E-O-2

A STUDY ON SIMULTANEOUS COAGULATION AND DISPERSION IN A CONCENTRATED AEROSOL CLOUD

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Introduction

Aerosols released in high concentrations (nucleation bursts, industrial stack emissions, vehicular exhausts, biomass burning, large scale use of fire works etc.) into the atmosphere undergo simultaneous coagulation and dispersion before becoming a part of background aerosols. For such dense releases, coagulation, which is generally neglected in most atmospheric dispersion models, will have a strong bearing on their concentrations, particle sizes and residence times in the environment. Simons (1986) is one of the few to carry out analytical investigations on problems involving simultaneous coagulation and dispersion equation. He combined the scaling theory with moments method to reduce this equation it to simpler nonlinear form of recombination-dispersion equation. Starting with early work of Jaffe, a semi-analytical technique using diffusion approximation is used for solving ion-recombination equation [e.g. Mayya and Hollander, 1995]. The present study combines these two approaches to obtain approximate analytical formula to estimate survival fraction and effective size of particles. Numerical solutions are discussed to assess the margin of error in analytical results over reasonable set of values.

Formulation of the problem

Let $n(v,r,t)\,dv$ be the number of particles with volumes lying between v and $v+dv$ per unit volume of fluid at position r and time t. Then the general equation governing n which undergoes simultaneous coagulation and diffusion is given by

$$\frac{\partial n}{\partial t} = D\,\nabla^2 n + \frac{1}{2}\int_0^v K(u, v-u)\, n(u)\, n(v-u)\, du - n(v)\int_0^\infty K(u,v)\, n(u)\, du - \lambda(v) n \qquad (1)$$

where, D is the diffusion coefficient of the aerosol particle independent of size. $K(u,v)$ is the coagulation kernel and *(v)* is the removal rate of particles. In the initial phase of aerosol growth, the removal processes are weak compared to coagulation and diffusion, and hence (*v*) is set to zero in the present study. Assuming a solution of self-preserving form (Simons and Simpson, 1988),

$$n(v,r,t) = \frac{\phi(r,t)}{V^2(r,t)}\, g\left(\frac{v}{V(r,t)}\right) \qquad (2)$$

where $\phi(r,t) = N(r,t)V(r,t)$ is the volume fraction, $N(r,t)$ is the number fraction, $V(r,t)$ is the mean particle volume at position r and time t, and g(w) is the scaling function satisfying the relations $\int_0^\infty g(w)\, dw = 1$ and $\int_0^\infty w g(w)\, dw = 1$.

Substituting Eq. (2) in Eq. (1), and taking zeroth and first moments of Eq. (1) w.r.t. *v*:

$$\frac{\partial \phi}{\partial t} = D\,\nabla^2 \phi \qquad (3) \qquad \text{and} \qquad \frac{\partial N}{\partial t} = D\,\nabla^2 N - R\phi^{\alpha}\, N^{2-\alpha} \qquad (4)$$

where, $R = \frac{1}{2}\int_0^\infty \int_0^\infty K(w, w')\, g(w)\, g(w')\, dw\, dw'$. The initial conditions are:

$$\phi(r,0) = N(r,0)V(r,0) \text{ and } N(r,0) = N_0 \frac{1}{\pi^{3/2} b^3} e^{-r^2/b^2},$$

where N_0 is the total number of particles time t=0 and b is the width of the cloud.

Analytical approach using Jaffe's approximation technique: The survival fraction is defined as the fraction of the initial particles surviving at time *t*. To estimate this, Jaffe's approximation technique assumes that the shape of the dispersing plume essentially evolves by diffusion and coagulation affects only the survival fraction *f(t)*. With this, the survival fraction for a spherical cloud for constant kernel (=0), is

$$f(t) = \left[1 + A\left\{1 - \left(1 + 4Dt / b_0^{\,2}\right)^{-1/2}\right\}\right]^{-1}, \text{ where, } A = \frac{K_0\, g_0}{4(2\pi)^{3/2} D b_0} \qquad (6)$$

Numerical methods

a. Solution for Integro-differential equation (1)

