational growth on a fixed grid directly. The former was done by, for example, Mitrakos et al. [20], using a third order polynomial for the interpolation, which is argued to lead to significant reduction of numerical diffusion compared to lower order interpolations. However, only the zeroth moment of the distribution is preserved. Fixed grid methods (e.g., see [31–33]) show an analogy with the Finite Volume (FV) treatment of a convective term: fluxes of droplets growing from one section to its neighbor are determined by interpolation to section interfaces of the approximate size distribution. As shown in [30], these methods suffer from instabilities. Moreover, they reduce the global numerical time step size significantly, when condensation is rapid.