International Symposium
On Turbulence and Shear Flow
Phenomena (TSFP-9)
June 30 - July 3, 2015 *Melbourne, Australia*

SIMULATION OF A CONDENSING AEROSOL IN HOMOGENEOUS ISOTROPIC TURBULENCE

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ABSTRACT

The nucleation, growth, and coagulation of liquid droplets in three-dimensional homogeneous isotropic turbulence at $\mathrm{Re}_\lambda \approx 150$ is simulated. Patches of dry and cold gas mix with patches of hot gas saturated with vapor of a condensable species, inducing nucleation of particles due to supersaturation. The simulation consists of a three-dimensional direct numerical simulation of homogeneous isotropic turbulence with a statistically stationary forced velocity field. All length and time scales of fluid motion and scalar mixing are resolved adequately. For the droplets phase, a model based on the quadrature method of moments and a Lagrangian scheme for the solution of the moment transport equations are employed. Results show that droplets form early in the evolution of the flow field and their concentration peaks on the cold side of the mixing layers separating the patches of hot and cold gas, where droplets nucleate most intensely. Conversely, the droplets grow most rapidly on the hot side of the mixing layers. As turbulent mixing displaces the droplets into regions of hot and moist gas, the droplets' size increases markedly. Conditional statistics of the aerosol phase in the mixture fraction space are employed to investigate this trend.

INTRODUCTION

The formation of aerosol particles in turbulent, spatially inhomogeneous flows is a fundamental process in nature and is relevant in many technological applications. These include cloud formation (Pruppacher & Klett, 2010), the production of advanced powders (Kodas & Hampden-Smith, 1998), and soot formation (Attili *et al.*, 2014).

In this work, we consider a flow configuration in which aerosol particles form (nucleate) from a supersaturated vapor and supersaturation is induced by the turbulent mixing of two streams (a saturated stream and a cold one). As the rates of nucleation and other aerosol processes are highly sensitive to the scalar fields, turbulence has a strong effect on aerosol dynamics. In the present work, a simulation of the formation and evolution of a condensing aerosol in homogeneous isotropic turbulence is performed and analyzed. The aerosol evolves due to the formation, growth, and coagulation of dibutyl phthalate (DBP) droplets. This investigation is an extension of a previous work by the group, where aerosol dynamics were simulated in a turbulent spatially-evolving mixing layer (Zhou *et al.*, 2014). In this work, the homogeneous isotropic turbulence configuration is selected to identify the Reynolds number of the flow unequivocally, thereby facilitating parametric studies. Moreover, the flow field is characterized by well-defined time scales, e.g. the eddy turnover time, which may be compared to the time scales of the processes responsible for the formation and growth of the aerosol.

FORMULATION

Simulation of the Gaseous Phase

A direct numerical simulation (DNS) is employed for the simulation of velocity, temperature, and vapor mass fraction fields. The incompressible Navier-Stokes equations are solved with a finite difference method on a spatially and temporally staggered grid with the semi-implicit fractional step method of Kim & Moin (1985). Velocity is linearly forced using the method of Rosales & Meneveau (2005) in order to achieve a statistically-stationary velocity field. The transport equations of the scalars (temperature and mass fraction of DBP vapor) are discretized using the third-order WENO scheme (Liu *et al.*, 1994). The computational domain of size $L = 6.25\,\mathrm{mm}$ is discretized into 134 million grid points (512^3). This results in a grid size $\Delta x = 11.9\ \mu\mathrm{m}$ smaller than the Kolmogorov length scale $\eta = 12.5\ \mu\mathrm{m}$. The solution is advanced with a time step $\Delta t = 0.5\ \mu\mathrm{s}$, maintaining a unity CFL condition. The parallel flow solver "NGA" (Desjardins *et al.*, 2008) developed at Stanford University is used to solve this system.

Due to the small size of droplets and low volume load, the effects of aerosol phase on the momentum and the temperature of the gaseous phase are neglected. On the other

hand, the effect on the mass fraction of the vapor is considered, i.e.,

$$\frac{\partial Y}{\partial t} + \vec{u}\cdot\nabla Y - D\nabla^2 Y = S_{\text{nuc.}} + S_{\text{grow}}, \qquad (1)$$

where, $S_{\text{nuc.}}$ and S_{grow} account to for gas-to-particle conversion rates due to the nucleation and growth by condensation, respectively. The simulation is carried out with unity Lewis and Schmidt numbers, and constant density.

