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Project Summary

Modal Aerosol Dynamics Modeling

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The report described by this Project Summary presents the governing equations for representing aerosol dynamics, based on several different representations of the aerosol size distribution. Analytical and numerical solution techniques for these governing equations are also reviewed. Described in detail is a computationally efficient numerical technique for simulating aerosol behavior in systems undergoing simultaneous heat transfer, fluid flow, and mass transfer in and between the gas and condensed phases. The technique belongs to a general class of models known as *modal aerosol dynamics* (MAD) models. These models solve for the temporal and spatial evolution of the particle size distribution function. Computational efficiency is achieved by representing the complete aerosol population as a sum of additive overlapping populations (modes), and solving for the time rate of change of integral moments of each mode. Applications of MAD models for simulating aerosol dynamics in continuous stirred tank aerosol reactors and flow aerosol reactors are provided. For the application to flow aerosol reactors, the discussion is developed in terms of considerations for merging a MAD model with the SIMPLER routine described by Patankar (1980). Considerations for incorporating a MAD model into the U.S. Environmental Protection Agency's Regional Particulate Model are also described.

Numerical and analytical techniques for evaluating the size-space integrals of the modal dynamics equations (MDEs) are described. For multimodal lognormal distributions, an analytical expression for the coagulation integrals of the MDEs, applicable for all size regimes, is derived, and is within 20% of accurate numerical evaluation of the same moment coagulation integrals. A computationally efficient integration technique, based on Gauss-Hermite numerical integration, is also derived.

This Project Summary was developed by EPA's Atmospheric Research and Exposure Assessment Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

For processes in the atmosphere, computer models are indispensable tools for assessing the environmental impact of anthropogenic and biogenic sources of gases, vapors, and particles, because real-world emission rates are not easily changed and comprehensive data sets are difficult to obtain. Atmospheric models that are calibrated with existing data can be used to assess the impact of various proposed strategies for altering atmospheric emissions of air pollutants.

Many atmospheric aerosol models have been developed using finite-difference techniques. Finite-difference techniques are commonly used because they allow very general models to be constructed, and the accuracy and computational effort can be altered by adjusting the grid struc-

ture. The concept of a "grid-independent" solution implies a model whose accuracy has converged within "reasonable" limits to the true solution. Increased computational speed with such models, however, is usually obtained by a deliberate sacrifice of accuracy, such as by approximating a truly three-dimensional situation with a two-dimensional simulation, and/or by implementing a coarse grid system to maintain the resulting simulation within the capabilities of the computer used.

High computational speed and accuracy can also be obtained by sacrificing the flexible structure of finite-difference techniques, and building a modeling structure based on a physical representation of the dynamic system modeled. For example, it has often been observed that aerosol size distributions in the atmosphere and elsewhere can accurately be approximated by multimodal lognormal functions. This provides a motivation for developing aerosol dynamics models that assume distributions to be lognormal, which facilitates the derivation of moment equations that are relatively straightforward to integrate.

Because the computational advantage of the moment modeling technique is so great, moment models for a variety of aerosol systems have been developed and used by many researchers. This report surveys the literature on aerosol dynamics models, and presents the general form of the governing equations used in these models. It also provides a detailed derivation of the governing equations used in modal aerosol dynamics models.

Simulating the Dynamics of Aerosol Systems

Modeling aerosol dynamics has at least three challenging aspects: a distribution of particle sizes must be considered; coagulation introduces summations and integrals over the size distribution; and particle sizes of interest in many systems span the free-molecule, transition, and continuum size regimes, requiring generalized expressions that apply to all size regimes to be used. Objectives of aerosol dynamics modeling include determining the particle size distributions and mixing characteristics of all chemical compounds as functions of time and position, and determining the magnitude and location of particle deposition.

The dynamics equations for the aerosol phase are formulated by expressing mathematically the effect of each process that represents interactions with the aerosol phase. The governing differential equation is commonly referred to as the general dynamics equation (GDE) (Friedlander, 1977). In this report, various forms of the GDE for representing the time dependence of the aerosol size distribution are presented. These are broadly classified as discrete, sectional, continuous-spline, and modal. Aerosol dynamics models based on the different forms of the GDE are reviewed and compared. The derivation of a computationally efficient aerosol dynamics modeling technique—modal aerosol dynamics (MAD) modeling—is given in detail, and an application of this modeling technique for simulating atmospheric aerosol processes is presented.

