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Simulation of aerosol nucleation and growth in a turbulent mixing layer

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A large-scale simulation of aerosol nucleation and growth in a turbulent mixing layer is performed and analyzed with the aim of elucidating the key processes involved. A cold gaseous stream is mixed with a hot stream of vapor, nanometer sized droplets nucleate as the vapor becomes supersaturated, and subsequently grow as more vapor condenses on their surface. All length and time scales of fluid motion and mixing are resolved and the quadrature method of moments is used to describe the dynamics of the condensing, non-inertial droplets. The results show that a region of high nucleation rate is located near the cold, dry stream, while particles undergo intense growth via condensation on the hot, humid vapor side. Supersaturation and residence times are such that number densities are low and neither coagulation nor vapor scavenging due to condensation are significant. The difference in Schmidt numbers of aerosol particles (approximated as infinity) and temperature and vapor (near unity) causes a drift of the aerosol particles in scalar space and contributes to a large scatter in the conditional statistics of aerosol quantities. The spatial distribution of the aerosol reveals high volume fraction on the hot side of the mixing layer. This distribution is due to drift against the mean and is related to turbulent mixing, which displaces particles from the nucleation region (cold side) into the growth region (hot side). Such a mechanism is absent in laminar flows and is a distinct feature of turbulent condensing aerosols.

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I. INTRODUCTION

The nucleation and growth of particles from supersaturated vapor is a key process in aerosol-laden flows, and occurs both in nature^{1–3} and industrial applications.⁴ In realistic flow configurations, the vapor concentration and temperature fields are not uniform in space and time, and an inhomogeneous spatial distribution of saturation ratios results from heat and mass transport. This mechanism is often referred to as “supersaturation by mixing.” Alternative mechanisms for the formation of aerosols include chemical reactions, radiative heat loss, surface cooling, and rapid (adiabatic) expansion of the gaseous mixture.

As established by classical theories,⁵ the rates of particle nucleation and growth are highly nonlinear and depend strongly on saturation ratio. For example, for the low vapor pressure compound dibutyl phthalate (DBP) at 300 K and saturation ratio of 1200, a 18% reduction in the saturation ratio causes the nucleation rate to decrease by an order of magnitude. Consequently, issues related to the coupling between heat and mass transport and microphysical processes of nucleation and growth are important in determining the properties of the condensing aerosol.^{6–8} A detailed theory of the formation of aerosols by condensation in spatially inhomogeneous flows exists for laminar configurations, such as laminar flow condensers,^{7,8} laminar coaxial jets,⁹ and gas-flow diffusion chambers.¹⁰ On the other hand, most of the natural and industrial aerosols formed by mixing occur

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in the turbulent flow regime, which is characterized by unsteady, three-dimensional, and intermittent velocity and scalar fields.¹¹

Given the highly nonlinear dependence of the formation and growth rates on temperature and vapor concentration, turbulent fluctuations in the mixing fields complicate the interaction between fluid transport and aerosol processes beyond simple scaling considerations. It is well recognized^{4,12,13} that turbulent flow alters the product aerosol compared to the laminar mixing case, even though the composition and temperature of the inlet streams are unchanged. At present, a complete description of the mechanisms underlying the interaction between aerosol and turbulence in realistic flow configurations is lacking. In broad terms, the goal of the study reported here is to improve our understanding of the key processes involved in the formation and growth of a condensing aerosol in turbulent flows. This goal is achieved via a numerical simulation of nucleation and condensation in a canonical spatially developing, turbulent flow featuring the mixing of two streams with mean shear. The stream composition and temperature are selected to mimic those in a well-known experiment for which data are available.^{14,15}

There exist a number of experimental studies on the nucleation and growth of condensing aerosols in turbulent flows with mixing. Turbulent jets of hot condensable vapor mixing with ambient air are used by Amelin¹⁶ and by Higuchi and O'Konski¹⁷ to test the Becker-Döring theory⁵ of homogeneous nucleation. Detailed measurements of velocity, temperature, and vapor concentration in the near field, transitional, and fully developed regions of a turbulent round jet of vapor are reported by Hidy and Friedlander¹⁸ together with a discussion of the spatial distribution of the mean saturation ratio. The authors conclude that there exists a near-field region of supersaturated vapor without appreciable aerosol mass, where nucleation occurs. Further downstream, newly formed particles act as nuclei for condensation in the regions of the jet where the saturation ratio exceeds unity. The scaling behavior of the condensing aerosol observed by Hidy and Friedlander¹⁸ agrees with a previous theoretical analysis by Friedlander,¹⁹ who identifies a set of dimensionless groups controlling the size distribution of the molecular clusters formed by mixing according to the Becker-Döring theory. Among these, a Damköhler number for condensation is proposed.

The well-known experiment of Lesniewski and Friedlander^{14,15,20} remains the most comprehensive attempt to formulate a general theory for particle nucleation and growth in turbulent flows. The experiment of Lesniewski and Friedlander¹⁵ consists of a hot nitrogen stream at 413 K with DBP vapor (estimated by the authors to be in the range 100–500 ppm by volume) issuing into a coflow of slowly moving cold, dry air (290–295 K), thereby forming a turbulent round jet with condensing droplets. Under the assumption of unity Lewis number, these conditions lead to peak saturation ratios from 250 (in the case of 100 ppm and 295 K) to 1230 (in the case of 500 ppm and 290 K). These values are only estimates and the difference in the diffusion coefficients of heat and mass may lead to significantly lower levels of saturation.⁶ The Reynolds number based on the bulk velocity and round jet diameter varies between 4000 and 7000.

Lesniewski and Friedlander^{14,15,20} restrict their analysis to configurations with moderate nucleation rates (in the range 10^4 – $10^8/\text{cm}^3 \text{ s}$) and low concentrations of particles (less than $10^4/\text{cm}^3$), so that coagulation rates are negligible and the aerosol evolution is nucleation-controlled. At the lowest levels of DBP vapor concentrations at the inlet (less than 350 ppm in mole fraction), the authors report that the mean number density along the jet centerline in the fully-developed region of the jet decays as $\langle N \rangle \propto 1/x$, where x is the streamwise coordinate. This rate of decay is consistent with the entrainment process in a round jet in the absence of nucleation. The authors^{14,15} conclude that particles are formed by homogeneous nucleation in the near-field of the jet and further nucleation downstream is suppressed by a rapid decrease in saturation ratio caused by the entrainment of dry coflow air. For higher vapor mole fractions, nucleation occurs past the near field throughout the jet and important deviations from the $1/x$ scaling are reported. These modifications in the aerosol evolution are reflected in the particle size distributions sampled at various streamwise locations. The size distribution displays one, two, and eventually three modes at increasing diameters as the vapor mole fraction increases.

Although essential, the experimental characterization of turbulent flows with condensing aerosols is challenging and seldom quantitative due to uncertainties in the nucleation kinetics, physical properties of the vapor (e.g., surface tension), and insufficient information on the turbulent

fields.¹⁵ Most importantly, the concurrent, spatially resolved measurement of instantaneous fields of velocity, temperature, vapor concentration, and aerosol quantities (e.g., number density, volume fraction, and mean droplet diameter) is simply impossible with available experimental techniques. Joint statistics of the gas-phase and aerosol quantities are most valuable in elucidating the interplay between turbulent mixing and microphysical processes. In this context, the role of numerical simulations of aerosol formation in turbulent flows is invaluable and fills a large knowledge gap left by incomplete experimental databases.

Direct numerical simulation (DNS) for the momentum and gas-phase scalars and a nodal approach for aerosol dynamics²¹ have been employed successfully in the study of the effects of turbulent fluctuations on the evolution of aerosols.^{22–25} These papers detail the formation and growth of titanium dioxide nanoparticles in turbulent planar jets,²² and zinc nanoparticle dynamics (nucleation and coagulation without condensation) in turbulent round jets.²³ In these studies, the effects of turbulence on nanoparticle growth are investigated by comparing the formation and growth rates from DNS with those obtained from filtered quantities. In agreement with the theoretical analysis of Lesniewski and Friedlander,²⁰ neglecting the unresolved or subfilter scalar fluctuations leads to a significant overestimation of the mean formation and growth rates. A detailed analysis of the effects of unresolved scalar fluctuations on homogeneous nucleation in the context of large-eddy simulation is provided by Murfield and Garrick.²⁴

In the present work, a large-scale simulation of aerosol nucleation and growth in a turbulent mixing layer is performed and analyzed with the aim of elucidating the key processes involved. The spatially evolving mixing layer is a canonical turbulent flow configuration for which numerous results on the large and small scale statistics of velocity and scalars are readily available.^{26–30} The physical properties of the condensing vapor are those of dibutyl phthalate due to its low vapor pressure at near ambient temperatures and widespread usage in experiments. The temperature and stream composition are akin to those in Refs. 14 and 15.

The present simulation resolves all length and time scales of momentum and gas-phase scalars fully as in a DNS of turbulent flow, while the quadrature method of moments^{31,32} (QMOM) is used to model the dynamics of the condensing, non-inertial liquid droplets. The QMOM model for the aerosol phase is coupled to the gas-phase scalars fully. Among the various approaches available for aerosols,^{21,33,34} moment methods are the most suitable for use in large-scale simulations of three-dimensional turbulent flows. A Lagrangian particle scheme³⁵ is used to solve the moment equations. The Lagrangian particle scheme circumvents issues connected to the loss of the realizability of the moments common to conventional Eulerian advection schemes.³⁶

The work presented here makes three important contributions to the field of aerosol formation and growth in turbulent flows. First, we investigate and report on aerosol processes occurring in the fully-developed region of a canonical turbulent mixing layer. A moderately high Reynolds number flow is accomplished by considering a large computational domain, which enables the growth of the mixing layer, so that statistics are accumulated near the streamwise far field boundary. We estimate that, in the fully-developed region of the flow where the statistical analysis is carried out, the Reynolds number based on the Taylor microscale is $Re_\lambda = 110$.

