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Project Summary

Urban Aerosol Modeling

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Accurate numerical schemes for simulation of the coagulation and condensation/evaporation (with vapor conservation) processes of single component aerosols were developed. The schemes were incorporated into modules that permit simulation of these processes in models of atmospheric dispersion and transport.

This Project Summary was developed by EPA's Environmental Sciences Research 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

Urban aerosols are associated with various adverse effects such as reduced visibility, inadvertent weather modification, and increased incidence of respiratory disease. An understanding of these associations requires, among other factors, knowledge of ambient aerosol size and composition distributions. The dynamic processes that produce these distributions include, in addition to atmospheric dispersion and transport, nucleation, coagulation, condensation/evaporation, and deposition. Models that include these processes are necessary in order to develop aerosol air quality regulations and control strategies. There are currently no validated urban aerosol models that deal with the complexity of these processes.

The research project described in detail in the Project Report (see ordering information at back) involved development of urban aerosol models that include the dynamic processes shaping urban aerosol size and composition distributions. First, data bases and a K-Theory model for the super- and sub-micrometer aerosol mass concentrations were validated. This Project Summary describes our more recent work on the development of computer modules that accurately describe the coagulation and condensation/evaporation processes of single component aerosols. These modules can be employed in the K-Theory model or in other models.

Theoretical Framework

Modules that would incorporate aerosol growth processes in a general atmospheric dispersion and transport model were developed. In order to understand how growth modules operate, it is useful to illustrate their incorporation in a K-Theory model (as used in this project). For a single component aerosol, it is necessary to study the evolution of the particle number density function, n(m,x,y,z,t), where n(m,x,y,z,t)dm is the number of particles having masses in the range m,dm at downwind position x,y at height z at time t. The evolution equation is:

$$\frac{\partial n(m,x,y,z,t)}{\partial t} + U(z)\frac{\partial n(m,x,y,z,t)}{\partial x} + \frac{\partial}{\partial m}[\Psi(m,s)\, n(m,x,y,z,t)] = \frac{\partial}{\partial z} K(z) \frac{\partial n(m,x,y,z,t)}{\partial z} + \frac{\partial}{\partial y} K(y) \cdot \frac{\partial n(m,x,y,z,t)}{\partial y} + G_z(m)\frac{\partial n(m,x,y,z,t)}{\partial z} + \tfrac{1}{2}\int_0^m b(m-m', m')n(m-m')n(m')dm' - n(m)\int_0^\infty b(m,m')n(m')dm' \quad (1)$$

This equation is coupled to the following conservation equation for the vapor,

given in terms of the saturation ratio, s·

$$\frac{\partial s(x,y,z,t)}{\partial t} + U(z)\frac{\partial s(x,y,z,t)}{\partial x} = \frac{\partial}{\partial z} K(z) \frac{\partial s(x,y,z,t)}{\partial z} + \frac{\partial}{\partial y} K(y) \frac{\partial s(x,y,z,t)}{\partial y} - \frac{1}{c_v} \int_0^\infty \Psi(m,s) n(m,x,y,z,t) dm \quad (2)$$

In these equations, U(z) is the x component of the mean fluid velocity, K(z) and K(y) are the vertical and transverse eddy diffusivities, respectively; $G_z(m)$ is the gravitational settling speed for a particle of mass m, $\Psi(m,s)$ is the condensation/evaporation growth law for a particle, and b(m,m') is the coagulation coefficient. These quantities have been established based on the literature on growth and Brownian coagulation in noncontinuum regimes. Their forms are not detailed here

Eqs. (1) and (2) are subject to appropriate boundary and initial conditions. Explicitly the deposition velocity, $V_d(m)$, of a particle with mass m at ground level is:

$$n(m)\, V_d(m) = K\, \partial n(m)/\partial z, \; z = 0 \quad (3)$$

The deposition term plays an important role in aerosol simulation. It is not difficult to incorporate this condition in general atmospheric models; however, there is considerable uncertainty as to the correct parameterization of the deposition velocity. The other boundary and initial conditions are not detailed here

In the future, photochemical models will be included to provide a source term in Eq. (2); this will involve addition of a nucleation term to Eq. (1). These modifications are in progress.

In aerosol dynamics simulation, particles with radii covering approximately four orders of magnitude (10^{-8} to 10^{-4} cm) must be considered. A logarithmic transformation or particle mass was used to avoid problems associated with this large range:

$$m(J) = m(J_o)\exp(q(J-J_o)) \quad (4)$$

where J is a positive number greater than or equal to J_o, m (J_o) is the mass of a particle starting at J_o, and q is a numerical parameter that can be selected to give equally spaced integer J values. From the following definition of the density function,

$$n(m(J)) = g(J)/qm(J),$$

the growth rate terms in Eqs. (1) and (2) become, respectively:

$$\frac{\partial g(J,t)}{\partial t} = \int_{J_o}^{J_u} dJ'\, b_1(\tilde{J}, J')g(\tilde{J},t)g(J',t) - g(J,t)\int_{J_o}^{\infty} dJ' b(J,J')g(J',t) - \frac{\partial}{\partial J}[\Psi(J)g(J)/qm(J)] \quad (5)$$

and

$$J_u = J - \ln 2/q, \; J \geq 2;$$

$$\tilde{J} = J + (1/q)\ln[1-\exp(q(J'-J))];$$

$$b_1(\tilde{J},J') = (m(J)/m(\tilde{J}))\, b(m-m', m'). \quad (6)$$

The adjustable parameter, q, is a great advantage in "fine tuning" for mass or diameter spacings that increase numerical accuracy.

