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The structure of nanoparticle nucleation in planar jets

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ABSTRACT

The effects of large-scale mixing and vapor concentration on homogeneous nucleation rates are investigated via direct numerical simulation of dibutyl-phthalate (DBP) nucleation during cooling and mixing in three-dimensional planar jets. In the simulated cases, a heated jet doped with DBP issues into a co-flow of room-temperature air. As the two streams mix, the DBP vapor becomes highly supersaturated and particles are formed by nucleation. This particle formation takes place in the absence of condensation or coagulation. The simulation results provide a demonstration of how nucleation takes place in narrow regions where molecular mixing of the two streams occurs. When maximum nucleation rates occur in conditions where the nucleation rates are sensitive to ambient conditions, islands of nucleation form. There are two possible nucleation events: initial shear layer nucleation, and later nucleation in coherent structures or eddies generated by the velocity difference between the jet and the co-flow. A scatter plot diagram of observed dilution paths in temperature versus condensable vapor concentration space where nucleation rates are superimposed is shown to be a convenient tool for analyzing nucleation events. Convection by large-scale eddies gradually spreads the range of mixing paths in this space toward higher nucleation rates. The results also show that boundary conditions, including inlet concentration and velocity ratio, have both qualitative and quantitative effects on particle nucleation.

1. Introduction

In various processes, homogeneous nucleation that occurs in turbulent flow is a key issue in determining the properties of resulting aerosol. Examples can be found in nanoparticle synthesis, exhaust gas emissions, aerosol dilution and sampling techniques as well as in atmospheric processes. As nucleation rates are very sensitive to small changes in local ambient conditions, turbulence-induced fluctuations are an important consideration in the modeling of these phenomena. A case that is often encountered in practice is that of a hot jet or a plume that contains condensable vapors issuing into colder ambient air. Turbulent eddies are involved in transporting the vapors into cooler regions where they become supersaturated enough for homogeneous nucleation to occur. The overall mixing process involves advective transport by eddies and molecular mixing at the edges of these eddies. Roughly speaking, larger eddies are mainly responsible for the overall mixing in the large scale, while the coupled action of small-scale eddies and molecular diffusion is responsible for the micro-level

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mixing. In this paper, we focus on homogeneous nucleation that occurs when temperature and saturation pressure decrease as a result of turbulent mixing. Other important pathways for particle formation are nucleation due to chemical reactions, and nucleation due to a pressure drop in supersonic flow. Examples of nucleation by chemical reactions are titanium dioxide particle formation (Moody & Collins, 2003; Wang & Garrick, 2006; Xiong & Pratsinis, 1993) and soot particle inception (Kennedy, 1997; Zimberg et al., 1998). Converging/diverging nozzles provide an example of pressure drop induced nucleation (Bayazitoglu et al., 1996; Wegener & Pouring, 1964).

There are a number of studies, both experimental and theoretical, on the effect of turbulence on homogeneous nucleation. Lesniewski & Friendlander (1998) carried out a series of experiments on dibutyl-phthalate (DBP) nucleation and growth in turbulent round jet shear layers. In the analysis of their experiments, Lesniewski & Friedlander (1995, 1998) took advantage of the fact that, in round jet flows, RMS fluctuations, probability density functions (PDFs) and mean values of temperature and gaseous species concentration are known to depend only on the ratio of the radial position in the shear layer to the axial position. The PDFs do not have a Reynolds number dependence. In order to render the problem amenable to a simple computational analysis, they assumed that heat and the condensable vapor diffuse in a similar fashion, i.e. that the Lewis number *Le* is equal to one. In addition, nucleation was assumed to be limited to the shear layer. With these assumptions, shear layer nucleation was shown to be proportional to d_{jet}^3, where d_{jet} is the diameter of the jet, and to be independent of the jet velocity. With high enough DBP concentration, nucleation outside the shear layer was also observed in the experiments. In addition to the fundamental studies by Lesniewski & Friendlander (1998) and preceding studies of similar type summarized in their paper, there are a number of more applied studies on nucleation and aerosol dynamics in exhaust plumes. The simulations of Wu & Menon (2001) are the most relevant to this work. They simulated aerosol particle formation via binary nucleation of sulfuric acid–water vapor system using a linear eddy model. In the linear eddy model, a one-dimensional grid that is perpendicular to the plume is marched and stretched along the plume and turbulent mixing is represented with a series of stochastic mappings in this space. The peak number density was shown to be under-predicted by about 40% when micromixing was not included in the simulations. More recently, Housiadas et al. (2004) showed using data from a LES study of the planetary boundary layer that the effect of turbulence on the overall atmospheric nucleation rate in a sulfuric acid–water vapor system is not very large in the cases they considered. Also recently, Shaw (2004) demonstrated with asymptotic analysis that mean particle formation rates are essentially dependent only on the value of the PDF at which the exponential of the nucleation rate expression attains maximum.

A computational fluid dynamics technique called direct numerical simulation (DNS) is used to capture the mixing process in a model-free manner (Givi, 1989). Both the convective/advective action of the eddies and the molecular mixing processes are considered explicitly. No assumptions regarding flow structure or the nature of the molecular mixing process need to be made. Instead, the Navier–Stokes equations and other transport equations are solved down to the smallest scales. The methodology is general and not restricted to a single flow configuration. The disadvantage of DNS is the large amount of computational resources it requires. This limits the applicability of DNS to relatively simple cases, and to relatively low levels of turbulence. Even so, these cases are valuable as a mixing process, they contain most of the same salient features that exist in more complex flows. This makes it possible to infer or test models or theories of nucleation in turbulent flow. The visualization of the results provides an instructive and easily understandable insight into the fine-structure details of the nucleation process. A similar degree of information is not available from today's experimental techniques. The framework of DNS allows, in a relatively straightforward manner, the inclusion of other phenomena such as heterogeneous condensation, surface reactions, coagulation and intra-particle processes. The DNS methodology is potentially even more beneficial in dealing with the effects of micro-mixing when some of these mechanisms are significant as well, and coupled with each other, since such cases are beyond the scope of simplified models.

In this work, we analyze nucleation in the eddies formed by three-dimensional, cooling planar jet flows. The flow and the transport of vapor and particles are modeled with a three-dimensional DNS code. This configuration and modeling approach has been used in a number of University of Minnesota studies to simulate reacting flows (Das & Garrick, 2010; Garrick & Khakpour, 2004; Miller & Garrick, 2004). In the studies reported in this paper, homogeneous nucleation is considered to be the sole aerosol transformation mechanism. The goal of this work is to provide insights into homogeneous nucleation under various mixing intensities and its DNS modeling as well as to act as a stepping stone toward turbulent mixing cases that involve other phenomena as well. It is not the intention of this work to simulate the full formation and growth process as this has been considered previously (Das & Garrick, 2010; Garrick & Wang, 2011; Loeffler et al., 2011; Wang & Garrick, 2005). Dibutyl-phthalate (DBP) is adopted as an example species because it has been used widely in experimental nucleation research, modeling studies and as a large molecule it is representative of heavy organic compounds that are important in fine particle emission issues.

2. Formulation

2.1. Fluid field

The flow and temperature fields are represented by the Navier–Stokes equations for compressible non-isothermal flow:

$$\frac{\partial \rho}{\partial t} + \frac{\partial \rho u_j}{\partial x_j} = 0, \tag{1}$$