Second, an important contribution of this work lies in the detailed analysis of the effects of the high Schmidt number of droplets on growth. In studies of aerosol condensation in turbulent flows, it is seldom emphasized that the diffusion coefficient of aerosol droplets is several orders of magnitude lower than that associated with gas-phase species. The difference in diffusion rates between droplets and the gas-phase causes droplets to drift in the concentration and temperature spaces, with important effects on growth rates. The small scale statistics of turbulent mixing affect this process strongly, thereby influencing the growth rates of droplets. This and related issues are well-known to the combustion research community interested in sooting flames,^{35,37,38,68,69} but remain largely overlooked in the literature on aerosol condensation in inhomogeneous flows. This important mechanism is discussed in detail using Lagrangian statistics.

Third, the data generated are analysed in great depth and the statistics of velocity, mixing, and aerosol quantities are shown and discussed with the aim of providing a reference database, which is made available to the aerosol and fluid mechanics communities. The present numerical simulation offers complete and concurrent data on gas-phase and aerosol scalars of interest to model

development. The data generated, which may not be obtained experimentally, support the formulation of novel closure strategies for Reynolds Averaged Navier Stokes (RANS) and large-eddy simulation (LES) of aerosol formation and growth in the turbulent flow regime.

The paper is organized as follows. In Sec. II, the details of models and methods employed to describe the fluid dynamics and aerosol processes are provided. Results are discussed in Sec. III. Conclusions are presented in Sec. IV.

II. MODELS AND METHODS

A. Flow configuration

Nucleation and growth of droplets due to supersaturation induced by mixing are simulated in a spatially developing turbulent mixing layer. The flow configuration is similar to that considered in recent studies,^{27,28} where a detailed analysis of the small scale statistics of the velocity and passive scalar fields is given.

The streamwise velocities are $u_1 = 15$ and $u_2 = 5$ m/s for the top and bottom streams, giving $\Delta U = u_1 - u_2 = 10$ m/s and $U_c = (u_1 + u_2)/2 = 10$ m/s. At the inlet, the streamwise velocity profile is imposed by combining two Blasius laminar boundary layers. Random velocity fluctuations $0.4\omega_t u$, where ω_t is uniformly distributed with zero mean and unit variance and u is the streamwise inlet velocity, are superimposed on the streamwise velocity profile at the inlet. As a result of this forcing, the Kelvin-Helmholtz instability arises within a short distance downstream of the inlet, i.e., $x^* = x/\delta_{\theta,0} \approx 60$, where $\delta_{\theta,0} = 0.201$ mm is the momentum thickness at the inlet. The computational domain is $L_x^* \times L_y^* \times L_z^* = L_x/\delta_{\theta,0} \times L_y/\delta_{\theta,0} \times L_z/\delta_{\theta,0} = 640 \times 512 \times 214$, where the streamwise, crosswise, and spanwise directions are indicated with x , y , and z , respectively. The momentum thickness grows to $\delta_\theta = 3.596$ mm near the outlet of the domain at $x = L_x$. The Reynolds number based on ΔU and the momentum thickness at the inlet (outlet) is 111 (1998).

A free convective outflow³⁹ condition is used at the outlet in the streamwise x direction. The boundary conditions are free slip in the crosswise direction y and periodic in the spanwise direction z . The free slip condition is applied by imposing a zero crosswise velocity component at the crosswise boundaries. The normal derivatives of the streamwise and spanwise velocity components are set to zero. The domain in the crosswise direction y is large enough to avoid any important issues related to the free slip boundary conditions.

The top stream consists of hot nitrogen saturated with DBP vapor and the bottom stream is dry, cold nitrogen. At the inlet, a hyperbolic tangent profile is used for the temperature, i.e. $T(y^*) = (T_1 + T_2)/2 + (T_1 - T_2)\tanh(0.2y^*)/2$, where $y^* = y/\delta_{\theta,0}$, and $T_1 = 410$ and $T_2 = 290$ K are the temperature in the fast (top) and slow (bottom) streams, respectively. The profile for the vapor mass fraction is defined similarly with $Y_1 = 5000$ ppm and $Y_2 = 0$.

Given the temperature T_1 and DBP vapor mass fraction Y_1 , the saturation ratio in the hot stream is $S \approx 0.7$. The boundary conditions for the temperature and vapor concentration resemble the experimental settings for the maximum nucleation case studied by Lesniewski and Friedlander.¹⁵ Relevant parameters for the turbulent mixing layer configuration are summarized in Table I.

Two Blasius boundary layers are used as inlet velocity profiles because they promote a rapid transition to turbulence. The hyperbolic tangent profile for the scalars is preferred to a Heaviside function in order to avoid inadequate resolution of the aerosol source terms near the inlet. In a

TABLE I. Simulation parameters. The domain size is normalized by the momentum thickness at the inlet $\delta_{\theta,0} = 0.201$ mm. Y_1 and Y_2 are the mass fractions of DBP at the hot and cold inlet, respectively.

Reynolds number $\Delta U \delta_\theta / \nu$	111 (inlet) - 1998 (outlet)
Domain size $L_x^* \times L_y^* \times L_z^*$	$640 \times 512 \times 214$
Grid $N_x \times N_y \times N_z$	$768 \times 398 \times 256 \approx 80$ M
Inlet conditions (fast stream)	$u_1 = 15$ m/s, $T_1 = 410$ K, $Y_1 = 5000$ ppm
Inlet conditions (slow stream)	$u_2 = 5$ m/s, $T_2 = 290$ K, $Y_2 = 0$ ppm

realistic experimental configuration, velocity and scalars profiles similar to those prescribed here at the inlet occur at a short distance downstream of a thin splitter plate. It is well-recognized that the flow characteristics in laminar and transitional regions depend on the inlet conditions. It will be shown that most of the aerosol is formed in the fully developed turbulent region of the mixing layer and minor effects related to the inlet conditions do not alter the conclusions reported in this paper.

B. Gas-phase heat and mass transport

The simulation is performed by solving the incompressible Navier-Stokes equations. The density of the gas is set to a constant $\rho = 1 \text{ kg/m}^3$, thereby neglecting buoyancy effects. The kinematic viscosity in the momentum equation is also set to a constant $\nu = 1.8 \times 10^{-5} \text{ m}^2/\text{s}$.

The evolution of DBP vapor mass fraction Y and temperature T are described by

$$\frac{\partial Y}{\partial t} + \nabla \cdot (\mathbf{u}Y) = D\nabla^2 Y + S_Y, \quad (1)$$

$$\frac{\partial T}{\partial t} + \nabla \cdot (\mathbf{u}T) = D\nabla^2 T, \quad (2)$$

where $\mathbf{u}$ is the velocity vector, and both the thermal and mass diffusivities are set equal to the kinematic viscosity, $D = \nu = 1.8 \times 10^{-5} \text{ m}^2/\text{s}$.

In the DBP mass fraction equation (Eq. (1)), the source term S_Y denotes the vapor consumption due to gas-to-particle conversion (i.e., nucleation and condensation). In the temperature equation (Eq. (2)), the effect of aerosol formation and growth is neglected. This simplification is acceptable, since the vapor mass fraction is very low ($Y \leq 5000 \text{ ppm}$). *A posteriori* analysis of the flow configuration considered shows that a negligible amount of DBP vapor is converted to liquid. Thus, effects on the continuity, momentum, and temperature (energy) equations related to the presence of DBP in vapor or liquid phase are inconsequential and are not included in the model.

Under the simplifications adopted in this work (i.e., equal diffusivities and negligible effects of latent heats), the mixture fraction $\phi = (T - T_2)/(T_1 - T_2)$ may be computed from the temperature field. By definition, the mixture fraction is a scalar bound between zero and one, being $\phi_1 = 1$ in the hot stream and $\phi_2 = 0$ in the cold stream. In the limit of negligible vapor consumption ($S_Y = 0$), the vapor concentration is linearly related to the mixture fraction as $Y(\phi) = Y_1\phi$ and the saturation ratio is a function of mixture fraction.

The relative importance of natural and forced convection may be estimated from the ratio of the Grashof number Gr and the square of the Reynolds number Re , $Gr/Re^2 = g(T - T_2)L/(u^2T_2)$. In this expression, g is the gravitational acceleration and L is set equal to the momentum thickness at the outlet ($\delta_\theta = 3.596 \text{ mm}$). The estimated maximum value of Gr/Re^2 is $1.5 \times 10^{-4} \ll 0.1$ near the outlet, indicating that the effect of buoyancy on the flow is negligible. This estimate is based on the most unstable case (temperature gradient opposite to gravity) and the largest convective length scale.

C. Aerosol microphysical models

Aerosol particles are assumed to be spherical and are characterized by their diameter d . The size distribution of non-inertial particles $n(\mathbf{x}, t; d)$, where $\mathbf{x}$ indicates position and t time, satisfies the general dynamic equation (GDE):⁴⁰

$$\frac{\partial n}{\partial t} + \nabla \cdot n\mathbf{u} = \tilde{I} + \tilde{G} + \tilde{C}. \quad (3)$$

The three source terms on the right-hand side denote homogeneous nucleation, growth via condensation, and coagulation, respectively. The diffusion term is neglected owing to the large Schmidt number of aerosol particles. For example, under standard atmospheric conditions, spherical particles with a diameter of 10 nm (resp., 100 nm) have a Schmidt number equal to 290 (resp., 2.2×10^4).⁴⁰

Direct solution of the GDE is not practical in three-dimensional, unsteady flows due to the high-dimensionality of the equation. Established approaches for the simulation of the evolution of an aerosol include moment,^{31,41,42} sectional,²¹ and Monte Carlo^{33,34} methods. Among these approaches, moment methods are tractable computationally for the purpose of simulating aerosols in turbulent flows.