The diffusion part of Eq. (1) in spherical co-ordinates is solved by using finite difference technique and resulting matrix equation is solved by the Thomas algorithm. The boundary of the problem domain has been kept large enough to encompass the expanding cloud. The coagulation part of Eq.(1) is discretized and solved by nodal method (Prakash et al, 2003). The particles are distributed in two-dimensional grids, one in physical-space and other in the size domain. A program in C language is written to solve the two processes, diffusion and coagulation, independently at each time step (Δt).

b. Solution for the Simon-Simpson equations

Eq.(4) in spherical co-ordinates (assuming azimuthal symmetry) for the special case of α= 0 is given by,

$$\frac{\partial N}{\partial t} = D\frac{\partial^2 N}{\partial r^2} + \frac{2D}{r}\frac{\partial N}{\partial r} - \frac{K_0}{2} N^2 \qquad (9)$$

This equation is numerically solved by finite difference schemes.

Results and Discussions

The study has been carried out for two different constant coagulation rates. The results are shown in Fig.1. In both the cases, the behaviour of numerical solutions is similar. The error between numerical and analytical solution obtained by Jaffe-Simon-Simpson approximation is 4.9% and it increases as the survival fraction decreases. This deviation is mainly because of Jaffe's approximation. In fact, Simon-Simpson's approximation matches with exact numerical solution. The error further increases to 29% when the coagulation rate is increased.

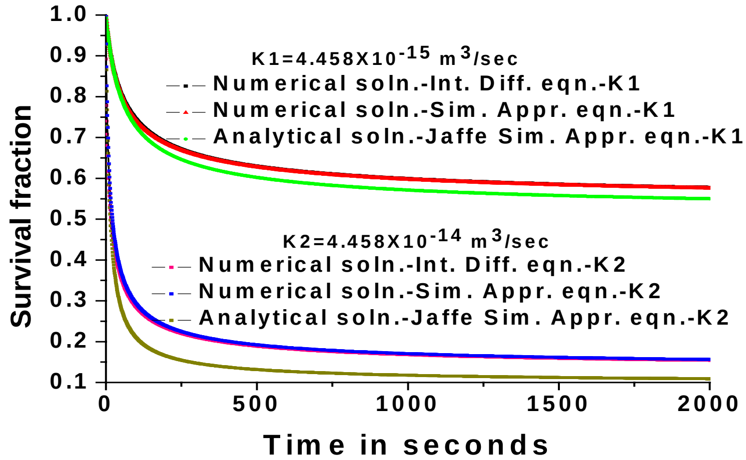

Fig. 1

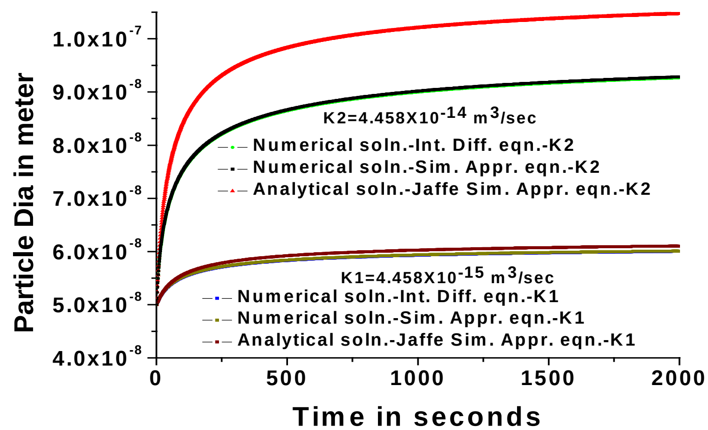

Fig. 2

The variation of particle size with time is shown in figure 2. The initial size of the mono disperse particles is 50 nm, finally the average particle size attains a constant value of 60 nm in one case (for coagulation constant $K_1 = 4.458 \times 10^{-15}$ m^3/sec) and 93.4 nm in the other (for coagulation constant $K_2 = 4.458 \times 10^{-14}$ m^3/sec).

Conclusion

Although a purely coagulating aerosol decays completely with time, that with diffusion eventually survives coagulation. This knowledge is useful for predicting the fraction of particles injected into the open environment from puff releases. The analytical formula although approximate describes the qualitative features fairly well. It has about 5% error at low concentrations and has about 30% error at high concentrations for predicting the survival fractions.

References

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