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The effect of mixing rates on the formation and growth of condensation aerosols in a model stagnation flow

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ABSTRACT

A steady, laminar stagnation flow configuration is adopted to investigate numerically the interaction between condensing aerosol particles and gas-phase transport across a canonical mixing layer. The mixing rates are varied by adjusting the velocity and length scales of the stagnation flow parametrically. The effect of mixing rates on particle concentration, polydispersity, and mean droplet diameter is explored and discussed. This numerical study reveals a complex response of the aerosol to varying flow times. Depending on the flow time, the variation of the particle concentration in response to varying mixing rates falls into one of the two regimes. For fast mixing rates, the number density and volume fraction of the condensing particles increase with residence time (nucleation regime). On the contrary, for low mixing rates, number density decreases with residence time and volume fraction reaches a plateau (condensation regime). It is shown that vapor scavenging by the aerosol phase is key to explaining the transition between these two regimes. The results reported here are general and illustrate genuine features of the evolution of aerosols forming by condensation of supersaturated vapor from heat and mass transport across mixing layers.

1. Introduction

The formation of particles from supersaturated vapor is a fundamental process in aerosol-laden flows in nature and technological applications. Examples in environmental flows include clouds (Pruppacher & Klett, 2010) and atmospheric aerosols (McMurry, 2000). In the materials processing industry, the condensation of a precursor vapor (e.g. a metallic chloride) into monodisperse liquid droplets by mixing with a cold inert gas is a preliminary step in the production of advanced powders (Kodas & Hampden-Smith, 1998).

In many of these situations, the vapor concentration and temperature fields are not homogeneous in space. For example, a hot vapor stream may combine with a cold gas in laminar or turbulent jets, wakes, or mixing layers (Lesniewski & Friedlander, 1998). Local vapor supersaturation and droplet condensation therefore occur as a consequence of fluid, heat, and mass transport across mixing layers, which are thin compared to the largest scales of the flow. The nucleation and growth rates of particles are highly nonlinear and very sensitive to local temperature and vapor concentration. Thus, the yield and particle size distribution of the condensing particles depends critically on the coupling between the aerosol

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microphysical processes and heat and mass transport in the spatially inhomogeneous mixing field (Clement, 1985; Nguyen et al., 1987; Pesthy et al., 1983).

The purpose of the present study is to investigate the interaction between condensing aerosols and transport in laminar flows. Our approach differs from previous studies in its choice of the hydrodynamic configuration: a laminar mixing layer established across a stagnation flow. As will be shown, the stagnation flow is a canonical flow configuration, which allows for a unique parametrization of the response of the condensing aerosol to varying rates of heat and mass transfer.

A detailed theory of the formation of aerosols by condensation in inhomogeneous flows with heat and mass transfer has been developed mostly in the context of laminar flow condensers. In a flow condenser, vapor flows through a tubular reactor where the temperature of the gas is lowered by conduction to the walls or by injection of a cold gas, causing the formation of particles (Nguyen et al., 1987; Pesthy et al., 1983). Alternative configurations include laminar coaxial jets (Brock et al., 1986) and gas-flow diffusion chambers (Anisimov & Cherevko, 1985). In these experiments, reported values for the particle number density lie between 10^2 and 10^8 cm^{-3} and particle diameters are in the range 1–10 μm.

Nguyen et al. (1987) investigate the formation of aerosol particles by homogeneous and heterogeneous nucleation of dibutyl phthalate in dry nitrogen carrier gas inside a condenser tube. A range of saturation temperatures, vapor concentrations, and seed properties are considered. They observe that the particle number density increases by several orders of magnitude due to an increase of only a few percent in the vapor concentration, while the size of the liquid droplets does not change significantly. Brock et al. (1986) perform an experimental study for the formation of condensation aerosols of phthalate esters in coaxial laminar jets. They note that as the particle number density increases downstream of the inception zone, the vapor condenses on existing particles and nucleation rates decrease due to vapor depletion. Similar effects are discussed by Pesthy et al. (1983), Pratsinis (1988), and Mikheev et al. (2000) in the context of condenser tubes.

Numerical approaches for condensing aerosols with heat and mass transfer have been proposed in the literature (Pesthy et al., 1983; Phanse & Pratsinis, 1989; Pratsinis, 1988; Pyykönen & Jokiniemi, 2000). Pesthy et al. (1983) propose a detailed model for the aerosol phase and solve the general dynamic equation along each streamline in a condenser tube. They consider nucleation and growth processes only and neglect coagulation because the particle concentration is low. They repeat the calculations for the classical Becker–Döring and the Lothe–Pound nucleation theories (Springer, 1978), with the latter providing a better comparison with experiments. Pratsinis (1988) formulates a moment method with closure based on a lognormal particle size distribution. Nucleation, condensation, and coagulation processes are included, together with heat and mass coupling to the gas-phase. The method is applied successfully to the simulation of aerosol generation in a model laminar flow condenser tube (Phanse & Pratsinis, 1989). Pyykönen & Jokiniemi (2000) develop and implement sectional approaches for the dynamics of aerosols in laminar flow condensers.

In this work, the flow configuration consists of two streams flowing from opposite directions, so that a steady mixing layer is established across a stagnation plane. The opposed flow belongs to the broad category of Hiemenz-type stagnation flows. One stream is hot and saturated with vapor, while the other is cold and dry. Large saturation ratios occur across the mixing layer, causing liquid droplets to form and grow, mostly at the stagnation plane. The temperature and composition at the inlets can be adjusted to obtain the desired distribution of saturation ratio in the mixing layer. In keeping with its common usage in experiments, the physicochemical properties of the condensing model vapor used in this work are those of the low vapor pressure compound dibutyl phthalate (DBP).

In the opposed flow configuration considered here, there is only one velocity and one length reference scale. These are the fluid velocity at the inlets and the separation distance between the two nozzles. Thus, the opposed flow is characterized by a unique, well-defined flow time scale, the inverse of a global strain rate, which can be varied by adjusting the ratio between the inlet velocity and the nozzle separation distance (Chapman & Bauer, 1975). This feature is exploited in this work to vary the hydrodynamic flow time parametrically, thereby changing the rates of heat and mass transfer and allowing the response of the condensing aerosol to be observed.

From the perspective of investigating condensing aerosols experimentally, the opposed flow configuration may possess some advantageous features. Firstly, steady mixing and aerosol condensation is achieved within a confined region of the flow away from walls and foreign surfaces, thereby simplifying the management of heat and mass transfer. Secondly, the external flow field and aerosol particles are accessible both by optical diagnostics and probe-based aerosol sampling techniques. Thirdly, the opposed flow configuration has been extensively studied experimentally and numerically in the context of premixed and non-premixed laminar flames (Law, 2006), so that many results are available. Finally, the velocity and scalar fields across the opposed flow are amenable to an inexpensive, quasi-one-dimensional representation, which facilitates the development of computational models and the comparison between numerical and experimental results (Kee et al., 1989).

The paper is organized as follows. In Section 2, the details of the configuration, physical models, and numerical methods are explained. Results for the velocity, vapor concentration, temperature, and aerosol properties are discussed in Section 3. A comprehensive parametric study is included in the analysis. Quantitative results for number density, volume fraction, and droplet diameter are discussed. Conclusions are presented in Section 4.

2. Configuration, methods, and models

The geometry of the flow considered in this work is shown in Fig. 1. The configuration is two-dimensional and axisymmetric. The fluid is introduced with uniform and equal velocity u_0 from two nozzles of infinite diameter separated by a distance $2L$. Let x be the axial coordinate placed along the axis of the geometry and r be the radial coordinate normal to it.