Solving for the temporal and spatial evolution of the size distribution, n, is tantamount to solving for the coefficients of the approximating functions. The mathematical representation of the size distribution function determines the form of the GDE solved, and hence the solution algorithms required to construct a complete model. Because the solution techniques used to solve for the function coefficients depend on the mathematical representation of n, aerosol dynamics models are commonly named according to the mathematical representation of n. Commonly used aerosol models that approximate the aerosol size distribution with a set of mathematical functions are the discrete, continuous-spline, sectional, and MAD models. Discrete, continuous-spline, and sectional models have been described in the literature, and are reviewed in this report. To the authors' knowledge, however, a basic derivation of the governing dynamics equations for MAD models does not exist in the literature.

The discrete form is the most general representation of the size distribution, where separate equations for the time-dependent concentration of each cluster size are formulated. The cluster size increments are determined by the smallest molecule causing cluster growth. Discrete models are useful for simulating nucleation processes, where the dynamics of clusters with relatively few i-mers are important.

The continuous-spline representation is so called because particle size-space is subdivided into a series of contiguous sections, and the section functions are splined smoothly together at the section boundaries. Cubic functions are commonly used, implying that four differential equations (one for each coefficient of the cubic equation) must be solved for each section.

The sectional representation is so called because the particle size space is subdivided into a series of sections and the distribution function of a prescribed moment of the distribution (usually the mass distribution) is assumed constant within each section. This assumption results in a size distribution with the appearance of a histogram. For sectional techniques, one differential equation is required for each section.

The modal representation is so called because the particle size range is divided into a small number of overlapping intervals (usually three or less), called modes. Overlapping intervals are used because the modal approximation originates from the assumption that the complete distribution is composed of multiple distinct aerosol populations. Within each mode, n is represented by a continuous distribution function. Because the modes overlap, the distribution functions for all modes are added to yield an approximation for the entire distribution function. A differential equation is required for each independent modal function parameter.

The above modeling techniques are presented in order of decreasing computational requirements. It should be noted, however, that the less computer-intensive methods are not necessarily less accurate. For example, if an actual system can be accurately simulated by a highly parameterized distribution function, then aerosol dynamics models that employ such a distribution function should accurately simulate the system behavior. The associated savings in computational resources can make the difference between a practical model and a model that is too complex to be solved even with the most sophisticated available supercomputer. This is especially true for complicated real-world problems such as the behavior of atmospheric aerosols, which is one reason why highly parameterized models are still in use today.

Modal Aerosol Dynamics Models

Because aerosol distributions often result from the additive effect of multiple sources, distributions composed of multiple distinct aerosol populations (modes) occur in many systems of interest, and MAD models are often appropriate for simulating the behavior of such systems. The central physical assumption of MAD models is stated as follows:

> An aerosol may be viewed as an assemblage of distinct populations of particles, distinguished by size or chemical composition. The size distribution of each population is approximated by an analytical distribution function.

In general, aerosol models solve for the temporal and spatial evolution of the coefficients of the size distribution. For MAD models, however, differential equations for the modal function coefficients are replaced by differential equations for moments of each mode (e.g., total number or mass concentration). Solving for the time rate of change of a sufficient number of moments of the distribution allows the size distribution at every point in the time and space of interest to be determined. This report presents a derivation for converting the differential equations for multiple overlapping modes into an equivalent set of differential equations for the moments of these modes.

To account for the effects of processes on the size distribution in MAD models, the net effect of each process on each mode must be determined. This requires evaluating integral expressions for the dynamics of each process, and including these effects in the governing dynamics equations for each mode. One of the principal tasks in developing MAD models, therefore, is to develop solution algorithms for evaluating these integral terms. Coagulation processes are especially difficult to handle, because they represent particle-particle interactions, and this translates into double integrals over the interacting populations. To maintain computational efficiency with MAD models, care must be taken when formulating and solving these integral expressions. This report discusses in detail the derivation of these integral terms, and algorithms for analytically and/or numerically evaluating the resulting mathematical integrals. Many analytical expressions for common dynamical processes are provided.

A new technique for evaluating multimodal coagulation integrals has been derived that is within 20% of accurate numerical evaluations of the same coagulation integrals. Techniques for numerically evaluating the modal integrals based on Gauss-Hermite numerical integration are also described. The report's presentation on numerical integration techniques is important because failure to implement efficient integration techniques can result in the computational effort of the resulting model approaching that of one of the more detailed aerosol dynamics models. The accuracy and domain of applicability of the numerical techniques presented in this work are therefore clearly presented, to guide the modeler in selecting the technique most suited to the particular modeling application.

References

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The complete report, entitled "Modal Aerosol Dynamics Modeling," (Order No. PB91-161 729/AS; Cost: $31.00, subject to change) will be available only from:

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