The k th ($k = 0, 1, 2, \dots$) order moment of the size distribution $n(\mathbf{x}, t; d)$ is defined as

$$M_k(\mathbf{x}, t) = \int_0^\infty n(\mathbf{x}, t; d) d^k dd. \quad (4)$$

Therefore, M_0 indicates the particle number density ($1/\text{m}^3$), M_1 the diameter density (m/m^3), πM_2 the surface area density (m^2/m^3), and $\pi M_3/6$ the volume fraction. The dynamic equations for the moment set are obtained from the GDE by integration in sample space to give:

$$\frac{\partial M_k}{\partial t} + \nabla \cdot (M_k \mathbf{u}) = S_k, \quad (5)$$

where S_k is the source term for the k th moment due to the microphysical processes.

Homogeneous nucleation generates DBP particles according to the Becker-Döring theory^{40,43} with nucleation rate I and critical diameter d^* (the size of nucleated particles):

$$I = \frac{P_v x_v}{k_B T} \sqrt{\frac{2\sigma}{\pi m}} \exp\left(-\frac{16\pi\sigma^3 m^2}{3(k_B T)^3 \rho_p^2 (\ln S)^2}\right), \quad (6)$$

$$d^* = \frac{4\sigma v_m}{k_B T \ln S}, \quad (7)$$

$$\tilde{I} = \int I \delta(d - d^*) dd. \quad (8)$$

In the expressions above, P_v is the vapor pressure, x_v is the mole fraction of the vapor, m is the mass of one DBP molecule, ρ_p is the density of liquid DBP, $S = P_v/P_{\text{sat}}$ is the vapor saturation ratio, k_B denotes the Boltzmann constant, σ is the surface tension, and $v_m = m/\rho_p$ is the effective volume of one DBP molecule. Semi-empirical formulas for ρ_p , σ , and P_{sat} are taken from Okuyama *et al.*⁴⁴ and reproduced in Table II. At the saturation conditions considered here, the critical diameter lies in the range $2 \leq d^* \leq 3$ nm.

Figure 1 shows the saturation ratio and the homogeneous nucleation rate as a function of mixture fraction (or equivalently of temperature) for the two streams considered in this work. The saturation ratio peaks at $\phi_m^{(S)} = 0.06$ and the nucleation rate is maximum at $\phi_m^{(I)} = 0.13$, corresponding to $T_m^{(I)} = 306$ K. It is apparent that the nucleation rate is highly sensitive to the local mixing conditions and resulting saturation ratio, varying by more than two orders of magnitude for minor changes in mixture fraction around $\phi_m^{(I)}$ (see inset in Fig. 1(b)). The sensitivity of nucleation rates to the DBP vapor mass fraction is explored by varying the DBP mass fraction in the hot stream ($Y = Y_1\phi$, $0.98Y_1\phi$, and $0.90Y_1\phi$), while maintaining a constant temperature T_1 . A 10% reduction in the vapor mass fraction with respect to $Y_1\phi$ causes the maximum nucleation rate to drop to one third of the peak value under nominal conditions. The temperature at which the peak nucleation occurs does not vary appreciably in response to changing DBP mass fraction in the range of values considered.

The functional form of the particle diameter growth rate due to condensation depends on the Knudsen number $\text{Kn} = 2\lambda/d$, where λ is the mean free path.⁴⁵ In the free molecular regime ($\text{Kn} \geq 10$),

TABLE II. Physical properties of dibutyl phthalate (DBP). Temperature T in Kelvin. Adapted from Okuyama *et al.*⁴⁴

Molecular weight (g/mol)	$M = 278.35$
Density (kg/m^3)	$\rho_p = 1063 - 0.826(T - 273.16)$
Surface tension (kg/s^2)	$\sigma = 3.53 \times 10^{-2} - 8.63 \times 10^{-5}(T - 273.16)$
Saturation vapor pressure (Pa)	$P_{\text{sat}} = 133.0 \exp[16.27 - 5099.0/(T - 109.51)]$

FIG. 1. (a) Saturation ratio $S = P_v/P_{\text{sat}}$ and (b) homogeneous nucleation rate (see Eq. (6)) as a function of mixture fraction ϕ and temperature T . In both figures, curves for various values of the DBP vapor mass fraction in the hot stream are shown: $Y = \phi Y_1$ is the base case, while $Y = 0.98\phi Y_1$ and $Y = 0.90\phi Y_1$ indicate a 2% and 10% reduction in the hot stream DBP mass fraction Y_1 . Inset in (b): nucleation rates in logarithmic scale. The short dashed line in the inset indicates the locus of peak nucleation rate.

the growth rate G is independent of the particle size, while in the continuum regime ($\text{Kn} \leq 0.1$), it is inversely proportional to the diameter. In the transition regime ($0.1 < \text{Kn} \leq 10$), a harmonic mean of the expressions for the two regimes is used. The expressions for G and $\tilde{G}$ are:

$$G = \begin{cases} \frac{2(P_v - P_p)v_m}{(2\pi m k_B T)^{1/2}}, & \text{Kn} \geq 10 \\ \frac{4D(P_v - P_p)v_m}{k_B T d}, & \text{Kn} \leq 0.1 \\ \text{harmonic mean,} & 0.1 < \text{Kn} < 10 \end{cases} \quad (9)$$

$$\tilde{G} = -\frac{\partial n G}{\partial d}. \quad (10)$$

In the expressions above, P_p is the equilibrium vapor pressure for droplets of size d , which is equal to the saturation pressure with the Kelvin correction, i.e., $P_p = P_{\text{sat}} \exp[(d^*/d) \ln S/d]$. The Kelvin correction accounts for the surface tension of the particle and is significant only when the droplet diameter is approximately equal to the critical diameter d^* (Eq. (7)).

Figure 2 shows the diameter condensation growth rate G for various values of temperature, DBP mass fraction, and particle diameter in the ranges characteristic of this study. The diameter growth rate is independent of particle size in the free molecular regime ($d < 20$ nm). As the diameter of the particles increases, the growth rate decreases in the transitional and continuum regimes. In the temperature and vapor mass fraction ranges considered, particles grow most rapidly at $\phi_m^{(G)} = 0.67$ (or $T = T_m^{(G)} = 370$ K), regardless of diameter (Fig. 2(b)). In the case of condensation, negligible differences are found by varying the DBP mass fraction at the hot stream inlet (not shown). Finally, it is worth noting that the harmonic mean formula produces almost identical results to the generalized Mason's formula⁴⁶ as shown in Fig. 2(a).

For $P_v < P_p$, Eq. (9) leads to $G < 0$, resulting in particle evaporation. Given the temperature and vapor concentration of the hot and cold streams, evaporation occurs solely for $\phi \geq 0.96$, i.e., in the under saturated, hot stream, which has a saturation ratio $S \approx 0.7$. Our results show that evaporation plays a negligible role in the overall statistics of the aerosol, since negligible amounts of liquid DBP are found at $\phi \geq 0.96$ at any time, anywhere in the domain. In order to avoid subtle computational issues that compromise the robustness of the moment method for negative growth rates, G is set to zero when $P_v < P_p$ ($\phi \geq 0.96$) as shown in Fig. 2(b).

FIG. 2. Diameter condensation growth rate G (Eq. (9)) (a) as a function of diameter at 325 and 375 K and (b) as a function of mixture fraction ϕ and temperature T for various values of diameter ($d = 10$ nm, $d = 200$ nm, and $d = 4 \mu\text{m}$). In (a), the generalized Mason's formula⁴⁶ and the harmonic mean are compared and found to be equivalent for practical purposes.

Coagulation is described by the Smoluchowski equation⁴⁰

$$\tilde{C} = \frac{1}{2} \int_0^d \beta n(\tilde{d}) n(d - \tilde{d}) d\tilde{d} - \int_0^\infty \beta n(d) n(\tilde{d}) d\tilde{d}. \quad (11)$$

The collision kernel function $\beta(d, \tilde{d})$ describes the rate at which particles of diameter d collide with particles of diameter $\tilde{d}$. In the free molecular and continuum regimes, β is obtained from well established theories:⁴⁰

$$\beta(d, \tilde{d}) = \begin{cases} \left(\frac{3k_B T}{\rho_p} \right)^{1/2} \left(\frac{1}{d^3} + \frac{1}{\tilde{d}^3} \right)^{1/2} (d + \tilde{d})^2, & \text{Kn} \geq 50 \\ \frac{2k_B T}{3\mu} \left(\frac{A(d)}{d} + \frac{A(\tilde{d})}{\tilde{d}} \right) (d + \tilde{d}), & \text{Kn} \leq 1 \\ \text{harmonic mean,} & 1 < \text{Kn} < 50 \end{cases} \quad (12)$$

where μ is the dynamic viscosity of the gas-phase, and the slip correction factor A is used to extend the friction coefficient to the near-continuum regime ($\text{Kn} < 1$) and is given by⁴⁰

$$A = 1 + \text{Kn}[1.257 + 0.4 \exp(-0.55 \text{Kn})]. \quad (13)$$

The harmonic mean^{41,47,48} is used in the transition regime ($1 < \text{Kn} < 50$).