Numerical Solution Strategies

Coagulation

The coagulation process can be numerically simulated with high accuracy. We chose methods that optimize both accuracy and efficiency. Accuracy was studied in two ways: comparison of numerical simulations with analytical solutions for restricted forms of b(m,m') and comparison with Brownian coagulation through tests for conservation of mass. Cubic spline was used for numerical quadrature and interpolation of the coagulation terms. Gear's method was used for time integration. These comparisons showed that simulation by these methods is accurate and reliable and that errors can be reduced to any desired level.

We found the method of fractional steps to be essential for incorporating the coagulation rate process into any atmospheric dispersion and transport model. A problem associated with this method is the question of where in the time splitting the source and sink terms (i.e., those associated with the aerosol dynamics processes) should be included. In our work, coagulation was included in the dispersion and transport model (Eq. (1)) by the following procedures:

(a) Solve the advection equation by a method such as Fromm's at each vertical level (at each interior collocation point) J_{max} times, where J_{max} is the number of size classes.
(b) Solve the diffusion equation by a method such as orthogonal collocation on finite elements at each grid point in x direction J_{max} times.
(c) Solve the coagulation equation at each grid point

The numerical coagulation results that we obtained prove that the method of fractional steps can accurately simulate aerosol coagulation and transport The desirability of this method can be seen by the fact that a fully implicit scheme for solving a three-dimensional aerosol model requires simultaneous solution of IM·JM·KM·J_{max} equations. For IM = 20 in x direction, JM = 20 in y direction, KM = 10 in z direction, and J_{max} = 30 size classes a simultaneous solution for the 120,000 unknowns would not be feasible; the method of fractional steps removes the necessity of carrying out simultaneous solutions for these unknowns. We have shown that the method of fractional steps is a reliable treatment for multidimensional nonlinear problems, including aerosol coagulation.

Condensation/Evaporation

The condensation/evaporation term in Eq. (1) is deceptively simple. In the J-space formulation, the condensation coefficient, Ψ, varies by ten order of magnitude. The Kelvin effect causes this coefficient to change in sign at a certain particle size for a given supersaturation; it has a negative value for evaporation and a positive value for condensation. These factors make the numerical solution of the condensation/evaporation term difficult.

Difficulties in the numerical solution of first-order hyperbolic equations such as the condensation/evaporation terms are evidenced by the numerous reports of attempts at numerical solutions of similar hyperbolic equations (e.g., advection). Most numerical schemes for hyperbolic equations give rise to numerical dispersion and diffusion. Numerical dispersion, caused by the combination of large phase errors and insufficient short wave damping, manifests itself by the unphysical wakes behind and ahead of the simulated regions of high concentration. Numerical diffusion lowers the peak values of the concentration distribution but increases the values around the peak. A robust numerical scheme for condensation/evaporation should be free of numerical dispersion and should minimize numerical diffusion. Eulerian numerical schemes create numerical dispersion, Lagrangian schemes do not. We combined Eulerian and Lagrangian methods to produce a numerical scheme in which numerical dispersion and diffusion for condensation/evaporation can be reduced to any desired level.

Incorporation of the condensation/evaporation term in an atmospheric dis-

persion and transport model is analogous to incorporation of coagulation; however; coupled equations such as Eqs. (1) and (2) are involved

The simultaneous solution of 10^4 to 10^5 unknowns in a dispersion and transport model by an implicit numerical scheme using a matrix technique (as would occur if a fully implicit solution of Eqs. (1) and (2) were attempted) is difficult. We used the method of fractional steps to incorporate advection, diffusion, and condensation/evaporation by the following procedures:

(1) Solve the advection and diffusion equation J_{max} times, where J_{max} is the number of size classes.
(2) Solve the condensation/evaporation equation by our algorithm at each grid point.
(3) Solve the advection and diffusion equation for s
(4) Calculate the integral term (a source term for vapor due to evaporation) in Eq. (2) and update the saturation ratio at that grid point.

This method of fractional steps decouples Eqs. (1) and (2). If the saturation ratio of the vapor does not change with position—that is, if it is constant—procedures 3 and 4 are unnecessary. The explicit nature of procedure 4 does not pose a problem because the source term is counterbalanced by the dilution effect of advection and diffusion

Recommendations

The general approach and numerical solution schemes outlined here represent one of the first comprehensive studies of aerosol growth in the context of a model of atmospheric dispersion and transport. The modules for coagulation and condensation/evaporation that we developed yield highly accurate simulations of aerosol growth processes in the context of an atmospheric model. These growth modules should be compared with proposed approximate methods for simulation of aerosol dynamics We feel that currently available approximate methods are not adequate for atmospheric simulations on the urban scale. We recommend that new approximate methods be developed.

The numerical schemes for coagulation and condensation/evaporation developed in this project need to be extended to multi-component aerosol dynamics. This will be essential to the study of chemical speciation. Similarly, approximate methods for multi-component aerosols should be developed on the basis of accurate simulation methods.

It is particularly important that field and laboratory aerosol data be obtained with sufficient resolution and accuracy to permit validation of atmospheric aerosol dynamics models. These data should include time and space resolved size distributions and chemical speciations.

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***H. M. Barnes** is the EPA Project Officer (see below).*
The complete report, entitled "Urban Aerosol Modeling," (Order No. PB 84-233 469; Cost: $8.50, subject to change) will be available only from:
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