Technical note

Coagulation in a spatially inhomogeneous plume: Formation of bimodal size distribution

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ABSTRACT

Aerosol systems are more often spatially heterogeneous than homogeneous due to various factors such as turbulence in the atmosphere, flow field in a pipe, and varying generation rates at the source. As a general result, we demonstrate that an initially monodisperse and spatially heterogeneous aerosol system evolves into a bimodal size distribution purely by coagulation. The spatial inhomogeneity in the particle number concentration initiates differential coagulation rates which lead to a distribution with larger size modes in regions with higher concentration. When averaged over space, this would appear as a bimodal size distribution. We show this effect through a free-molecular coagulation model for a spatially heterogeneous system combined with the scaling theory of self-preserving distributions. It is found that sharper the occurrence of spatial heterogeneity, more pronounced is the bimodal effect. The study clearly demonstrates spatial hetero-geneity as an additional factor for the origin of bimodality in aerosols.

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1. Introduction

Atmospheric aerosol size distributions of mass and number are normally multimodal as a result of new particle formation and various dynamical processes such as coagulation, condensation, and removal mechanisms taking place in the atmosphere (Seinfeld & Pandis, 2006). For example, most of the atmospheric mass or volume distributions are represented by accumulation and coarse modes. Accumulation mode ($\sim$(0.1–2) μm) is a result of primary emissions, condensation and coagulation mechanisms while the coarse mode ($\sim$(2–50) μm) particles are formed by the direct injection of particles such as sea salt sprays from the oceans, windblown dust, and other mechanical processes. Similarly, the number size distribution of aerosols in the atmosphere consists of modes at sizes < 10 nm, (10–100) nm, and > 100 nm termed as nucleation, Aitken and coarsemodes respectively. Although the aerosol processes combined altogether are responsible for the formation of multimodal distributions in the atmosphere, in some special cases, individual aerosol processes such as nucleation or coagulation also evolves the number size distribution to bimodal. One such example is the formation of a two-modal distribution during the nucleation growth process in an atmosphere with saturation ratio fluctuations in space and time (Kulmala et al., 1997). Similarly, the case of continuous source emission and coagulation in well mixed closed reactors and chambers also lead to the formation of bimodal size distribution (Landgrebe & Pratsinis, 1989; Seipenbusch et al., 2008; Anand et al., 2012).

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Another example widely discussed in the literature is the effect of differential coagulation rate due to spatial inhomogeneity of particle number concentration that leads to bimodal distribution in the atmosphere. The inhomogeneous aerosol concentration in the atmosphere is formed by various mechanisms such as turbulence, and the details of formation are discussed in Elperin et al. (2000). This spatial inhomogeneity of particle concentration introduces the differential coagulation rate or acceleration of coagulation growth (Kasper, 1984; Khain et al., 2000). Voloshuk and Sedunov (1975) noted the acceleration of gravitational coagulation growth of cloud droplets in the regions of inhomogeneous concentration. Pinsky & Khain (1997) also demonstrated in their work the influence of inhomogeneous drop concentration in the cloud on droplet spectrum broadening. However, simple models are required to understand the effect of spatial heterogeneity in a coagulating system that leads to the evolution of bimodal particle size distribution. The objective of the present study is to develop an analytical model based on scaling theory to demonstrate these effects.

2. Problem formulation

Let us consider a circular combustion source of radius a, (say, a circular tip of burning wood) kept in a flow field having small advection velocity normal to the source plane. In the earlier treatments (Anand & Mayya, 2009, 2011), we assumed Gaussian profiles for the spatial distributions under an implicit understanding that the diffusion process will eventually render all distributions into a Gaussian shape, after sufficient time. When particles are nucleated from finite size sources and diffusion is weak, their concentration will be highest at the centre of the source and would decrease rapidly at the periphery. Then the spatial number concentration profile will have the shape of a nearly flat top near the source. This shape represents high and uniform concentration in the central regions and a rapid fall near the periphery. Although one may construct a large number of mathematical functions having this shape, for simplicity the following form is chosen:

$$N(r,0) = N_0(r) = N_0(0)\mathrm{erfc}\left(\frac{r-a}{s}\right) \tag{1}$$

where $N_0(0)$ is the number concentration at the centre of the circle, and "s" is the width of the spatial zone across which the concentration falls rapidly to zero. For $s \ll a$, the concentration will remain uniform in the central region and fall rapidly at the edge of the source. As noted in the context of studying the coagulation induced flattening (Anand & Mayya, 2011), coagulation will be most pronounced at the centre of the source due to spatial inhomogeneity, giving rise to larger particles after a short time, as compared to those at the periphery. This differential coagulation rate will result in a bimodal distribution of particle sizes. We illustrate the possibility of this phenomenon below by a simple model that captures the essence of spatial inhomogeneity and coagulation.

3. Analytical model and solution

Let us assume that the initial size distribution of particles is monodisperse having volumes, u_0. Due to the flow field, these particles will be advected along say, X- direction normal to the plane of the source. The spatial coordinate (x) may be translated into time coordinate (t) through the relationship $t = x/v$, where v is the advection velocity. Let us assume that there is no lateral diffusion. In other words, we are going to consider only an early time process of a high concentration aerosol whose coagulational time-scale is far smaller than the diffusional time-scale near the source region. If the lateral diffusion is ignored, then the particles at different radial points will coagulate at different rates dictated by their concentration, $N_0(r)$. Let us denote their volume (u) distribution at a radial distance r from the centre-line as $n(r,u,t)$. If the initial concentrations are very large, then the particle volume distribution will attain self-preserving form (similarity form) having r-dependent characteristic parameters. From the standard similarity theory, this may be expressed as

$$n(u,r,t) = \frac{N^2(r,t)}{u_0 N_0(r)} \, g\left\{\frac{u N(r,t)}{u_0 N_0(r)}\right\} \tag{2}$$

where $N(r,t)$ is the size integrated number concentration at r, t

$$N(r,t) = \int_0^\infty n(u,r,t)du \tag{3}$$

and $g(\eta)$ is the scaling function having the property (Friedlander, 2000)

$$\int_0^\infty g(\eta)d\eta = 1 \quad \& \quad \int_0^\infty \eta g(\eta)d\eta = 1 \tag{4}$$

Since we are considering nano-particles, their coagulation process may be adequately described by a homogeneous, free-molecular kernel. The homogeneity index (α) of the kernel will vary depending upon the fractal dimension (d_f) of the particles ($1/6 < \alpha < 1/2$). $\alpha = 1/6$ corresponds to compact clusters ($d_f = 3$) and $\alpha = 1/2$ corresponds to clusters with fractal dimension ≤ 2. Upon solving the rate equation for the total number concentration derived from similarity theory for the