In the GDE (Eq. (3)), inertial and thermophoretic effects on the motion of particles are neglected. *A posteriori* analysis shows that both of these effects are negligible in this configuration. Particle inertial effects can be readily investigated by considering the Stokes number $\text{St} = \tau_p / \tau_f$, where τ_p and τ_f are characteristic times for the particle and the flow, respectively. In this simulation, the mean diameter is always less than $2 \mu\text{m}$. Given⁴⁰ $\tau_p = \rho_p d^2 / (18\mu)$, one obtains $\tau_p \leq 12 \mu\text{s}$. An estimate for the smallest characteristic time of the flow is provided by the minimum Kolmogorov time scale $(\nu/\epsilon)^{1/2}$ in the domain, so that $\tau_f \geq 67 \mu\text{s}$. Therefore, the Stokes number is $\text{St} = \tau_p / \tau_f \leq 0.18 \ll 1$ throughout the domain, indicating that inertial effects are negligible and particles move along flow pathlines. The thermophoretic velocity is estimated from the formula⁴⁰ $v_{\text{th}} = -3\nu \nabla T / [4(1 + \pi\alpha/8)T]$, where $\alpha = 0.9$ and the temperature gradient is $\nabla T \leq 10^5$ K/m, giving $v_{\text{th}} \leq 10^{-4}$ m/s. The thermophoretic velocity is much smaller than convective velocities and may be neglected.

D. Discretization and solution methods

1. Momentum, temperature, and vapor mass fraction

The solver⁴⁹ for the evolution of the momentum, temperature, and DBP vapor mass fraction implements a finite difference method on a spatially and temporally staggered grid with the second order semi-implicit fractional step method of Kim and Moin⁵⁰ with approximate factorizations. Velocity derivatives are approximated with a second order centered scheme. The convective term for the scalars is discretized with the third order WENO scheme⁵¹ and the diffusive term with a second order centered formula. A pressure-correction step involving the solution of a Poisson equation ensures mass conservation. The code decomposes the computational domain over a number of processors using the Message Passing Interface (MPI). The solution of the Poisson equation is performed by the library Hypra⁵² using the preconditioned conjugate gradient iterative solver (PCG) coupled with one iteration of an algebraic multigrid preconditioner.

2. Closure of the aerosol source terms

Moment methods require a closure approach and the quadrature method of moments^{31,32} is adopted in this work. The source term on the right-hand side of the moments equation (Eq. (5)) is written as

$$S_k = S_k^{(I)} + S_k^{(G)} + S_k^{(C)}, \quad (k = 0, 1, \dots, 2N_m - 1), \quad (14)$$

where^{31,53}

$$S_k^{(I)} = (d^*)^k I, \quad (15)$$

$$S_k^{(G)} = k \sum_{i=1}^{N_m} d_i^{i-1} W_i G, \quad (16)$$

$$S_k^{(C)} = \sum_{i=1}^{N_m} \sum_{j=1}^{N_m} W_i W_j \left[\frac{1}{2} (d_i^3 + d_j^3)^{k/3} - d_i^k \right] \beta(d_i, d_j), \quad (17)$$

and the weights W_i and abscissas d_i , ($i = 1, 2, \dots, N_m$) are defined through the Gaussian quadrature

$$M_k = \int_0^\infty n d^k dd = \sum_{i=1}^{N_m} d_i^k W_i, \quad (k = 0, 1, \dots, 2N_m - 1). \quad (18)$$

For a given set of $2N_m$ moments M_k ($k = 0, 1, \dots, 2N_m - 1$), the weights W_i and abscissas d_i are determined using the product difference algorithm.³¹ In this work, $N_m = 2$ and four moments (M_0 , M_1 , M_2 , and M_3) are used. In order to determine the weights and abscissas, the moment set must be realizable. A set of moments is realizable if there exists a size distribution, which satisfies the moment definition (Eq. (4)). Mathematically, the set of moments must satisfy the Stieltjes conditions to be realizable.⁵⁴

The source terms on the right-hand side of Eq. (14) are evaluated from Eqs. (15)–(17). It is worth pointing out that the nucleation term is independent of the weights and abscissas, i.e., independent of the particle size distribution. Condensation does not change the number density, i.e., $S_0^{(G)} = 0$. Coagulation reduces the number density and conserves the volume fraction, i.e., $S_0^{(C)} = -\frac{1}{2} \sum_{i=1}^{N_m} \sum_{j=1}^{N_m} W_i W_j \beta(d_i, d_j) < 0$, and $S_3^{(C)} = 0$.

3. Solution of the moment equations by a Lagrangian particle scheme

Lagrangian particle descriptions⁵⁵ are widely used in the analysis of discrete physical systems. Although not as popular as Eulerian schemes, a number of Lagrangian particle methods have been applied successfully to the solution of continuum systems^{35,56,57} (e.g., Navier-Stokes equations). A

recent review by Koumoutsakos⁵⁸ highlights the advantages of these approaches: adaptivity, multi-resolution capabilities, good stability properties, and scalability on massively parallel computers.

In the Lagrangian particle scheme applied to turbulent aerosol flows, a large number of notional particles are tracked, and the aerosol moments associated with notional particles evolve along the particles' trajectories:

$$\frac{d\mathbf{x}^p}{dt} = \mathbf{u}(\mathbf{x}^p), \quad p = 1, 2, \dots, N_p, \quad (19)$$

$$\frac{dM_k^p}{dt} = S_k^p, \quad k = 0, 1, \dots, 2N_m - 1, \quad (20)$$

where $\mathbf{x}^p$ and $\mathbf{u}(\mathbf{x}^p)$ denote the location and the fluid velocity at the location of notional particle p , and N_p is the number of Lagrangian particles. For each moment, the source term S_k^p is evaluated from Eq. (14). Note that the Lagrangian particles do not represent individual liquid droplets or aerosol particles; rather they are mathematical objects representing a fluid parcel evolving along pathlines. The Lagrangian particles' properties are evolved with a second-order Strang splitting approach,⁵⁹ comprising a half step of the aerosol source terms ($\Delta t/2$), a full step particle advection (Δt), a half step of the aerosol source terms ($\Delta t/2$). Equation (20) is integrated by the solver DOPRI5,⁶⁰ which implements a fifth-order explicit Runge-Kutta method. More details on the application of a Lagrangian particle method to the evolution of an aerosol in a turbulent flow may be found in Ref. 35.

The reconstruction of Eulerian moment fields from the Lagrangian particles uses

$$M_k(\mathbf{x}) = \frac{1}{N_\Omega} \sum_{p \in \Omega} M_k^p, \quad (21)$$

where Ω is the set of N_Ω particles located in a cube centered at $\mathbf{x}$ and having a side of length equal to three mesh spacings, equivalent to $3 \times 3 \times 3 = 27$ cells in the three-dimensional configuration.

Resolution issues related to the number of particles per cell and the width of the averaging (regularization) kernel have been addressed in Ref. 35. In the present configuration, a narrower kernel of length equal to one mesh spacing has been tested without observing any significant difference in the reconstructed fields. In addition, alternative averaging techniques (e.g., a Gaussian kernel) provide the same results.

The flow field velocity vector $\mathbf{u}$, temperature T , and DBP vapor concentration Y are advanced in a traditional Eulerian framework (see Secs. II B and II D 1), while aerosol moments are evolved along Lagrangian trajectories. The Eulerian and Lagrangian fields are coupled as follows. Tri-linear interpolation is used to recover Eulerian fields at the notional particle position. These are required to advance the particle position $\mathbf{x}^p$ and to evaluate the source term S_k^p from temperature and DBP vapor mass fraction. The sink term for DBP mass fraction in Eq. (1) is evaluated as

$$S_Y = -\frac{1}{N_\Omega} \sum_{p \in \Omega} \frac{\pi \rho_p}{6\rho} \frac{dM_3^p}{dt}. \quad (22)$$

Thus, the depletion rate of DBP vapor is proportional to the rate of change of the aerosol volume fraction ($\pi/6$) dM_3^p/dt and the DBP mass is conserved globally. Discrete conservation of DBP mass was assessed carefully. The error in the total flux of DBP, i.e., vapor plus liquid, is less than 1%.

In the context of the transport of aerosol moments, the Lagrangian particle scheme circumvents the issue of loss of the realizability of moments.³⁵ In the Eulerian framework, each moment is transported in physical space with a separate partial differential equation, which is affected by independent errors. The uncorrelated errors might compromise the consistency of the moments set, leading to a non-realizable distribution.³⁶ The Lagrangian approach is not affected by this issue because the advection process does not change the values of the moments associated with particles and the evolution of the moments is not polluted by errors due to discrete differential operators.