Simulation of the Aerosol Phase

All droplets are assumed to be spheres with a diameter ξ. Thus the aerosol phase, at some point in time t and space $\vec{x}$, is described by the particle size distribution (PSD) function $n(\vec{x},t;\xi)$, for which the raw moments $\mu_k(\vec{x},t)$ are defined as:

$$\mu_k(\vec{x},t) = \int_0^\infty \xi'^k n(\vec{x},t;\xi')d\xi'. \qquad (2)$$

The Quadrature Method of Moments (QMOM) (McGraw, 1997) is used to describe of the aerosol phase. The transport equations for the moments of the aerosol distribution are:

$$\frac{\partial \mu_k}{\partial t} + \nabla\cdot\mu_k\vec{u} = \left(\frac{\partial n}{\partial t}\right)_{\text{nuc}} + \left(\frac{\partial n}{\partial t}\right)_{\text{cond}} + \left(\frac{\partial n}{\partial t}\right)_{\text{coag}}, \qquad (3)$$

where, the three terms on the right-hand-side of the equation represent aerosol dynamics (nucleation, growth by condensation, and coagulation). The diffusion term is neglected due to the large Schmidt number of aerosol particles. For example, under standard atmospheric conditions, spherical particles with a diameter of 10 nm (100 nm) have a Schmidt number equal to 290 (2.2×10^4). The nucleation rate is modeled using the self-consistent correction to the classical nucleation theory (Girshick & Chiu, 1990), the growth rate of droplet by condensation is modeled by the generalized Mason's formula (Loyalka & Park, 1988), and the coagulation kernel is modeled by the Fuchs model (Fuchs, 1964).

The Lagrangian Particles Scheme

The transport of the aerosol moments is performed with a Langrangian particles method (Koumoutsakos, 2005; Attili & Bisetti, 2013). The Lagrangian scheme overcomes moments realizability problems encountered in Eulerian frameworks (McGraw, 2012; Wright, 2007). Additionally, the Lagrangian scheme has low numerical diffusion. This makes it easy to track the sharp front of the motion of aerosol particles, and makes it an ideal tool to investigate the differential diffusion of aerosol particles and the gas.

In the Lagrangian particles scheme, the movement of a large number of Lagrangian particles is tracked (a Lagrangian particle refers to a mathematical object rather than a physical aerosol droplet). Physical variables of interest (aerosol moments here) are tied to these Lagrangian particles and evolve along the Lagrangian trajectories. Within this framework, the conservation equationsare separated into two parts, convection and unsteady evolution.The convection is dealt with through an averagescheme over particles, and the unsteady evolution ofthe variables along a trajectory is obtained by solving thecorresponding control equations,which are only ordinary differential equations (Attili & Bisetti, 2013).

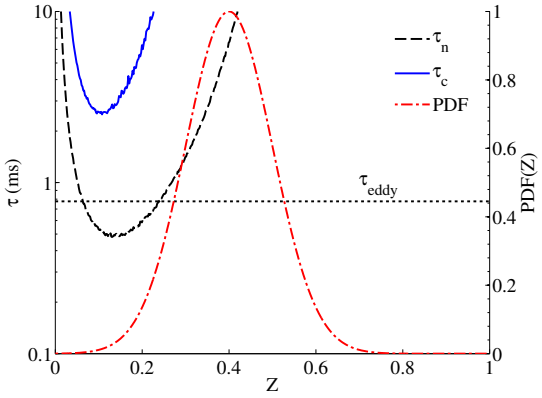


Figure 1: Distributions of the time scales (τ_n, τ_c, and τ_{eddy}), the initial PDF of Z in mixture fraction space.

The domain is populated with 1.3 billion Lagrangian particles (an average of ten particles per cell). The simulation is performed on the IBM Blue Gene/P supercomputer, "Shaheen", available at King Abdullah University of Science and Technology, using 8,192 cores. Time advancement over one eddy turnover time requires 1.8 million core hours.

RESULTS

Mixture fraction is related to temperature T and DBP mass fraction (in the absence of consumption) as:

$$T = T_c + Z(T_h - T_c), \qquad (4)$$

and

$$Y = Y_h Z, \qquad (5)$$

where $T_c = -20\,^\circ$C and $T_h = 112\,^\circ$C are the temperatures of the cold and hot streams being mixed, respectively. $Y_h = 1.43\times10^{-3}$ is the mass fraction of DBP vapor in the hot stream corresponding to a fully saturated state.