If two-way coupling between the aerosol and the gas-phase is neglected, i.e., no vapor consumption due to the formation and growth of particles, the aerosol dynamics due to microphysical

processes on each Lagrangian particle are not affected by the resolution of the reconstructed Eulerian moment fields. Conversely, if vapor consumption is important, there may be an indirect effect on the aerosol solution due to the resolution of the DBP mass fraction source term S_Y , which depends on the reconstructed moment fields as in Eq. (22). In the present configuration, vapor consumption is negligible and the DBP mass fraction field is well resolved, so that the evolution of the aerosol moments on each Lagrangian notional particle is independent of that on the other notional particles. Finally, we note that the resolution of the Lagrangian approach employed here is superior to that of an Eulerian method, since the large number of Lagrangian particles per cell leads to a local refinement of the moment fields. A detailed discussion addressing these and related issues is provided by Attili and Bisetti³⁵ and Lagaert *et al.*⁶¹

4. Discretization details

The domain is discretized with 80×10^6 grid points ($N_x \times N_y \times N_z = 768 \times 398 \times 256 \approx 80$ M). The dimensions of the grid cells are the same along the three axes with $\Delta x^* = \Delta y^* = \Delta z^* = 0.85$ in the center of the mixing layer for $|y^*| \leq 124$. For $|y^*| > 124$, the grid is linearly stretched along y with a ratio 1.05 and then uniform with $\Delta y^* = 3.4$. *A posteriori* analysis of the velocity field shows that $\Delta x = \Delta y = \Delta z < 3\eta_{\min}$, where the minimum value of the Kolmogorov length scale in the domain, $\eta_{\min}$, is calculated as follows. Given the statistical stationarity and homogeneity of the flow along the spanwise direction, $\epsilon = \epsilon(x, y) = 2\nu \langle S_{ij}S_{ij} \rangle$, where ϵ is the mean kinetic energy dissipation rate, S_{ij} is the instantaneous rate of strain tensor, and the brackets $\langle \rangle$ indicate the average in time and the homogeneous spanwise direction. The mean Kolmogorov scale $\eta(x, y) = \nu^{3/4} \epsilon^{-1/4}$ is computed everywhere in the domain and its minimum value $\eta_{\min}$ is used to assess resolution requirements. The time step size is selected so that the maximum Courant-Friedrichs-Lewy number is always less than 0.8.

Lagrangian particles are injected at the inlet plane at a rate proportional to the inlet velocity. The injection rate is selected to obtain 15 particles per cell on average. Particles are injected in the crosswise range $-120 < y^* < 120$ for the sake of computational cost, as turbulent mixing and aerosol growth are limited to this region. Approximately $N_\Omega = 400$ particles are used in the reconstruction of the Eulerian fields of the aerosol moments at each grid point (Eq. (21)) and the DBP vapor source term (Eq. (22)). Overall, the moment fields are described by 500×10^6 Lagrangian particles. The number of particles used limits the reconstruction error to a satisfactory level.

The simulations were performed on the IBM Blue Gene/P system “Shaheen” at King Abdullah University of Science and Technology using approximately 1×10^6 CPU hours over 4096 compute cores. After a statistically steady state is reached, the physical time simulated is 2τ , where $\tau = L_x/u_2 = 25.6$ ms corresponds to one flow through time of the slow stream. All statistics are obtained by averaging over different time instants (50 datasets separated by 1 ms time intervals) and the periodic z direction ($N_z = 256$), yielding more than 10 000 samples at each (x, y) grid location.

In closing, we note that the present simulation resolves all length and time scales of momentum and gas-phase scalars fully. Conversely, the evolution of liquid droplets is described statistically by a moment method with an adequate closure, so that the dynamics of individual droplets are not resolved. The high Schmidt number aerosol moments are solved by the Lagrangian particle method, which retains robustness and accuracy in the description of aerosol transport at the resolution afforded by a very large, albeit finite number of notional particles. As such, the numerical approach adopted here is best described as a DNS for the momentum and gas-phase scalars coupled with a robust and efficient model consisting of a Lagrangian scheme applied to the evolution of the moments of the aerosol.

III. RESULTS AND DISCUSSION

A. Overview

The hot fast stream saturated with DBP vapor and the cold slow stream of nitrogen mix in the turbulent layer. Locally, peak saturation ratios of 1270 are attained, promoting homogeneous nucleation and subsequent growth of the aerosol due to vapor condensing on the surface of newly

FIG. 3. Instantaneous fields of (a) mixture fraction ϕ , (b) number density N ($1/\text{cm}^3$), (c) volume fraction F , and (d) mean diameter $d = M_1/M_0$ (μm) on a $x-y$ plane. In (b), the solid white line along the bottom interface marks the $\phi = \phi_m^{(l)} = 0.13$ (or $T = T_m^{(l)} = 306$ K) isocontour, where nucleation is most intense (see Fig. 1(b)). A detailed view of the region in the square box marked in (c) and (d) is shown in Fig. 4.

nucleated particles. An instantaneous realization of the mixture fraction field is shown in Fig. 3(a). The mixing field starts with a laminar region near the inlet ($x^* = x/\delta_{\theta,0} \leq 60$). As the flow evolves in the streamwise direction, large-scale structures associated with the Kelvin-Helmholtz instability develop and grow in the transition region ($60 \leq x^* \leq 320$). The flow field is fully turbulent and its statistics are self-similar for $x^* > 320$. In the fully-developed region, the probability density function (PDF) of mixture fraction is “marching,” i.e., at any crosswise location within the turbulent region of the layer, the most probable and the mean values of mixture fraction are equal²⁶ (not shown). Also, the PDF of mixture fraction is well described by a beta-PDF in agreement with well-known results²⁶ (not shown).

Figures 3(b)–3(d) show the number density, volume fraction, and mean diameter fields, respectively. Although the figures reproduce a particular realization of the flow field, they represent the dominant trends of the aerosol evolution in the mixing layer. The condensing aerosol is confined in the turbulent region of the flow between the top and bottom interfaces, marked by $\phi = 0.95$ and $\phi = 0.05$ in Fig. 3(a). In the transition region ($60 \leq x^* \leq 320$), particles abound inside the large, quasi two-dimensional structures characteristic of the Kelvin-Helmholtz instability. The aerosol fields appear highly intermittent and are characterized by smaller spatial structures than those of the mixture fraction field. Significant number densities of aerosol particles emerge early in the laminar and transition regions of the mixing layer and peak number densities of $2 \times 10^5/\text{cm}^3$ are apparent at selected locations near the bottom interface (Fig. 3(b)).

Unlike number density, volume fraction grows appreciably only beyond the transition region of the flow for $x^* > 320$, and patches of fluid characterized by up to 0.03 ppm of liquid DBP appear near the domain boundary. The qualitative appearance of the volume fraction field resembles the “wedge of fog” noted by Hidy and Friedlander¹⁸ in their study on vapor condensation in turbulent

jets, whereby the aerosol cloud forms at a certain distance downstream from the injection plane. In between the cloud and the injection location, homogeneous nucleation leads to the formation of numerous, tiny droplets, which grow by condensation as they move downstream.

Figure 3(d) shows the mean diameter $d = M_1/M_0$. The smallest particles populate the region of the flow near the bottom interface, while the largest particles (diameter up to $1.2 \mu\text{m}$) are found close to the top interface between the turbulent region and the hot, fast stream laden with DBP vapor. Recall that the local values of temperature and vapor concentration are most favorable to nucleation near the bottom interface, while they are most favorable to condensation near the top interface (compare Figs. 1(b) and 2(b)).

A detailed view of number density, nucleation rate, volume fraction, and growth rate is shown in Fig. 4. In all plots, the $\phi_m^{(I)} = 0.13$ and $\phi_m^{(G)} = 0.67$ isocontours identify the location of peak nucleation, and peak diameter condensation growth rate, respectively (see Figs. 1(b) and 2(b)). Along the $\phi = 0.13$ mixture fraction contour, the supersaturation is around 518, resulting in a peak nucleation rate of $9.8 \times 10^7/\text{cm}^3\text{s}$, so that low maximum number densities of particles are attained in the mixing layer ($N \leq 2 \times 10^5/\text{cm}^3$). Even though nucleation is important in narrow regions of the flow along the contour $\phi = 0.13$ (see Fig. 4(b)), patches of peak particle density populate a much wider region of space. At times, these patches of high number density lie on or near the $\phi = 0.13$ isocontour, but not always. The number density field in Fig. 4(a) suggests that particle densities are highest on the lower half of the mixing layer, as confirmed by the mean statistics, which will be discussed in Secs. III B and III C.

As for number density, volume fraction F appears in patches (Fig. 4(c)). Unlike number density, however, most of the aerosol volume fraction is present on the upper half of the mixing layer, with

FIG. 4. Detail of the region of the flow marked by the square box in Figs. 3(b)–3(c): (a) number density N ($1/\text{cm}^3$), (b) nucleation rate I ($1/\text{cm}^3\text{s}$), (c) volume fraction F , and (d) volume condensation growth rate $S_3^{(G)}$ (ppm/s) as defined in Eq. (16). The solid white line indicates the isocontour of $\phi_m^{(I)} = 0.13$ (or $T = T_m^{(I)} = 306 \text{ K}$), which corresponds to the location of peak nucleation rate. The gray line indicates $\phi_m^{(G)} = 0.67$, which corresponds to the location of peak diameter condensation growth rate. The black solid line indicates $\phi = 0.95$, which highlights the turbulent/non-turbulent interface on the fast stream side at the top of the mixing layer.

TABLE III. Momentum thickness δ_θ at various streamwise locations. The streamwise location is normalized by the momentum thickness at the inlet, $\delta_{\theta,0}$.

x (mm)	$x^* = x/\delta_{\theta,0}$	δ_θ (mm)	$\delta_\theta/\delta_{\theta,0}$
0	0	0.201	1.00
8.4	42	0.233	1.16
16.8	83	0.367	1.84
25.3	125	0.676	3.38
50.7	249	1.418	7.09
84.6	415	2.423	12.1
118.4	581	3.206	16.0

an apparent bias for the region near the isoline $\phi = 0.67$. The growth rate $S_3^{(G)}$ (Fig. 4(d)) correlates strongly with the volume fraction as a large volume fraction corresponds to high values of surface density and related condensation-based growth rates. Patches of aerosol particles with large values of volume fraction populate the middle of the layer.

The spatial structures of the number density and volume fraction fields are governed by the combined effects of the high Schmidt number of aerosol scalars and the nonlinearity of the source terms with respect to the saturation ratio and, in the case of growth rate, the aerosol surface density. Similar fields characterized by patches are observed by Das and Garrick²² in the far field of a turbulent planar jet in which titanium dioxide nanoparticles nucleate and grow as well as in turbulent nonpremixed sooting flames simulated by Attili *et al.*³⁸ By virtue of the high Schmidt number characteristics of aerosol transport, there exist spatial scales smaller than those observed in the reconstructed Eulerian moments' fields. However, an analysis of the sensitivity of the size and shape of these structures to the grid resolution reveals that while minor details vary using finer grids, the size and main geometrical features of the structures do not change. Most importantly, the statistics in physical and mixture fraction spaces are grid-independent.³⁸ In Ref. 38, a comprehensive analysis of the dependence of the solution on grid size is discussed in the context of soot formation and growth in turbulent flames.

Although the fields in Figs. 3 and 4 portray a single realization of an unsteady, three-dimensional field, they support qualitative conclusions on the evolution of the aerosol. Particles nucleate on the cold, dry side of the layer and drift towards the middle of the layer, where they encounter vapor and temperature conditions favorable to growth by condensation. The proposed dynamics of aerosol formation and growth agree with the mechanism advanced by Hidy and Friedlander¹⁸ and later by Lesniewski and Friedlander.¹⁵ Flow statistics are presented and discussed next in Secs. III B and III C.

B. Spatial statistics

The mean number density and volume fraction in the crosswise direction $y^+ = y/\delta_\theta$ are shown in Fig. 5 at various streamwise locations $x^* = x/\delta_{\theta,0}$. A summary of the momentum thickness δ_θ is shown in Table III. The mean number density profiles vary qualitatively depending on the streamwise location. Among the streamwise locations shown, the mean number density is highest in the laminar flow region at $x^* = 42$ with a peak value equal to $2 \times 10^5/\text{cm}^3$. At this streamwise location, aerosol particles are concentrated in a thin spatial region, where supersaturation is highest and nucleation is most intense near the bottom interface ($y^+ < 0$). As the flow field transitions from laminar to turbulent, the maximum mean number density across the layer decreases and moves towards the middle of the layer at $y^+ = 0$ for $x^* = \{83, 125, 249\}$. In the fully developed region, the mean number density profile is maximum at $y^+ \approx -2$ with a peak value close to $0.5 \times 10^5/\text{cm}^3$. The crosswise location and value of the maximum are comparable at the two streamwise locations in the fully developed turbulent flow region ($x^* = 415$ and $x^* = 518$).

The peak value of the mean number density decreases severalfold in the streamwise direction in concomitance with the transition of the flow from laminar to turbulent. It is important to recognize that this decrease in the mean number density is not due to coagulation, rather to the unsteadiness of

FIG. 5. (a) Mean number density $\langle N \rangle$ and (b) mean volume fraction $\langle F \rangle$ in the crosswise direction $y^+ = y/\delta_{\theta}$ at various streamwise locations $x^* = x/\delta_{\theta,0}$, where δ_{θ} is the value of the momentum thickness at the corresponding streamwise location and $\delta_{\theta,0}$ is the momentum thickness at the inlet. See Table III for a list of the values of δ_{θ} at all streamwise locations.

the flow field and its effect on mean statistics. As will be argued in detail, the range of supersaturation values in the flow field does not lead to number densities for which coagulation is significant. In this work, the aerosol regime is akin to the nucleation-controlled case investigated in Lesniewski and Friedlander.¹⁵

The crosswise profiles of mean volume fraction shown in Fig. 5(b) reveal a markedly different behavior compared to that of number density. First, the peak value of $\langle F \rangle$ across the mixing layer increases monotonically along the streamwise direction, with its location remaining between $y^+ \approx -1$ and $y^+ \approx 0$ throughout the domain. Near the outlet, the mean volume fraction peaks at 0.013 ppm.

It is well-known that the center of the mixing layer shifts towards the low-speed side in the streamwise direction.^{27,62} Since the shift is linear with the streamwise distance x , the position of the center of the layer in terms of the normalized crosswise coordinate y^+ remains constant. This normalized position is usually at $y^+ \approx -0.5$,²⁷ approximately at the same location where the peak aerosol volume fraction is observed in the present configuration.

Second, the mean values of volume fraction in the laminar and transition regions are severalfold smaller than those in the fully developed region. The monotonic growth in volume fraction reflects residence time effects on the aerosol, as particles grow by vapor condensation inside the turbulent region of the mixing layer.

The streamwise evolution of the peak values of the mean number density and volume fraction across the layer are shown in Fig. 6(a). After the laminar and transitional flow regions of the mixing layer ($x^* \geq 200$), both peak number density and volume fraction increase monotonically, albeit at different rates. The growth rate of the peak number density is linear with x^* , while that of the peak volume fraction is nonlinear due to the enhancement of the growth rate associated with the increase in the surface area of the growing droplets.

Further insight into the spatial evolution of the aerosol is gained by considering the mean streamwise flux of number density and volume fraction (see Fig. 6(b)). Both mean fluxes increase monotonically, suggesting that nucleation and growth persist throughout the domain. As already shown for the peak value of the mean statistics, the rate of increase of the volume fraction flux is greater than that of nucleation, owing to the effect of increasing surface density resulting from volumetric growth.

The distribution of the mean streamwise fluxes along the streamwise direction clarifies the fraction of aerosol generated in the laminar, transitional, and fully-turbulent regions. Towards the end of the transition region ($x^* \approx 200$), the number density flux is 20% of the flux at the outlet, indicating that 80% of the particles at the exit plane originate in the fully developed region of the turbulent mixing layer. The volume fraction flux at the same streamwise location is negligible (less than 2%), implying that the majority of vapor to liquid conversion occurs in the fully-turbulent

FIG. 6. Streamwise evolution of (a) the peak value of the mean number density $\langle N \rangle^{\max}$ and volume fraction $\langle F \rangle^{\max}$ and (b) the mean streamwise flux of number density and volume fraction (normalized). The streamwise coordinate is normalized with the momentum thickness at the inlet, i.e., $x^* = x/\delta_{\theta,0}$.

flow ($x^* > 320$). We may conclude that the aerosol statistics near the end of the domain reflect the interaction between turbulence and aerosol growth processes and are minimally affected by the laminar flow region and its transition to turbulence. Thus, the statistics presented here illustrate the general features of aerosol formation and growth in the presence of turbulent fluctuations in temperature and vapor concentration. In all likelihood, the conclusions based on our analysis in the fully developed region of the mixing layer are applicable to the evolution of condensation aerosols in fully developed shear turbulence with a mean mixing gradient.

The stream composition considered in this study is similar to the experimental conditions in Ref. 15. Nonetheless, the authors¹⁵ consider a turbulent round jet, while our flow configuration is a spatially developing turbulent mixing layer. Lesniewski and Friedlander¹⁵ postulate that the saturation ratio of the mixture drops quickly in the streamwise direction due to the sharp decrease in vapor concentration brought about by mixing, inhibiting homogeneous nucleation. As a result, the mean number density of particles decreases along the jet centerline, while the mean of the number density flux remains constant. Their conclusions agree with the well-known results on mixing statistics in a jet and with a previous analysis.¹⁸ In our flow configuration, nucleation occurs at all streamwise locations and contributes to an increasing mean number density flux.

The nucleation rate and diameter condensation growth rate across the mixing layer are shown in Fig. 7, where the mean rates and the rates computed from the mean fields are compared. As

FIG. 7. Nucleation rate and diameter condensation growth rate in the crosswise direction $y^+ = y/\delta_\theta$ at various streamwise locations $x^* = x/\delta_{\theta,0}$. (a) nucleation rate: $\langle I \rangle = \langle I(T, Y) \rangle$ and $\bar{I} = I(\langle T \rangle, \langle Y \rangle)$. (b) diameter condensation growth rate: $\langle G \rangle = \langle G(T, Y) \rangle$ and $\bar{G} = G(\langle T \rangle, \langle Y \rangle)$. The diameter condensation growth rate is calculated with the expression for the free molecular regime, which is independent of particle diameter.

discussed in previous works,^{20,24,63,64} the difference between $\langle I \rangle$ and $\bar{I} = I(\langle T \rangle, \langle Y \rangle)$ is due to the fluctuations of temperature and vapor concentration. In contrast, the effect of scalar fluctuations on diameter condensation growth rate is modest. While $\langle I \rangle$ and $\bar{I}$ differ severalfold, $\bar{G} = G(\langle T \rangle, \langle Y \rangle)$ appears to be a good approximation to $\langle G \rangle$ in the configuration at hand. This behavior may be related to the significant nonlinearity of the nucleation rate with respect to mixture fraction (see Fig. 1(b)) and the quasi-linear behavior of the diameter condensation growth rate away from peak growth at $\phi = 0.67$ (see Fig. 2(b)).

The usage of $\bar{G} = G(\langle T \rangle, \langle Y \rangle)$ as a suitable approximation to $\langle G \rangle$ has important implications from a practical perspective. First, the measured or computed mean fields $\langle T \rangle$ and $\langle Y \rangle$ may be employed in a post-processing step to estimate growth rates in the flow domain without knowledge or consideration of turbulent fluctuations. Second, in the context of Reynolds-Averaged Navier-Stokes (RANS) and large-eddy simulation (LES), neglecting effects related to turbulent fluctuations in the temperature and vapor fields simplifies the formulation of closure models for the mean condensation rate greatly.

C. Conditional statistics

The spatial statistics of aerosol quantities and related growth rates provide an informative picture of the spatial distribution of the aerosol in the mixing layer. Nevertheless, important aspects of the evolution of the aerosol are associated with droplet formation and growth rates. In this section, we explore aerosol statistics conditioned on the local value of the mixture fraction. Recall that, if vapor consumption is negligible, temperature and vapor mass fraction are related linearly to the mixture fraction and a unique functional relation between saturation ratio and mixture fraction exists. Given that droplets are formed and grow rapidly for large values of saturation ratio, conditioning on mixture fraction allows to sort the aerosol statistical samples with respect to the droplet growth rates associated with the local gas-phase temperature and vapor concentration.

Figure 8 shows the mean of number density, volume fraction, mean diameter, and surface density conditioned on mixture fraction. In the laminar flow region ($x^* = 42$), particles lie in a narrow range of mixture fraction values near the location of peak nucleation at $\phi_m^{(I)} = 0.13$. During the onset of the Kelvin-Helmholtz instability and in the transition region ($x^* = \{42, 125\}$), particles spread over a wide range of mixture fractions, mostly at larger values of ϕ towards the hot and humid stream laden with vapor. In the fully-developed region of the flow ($x^* = \{249, 415, 581\}$), the conditional number density is non-zero at all values of mixture fraction with a persistent maximum at the location of peak nucleation.

The conditional mean of the volume fraction shows a broader distribution in mixture fraction space compared to number density and displays a peak in the middle of the layer at $\phi \approx 0.5$ for all locations in the fully-developed region. With the exception of the laminar flow region, significant scatter around the mean is apparent for both statistics. Although the maximum values of the conditional means continue increasing in the streamwise direction, their distribution in mixture fraction space attains a quasi-similar shape in the developed region of the mixing layer.

Consistently with the scatter observed in Fig. 8, the conditional PDFs of number density and volume fraction are very broad (not shown). The broad distribution of the aerosol samples around the conditional means has significant modeling implications for the formulation of adequate closure approaches in Reynolds Averaged Navier Stokes (RANS) calculations and large-eddy simulation (LES) of turbulent aerosols. The scatter indicates that conditioning on the local composition and temperature of the gas-phase alone does not provide definitive information on the instantaneous values of number density and aerosol volume fraction. Thus, connecting the statistics of number density and volume fraction to those of the mixture fraction field, which is well characterized in canonical turbulent flows and accessible via adequate simulation techniques such as LES, is not a fruitful modeling approach. Examples of effective closure strategies based on the statistics of mixture fraction include the family of flamelet-based methods and the conditional moment closure approach in use in turbulent combustion modeling.^{65–68} As it will be explained in detail in Sec. III D, the

FIG. 8. Conditional mean of aerosol quantities versus mixture fraction along the streamwise direction: (a) number density $\langle N|\phi \rangle$, (b) volume fraction $\langle F|\phi \rangle$, (c) mean diameter, $\langle d|\phi \rangle = \langle M_1/M_0|\phi \rangle$, and (d) surface density $\langle \pi M_2|\phi \rangle$. A small subset of all data points used to compute the conditional statistics is shown as scatter for $x^* = x/\delta_{\theta,0} = \{42, 581\}$.

occurrence of scatter in the aerosol's conditional statistics is due to turbulent mixing in combination with the high Schmidt number of aerosol particles. Furthermore, it will be argued that this is a general feature of the evolution of aerosols in turbulent flows. In the absence of turbulent fluctuations, such as in the near field of the mixing layer at $x^* = 42$, scatter in the aerosol's conditional statistics does not occur.

The differences in the distribution of the number density and volume fraction reflect the fact that the diameter condensation growth G (Eq. (9)) is less localized in mixture fraction space than homogeneous nucleation. While particles nucleate solely in a narrow region near $\phi_m^{(I)} = 0.13$, they grow at all compositions throughout the layer and most intensely around $\phi_m^{(G)} = 0.67$ (Fig. 2(b)). The value of the conditional mean diameter $\langle d|\phi \rangle$ grows monotonically up to its peak around $\phi = 0.7$, which slightly exceeds $\phi = 0.67$ (corresponding to the peak diameter condensation growth rate), since particles drifting beyond $\phi = 0.67$ towards higher mixture fractions continue to grow. The profiles of mean surface density are very similar to the volume fraction, except that the peak values of the surface density span a narrower region than for the volume fraction.

Conditional statistics of nucleation rates in the transitional and fully developed flow regions are shown in Fig. 9(a). The conditional mean does not change appreciably at the various streamwise locations and very limited scatter in the data occurs. Droplets with a critical diameter $d^* = 2.1$ nm are formed at a peak rate equal to $9.8 \times 10^7/\text{cm}^3 \text{ s}$, which compares well with the low formation rates in the nucleation-controlled regime estimated by Lesniewski and Friedlander.¹⁵ Given the range of vapor supersaturation and flow time scales of the present flow configuration, DBP vapor consumption is negligible, explaining the lack of scatter in the conditional statistics of the nucleation rate.

FIG. 9. Conditional mean of aerosol growth rates along the streamwise direction: (a) nucleation rate $\langle I | \phi \rangle = \langle S_0^{(I)} | \phi \rangle$, (b) coagulation rate $\langle S_0^{(C)} | \phi \rangle$. A small subset of all data points used to compute the conditional statistics is shown as scatter for $x^* = x/\delta_{\theta,0} = \{42, 581\}$.

Figure 9(b) presents the conditional mean of the coagulation rate. It is apparent that the rate of coagulation is six orders of magnitude smaller than the nucleation rate. Thus, coagulation is negligible due to low values of particle density.

Figure 10(a) shows the conditional mean of the normalized DBP vapor deficit due to vapor scavenging by particle growth. Note that $Y = Y_1\phi$, should there be no vapor consumption, since a Lewis number of unity is assumed. The relative vapor deficit (normalized by the local vapor mass fraction) is largest around $\phi = 0.2$. The maximum mean relative deficit is around 0.8%. This is equivalent to reducing the DBP vapor mass fraction at the hot inlet by 0.8%.

The process of vapor scavenging has been well documented in various laminar configurations^{8,41} and is responsible for suppressing homogeneous nucleation in laminar mixing layers for long residence times and high values of supersaturation. In the present configuration, this feedback mechanism is not important as shown by the conditional PDF of nucleation rate at the mixture fraction of maximum nucleation rate $\phi_m^{(I)} = 0.13$ ($T = T_m^{(I)} = 306$ K) in Fig. 10(b). In response to vapor consumption, the conditional PDF of nucleation rate shifts towards lower values moving downstream, although the most probable value of nucleation rate near the outlet is only 10% lower than the value of $9.8 \times 10^7/\text{cm}^3 \text{ s}$, calculated neglecting vapor consumption.

FIG. 10. (a) Conditional mean of the normalized DBP mass fraction deficit $\langle Y_1\phi - Y | \phi \rangle / Y_1\phi$ at various streamwise locations. A small subset of all data points used to compute the conditional statistics is shown as scatter for $x^* = x/\delta_{\theta,0} = 581$. (b) Conditional PDF of nucleation rate at $\phi = \phi_m^{(I)} = 0.13$ (or $T = T_m^{(I)} = 306$ K), where $\phi_m^{(I)}$ and $T_m^{(I)}$ are the mixture fraction and temperature at which maximum nucleation occurs (see Fig. 1(b)). $I^{\max}$ is the maximum nucleation rate without vapor consumption. The conditional PDF is shown at various streamwise locations.

FIG. 11. Conditional condensation growth (a) diameter growth rate (Eq. (9)) and (b) volume growth rate $\langle S_3^{(G)}|\phi \rangle$ (Eq. (16)). A small subset of all data points used to compute the conditional statistics is shown as scatter for $x^* = x/\delta_{\theta,0} = \{42, 581\}$.

Figure 11(a) shows the conditional mean of the particle diameter condensation growth rate. $\langle G|\phi \rangle$ is computed as in Eq. (9) using the mean diameter $d = M_1/M_0$. All profiles are very similar to the growth rate function shown in Fig. 2(b) with a peak at $\phi_m^{(G)} = 0.67$. Along the streamwise direction, the growth rate decreases, since particles increase in size moving downstream and growth is inversely proportional to particle size in the continuum regime (Eq. (9)). The moderate scatter in the plots reflects the variation of the mean diameter in mixture fraction space. The conditional mean of the volume condensation growth rate (Fig. 11(a)) peaks at different mixture fraction values at different streamwise locations. Unlike the diameter growth rate G , the volume growth rate $S_3^{(G)}$ depends on the local surface density of particles and $\langle S_3^{(G)}|\phi \rangle$ closely resembles the surface density shown in Fig. 8(d).

To conclude, the scatter of conditional statistics of number density and volume fraction in mixture fraction space is not explained by conditional fluctuations of the nucleation and condensation growth rates alone, as vapor consumption is hardly significant in the present configuration. These features point to the importance of turbulent transport and related drift of aerosol particles in mixture fraction space. Both of these issues are best addressed in the context of Lagrangian statistics of the aerosol quantities.

D. Lagrangian statistics of the aerosol evolution

The Lagrangian particle method entails evolving aerosol quantities associated with “notional particles” or “fluid parcels” along flow pathlines. As such, the method provides a unique opportunity to investigate the evolution of a parcel of fluid laden with non-inertial particles as they are transported in the turbulent mixing layer. Figure 12 shows the evolution of mixture fraction and aerosol quantities along a sample Lagrangian trajectory. As is apparent from the value of the mixture fraction at time $t = 0$, the fluid parcel originates in the cold stream. At 5 ms, the parcel is entrained in the turbulent region and undergoes a rapid increase in mixture fraction. As the mixture fraction increases through the value most conducive to nucleation ($\phi = \phi_m^{(I)} = 0.13$ or $T = T_m^{(I)} = 306$ K), a short-lived burst of nucleation results in a minor increase in number density. For $5 \leq t \leq 10$ ms, the aerosol within the fluid parcel grows, but there is no additional nucleation, and aerosol particles reach a mean diameter equal to $0.4 \mu\text{m}$. Around $t = 10$ ms, the fluid parcel moves into lower mixture fractions and nucleation resumes as the parcel remains at $\phi = \phi_m^{(I)}$ for 5 ms ($10 \leq t \leq 15$ ms), resulting in a severalfold increase in number density and a sharp decrease in mean diameter due to the rapid addition of newly nucleated, tiny particles. Finally ($t > 15$ ms), the fluid parcel settles in a region near the cold stream interface ($T \approx 300$ K), where nucleation is suppressed. From $t = 15$ ms onwards, the aerosol volume fraction increases due to the increasing surface density available for condensation.

The rapid variations of temperature and vapor concentration and related changes in formation and growth rates along the Lagrangian trajectory of aerosol parcels are important to the overall

FIG. 12. Evolution in time of gas-phase and aerosol properties along a sample Lagrangian trajectory: (a) mixture fraction ϕ , where $T_m^{(I)}$ and $T_m^{(G)}$ correspond to maximum nucleation rate and diameter condensation growth rate, respectively, (b) nucleation rate I , (c) number density N , (d) diameter condensation growth rate G , (e) mean diameter d , and (f) volume fraction F .

spatial distribution of particles and the properties of the aerosol. These rapid variations are due to two concurring effects.

First, the transport of aerosol parcels is related to advection along pathlines (see Eq. (5)), while temperature and vapor concentration are characterized by advection and diffusion. This difference in the transport of aerosol parcels and scalars is due to the high Schmidt number associated with aerosol particles ($Sc \gg 100$) and unity Schmidt number for temperature and vapor ($Sc = \mathcal{O}(1)$). The difference in Schmidt number causes a drift of the aerosol particles in scalar space (i.e., mixture fraction ϕ). The drift is quantified by the material derivative of mixture fraction, $D\phi/Dt$, which is readily available as part of the mixture fraction time history $\phi(t)$ along a Lagrangian trajectory, i.e., $D\phi/Dt = \phi'(t)$. Alternatively, the material derivative of ϕ may be obtained from the transport equation for mixture fraction, i.e., $D\phi/Dt = D\nabla^2\phi$. A positive value of $D\phi/Dt$ indicates that the fluid parcel is drifting towards higher values of ϕ (towards the hot stream), while a negative value of the material derivative indicates the opposite (drift towards the cold stream).

The second important effect is related to turbulent mixing. As well-known, the turbulent scalar mixing field is characterized by a wide range of time and length scales. The gradient of mixture fraction, i.e., the scalar dissipation rate $\chi = 2D|\nabla\phi|^2$, is highly intermittent.¹¹ The statistics of scalar dissipation rate affect the transport of aerosol particles in mixture fraction space (as well as in temperature and vapor spaces) and have a strong influence on the nucleation and growth of aerosol particles. Strong scalar dissipation events are associated with large drift events of aerosol particles in mixture fraction space. One of these events is apparent in Fig. 12 at 5 ms, when the fluid parcel moves quickly from $\phi = 0$ to $\phi = 0.4$, experiencing very different rates of nucleation and growth at the two locations in mixture fraction space.

Figure 13 shows the mean of $D\phi/Dt$ conditioned on the value of ϕ at various streamwise locations. Recall that negative values indicate a movement towards the cold stream and positive values indicate a drift towards the hot stream. At all streamwise locations, $\langle D\phi/Dt|\phi \rangle$ changes sign from positive to negative at $\phi \approx 0.5$. The trend is expected, as the large scale mixing process in the mixing layer results in the passive scalar ϕ converging towards the value 0.5. It is apparent that the conditional statistics depend on the streamwise location and that in the laminar flow ($x^* = 42$) and transition ($x^* = \{83, 125, 249\}$) regions, $\langle D\phi/Dt|\phi \rangle$ is significantly larger than in the fully developed region ($x^* = \{415, 581\}$). In the fully developed region, the conditional statistics are independent of the streamwise location. The statistics in the laminar and transition regions explain the sudden shift in the maximum conditional means of number density and volume fraction towards

FIG. 13. Conditional mean of $D\phi/Dt$ at various streamwise locations. The dotted line marks $D\phi/Dt = 0$.

larger values of ϕ at locations $42 \leq x^* \leq 249$ shown in Fig. 8. Similarly, in the fully developed region, the conditional statistics of drift are independent of the streamwise location, contributing to a quasi-self-similar behavior of the aerosol conditional statistics for $x^* = \{415, 581\}$ observed in Fig. 8.

A more complete picture of the statistics of drift must take into account turbulent fluctuations. The PDF of $D\phi/Dt$ conditioned on mixture fraction is provided in Fig. 14. The mean associated with the conditional PDF is shown in Fig. 13, where $\langle D\phi/Dt | \phi \rangle > 0$ for $\phi \leq 0.5$ and $\langle D\phi/Dt | \phi \rangle < 0$ for $\phi \geq 0.5$. The most important feature of the drift statistics is that, due to turbulence, the probability of drift against the mean is non-negligible. As an example, consider the mean value of $D\phi/Dt$ at $\phi = 0.7$. Although $\langle D\phi/Dt | \phi = 0.7 \rangle < 0$, the PDF in Fig. 14 shows that positive drift towards larger values of ϕ occurs with a non-zero probability.

The occurrence of drift against the mean is due to turbulent mixing and is critical in explaining the spatial distribution of the aerosol volume fraction. If aerosol particles were to drift in mixture fraction space solely due to mean scalar transport, such as in a laminar flow, the aerosol would occupy only the bottom half of the mixing layer at locations where $\phi \leq 0.5$. Upon nucleating at $\phi_m^{(I)} = 0.13$, the particles would proceed to grow at compositions with $\phi \leq 0.5$. As shown, the aerosol evolution in a turbulent mixing layer is more complex. Particles are present at all mixture fractions (see Fig. 8(a)) and growth is most intense at $\phi_m^{(G)} = 0.67$ (see Fig. 11(a)). Turbulent fluctuations superposed on the mean drift process are responsible for displacing particles from the nucleation region into the growth region. We may conclude that turbulent mixing enables vapor to particle conversion at $\phi \geq 0.5$, providing droplets for condensation. In the absence of turbulent fluctuations in scalar mixing, vapor to liquid conversion would not occur at $\phi \geq 0.5$.

FIG. 14. The PDF of $D\phi/Dt$ conditioned on various values of ϕ at streamwise locations (a) $x^* = 125$ and (b) $x^* = 415$. The mean values associated with the conditional PDF are shown in Fig. 13.

IV. CONCLUSIONS

A large-scale simulation of aerosol nucleation and growth in a turbulent mixing layer has been performed and analyzed with the aim of elucidating the key processes involved. The configuration features a cold, dry stream of nitrogen mixing with a hot stream of nitrogen laden with DBP vapor. The stream conditions are selected to mimic experiments on condensing aerosols in turbulent flows.^{15,18}

All length and time scales of fluid motion and mixing are resolved as in a DNS of turbulent flow. The quadrature method of moments describes the dynamics of the condensing, non-inertial droplets and is coupled to the gas-phase fully. A Lagrangian particles scheme is used to advance the moment set, circumventing issues connected to the loss of realizability of moments, which compromise the robustness of conventional Eulerian advection schemes.

Particles nucleate on the cold, dry side of the layer and drift towards the middle of the layer, where they encounter vapor and temperature conditions favorable to growth by condensation. The dynamics of aerosol formation and growth agree qualitatively with the analysis advanced previously.^{15,18}

Beyond the laminar region of the flow near the inlet, the peak values of the mean number density and volume fraction increase in the streamwise direction due to persistent nucleation and particle growth. Given the values of particle densities in the present configuration, coagulation is negligible and number density is solely affected by nucleation. The continuous aerosol growth is reflected in the mean streamwise number density and volume fraction fluxes, which increase in the downstream direction. For the most part, aerosol formation and growth occur in the fully developed region of the mixing layer, so that the statistics near the end of the domain reflect the interaction between turbulence and aerosol growth processes and are affected minimally by the laminar flow region and the transition of the flow to turbulence.

The statistics of aerosol quantities, such as number density, volume fraction, mean diameter, and surface density are conditioned with respect to mixture fraction, upon which aerosol growth rates depend. The conditional statistics show that the mean number density peaks at the location of the most intense nucleation near the cold interface. The conditional mean volume fraction reaches a maximum around $\phi = 0.5$, reflecting the fact that particles grow in a wide range of mixture compositions and temperatures, with most of the aerosol volume occupying the middle of the mixing layer. The effect of turbulent fluctuations on the conditional statistics of growth rates is minimal, due to negligible vapor consumption. Conversely, the conditional statistics of number density and volume fraction display large scatter, which points to the importance of turbulent transport and related drift of aerosol particles in mixture fraction space.

The evolution of aerosol quantities along a sample Lagrangian trajectory has been discussed in detail. The rapid variation of temperature and vapor concentration due to intermittent mixing results in sudden changes in the nucleation and growth rates along the trajectory of aerosol parcels. This variation is due to differential diffusion effects between aerosol particles and gas-phase scalars, so that aerosol particles drift in mixture fraction space. The drift in the turbulent flow field is investigated quantitatively using the Lagrangian statistics of the material derivative of mixture fraction. The occurrence of drift against mean mixing is key in explaining the presence of aerosol droplets on the hot and humid side of the mixing layer, where the rate of particle growth is highest. Thus, the spatial distribution of condensing aerosols in turbulent flows is shown to be fundamentally different from that found in laminar flows, which lack fluctuating mixing fields.

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