In order to characterize the evolution of the aerosol phase, we introduce two relevant quantities, τ_n and τ_c, which represent the time scales for nucleation and coagulation. Estimates for these two time scales are obtained by evolving the aerosol in the absence of gas-phase mixing, effectively considering an ensemble of zero-dimensional reactors at various conditions, but accounting for the consumption of gas-phase condensable vapor. In particular, τ_n is the time for number density to reach the peak in the reactor, and τ_c is the time needed for coagulation to decrease number density back to half of the peak value. By virtue of the strong dependence of nucleation rates on the gas composition and temperature, τ_n and τ_c are smallest at the location of peak nucleation rate at $Z = Z_m = 0.11$. This is shown in fig. 1, where the minimum values are $\tau_n = 0.5$ ms and $\tau_c = 2.5$ ms for the present streams.

The length and velocity scales of fluid motion and mixing are chosen to have a significant overlap between the

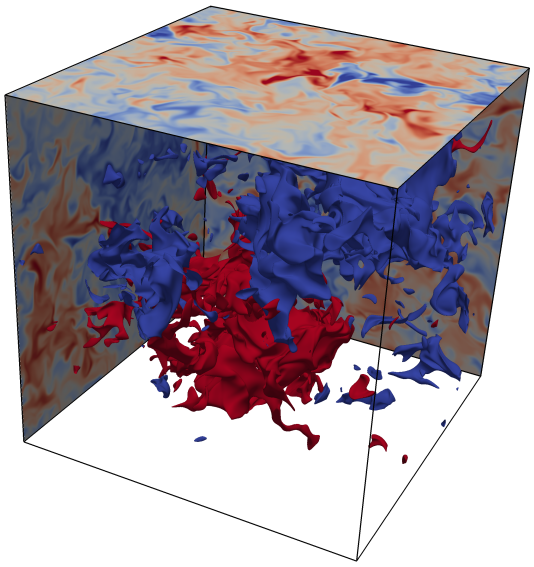


Figure 2: The initial distribution of the mixture fraction Z. The blue isosurface marks $Z = 0.15$ and the red isosurface marks $Z = 0.65$.

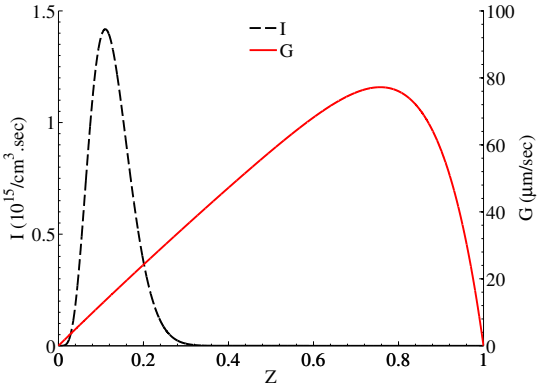


Figure 3: Distribution of the nucleation rate, and growth rate in the free molecular regime in mixture fraction space.

aerosol and the turbulent timescales. The choice of the stream conditions is such that τ_n and τ_c at $Z = Z_m = 0.11$ are of the same order. The Reynolds number based on the Taylor scale is $Re_\lambda = 150$. The Taylor scale is $\lambda = 0.28$ mm, the eddy turnover time scale is $\tau_{\text{eddy}} = 0.78$ ms, and the Kolmogorov time scale is $\tau_\eta = 11.3\ \mu$s.

The mixture fraction at the onset of the simulation is a rescaled snapshot obtained from a previous simulation of a decaing scalar after reaching a self-similar state, i.e., a Gaussian distribution (Warhaft, 2000). The initial distribution of the temperature and the mass fraction of DBP vapor are mapped from Z with a mean value of 0.4 and a standard deviation of 0.1 (see fig. 2) according to Eq. (4) and Eq. (5).

Nucleation is maximum on the cold side at $Z = 0.11$, whereas the growth rate of particles increases on the hot side and peaks at $Z = 0.76$ as shown in fig. 3. Thus, droplets nucleate on the cold side near the peak nucleation rate location first.

By virtue of temperature being a conserved scalar, the

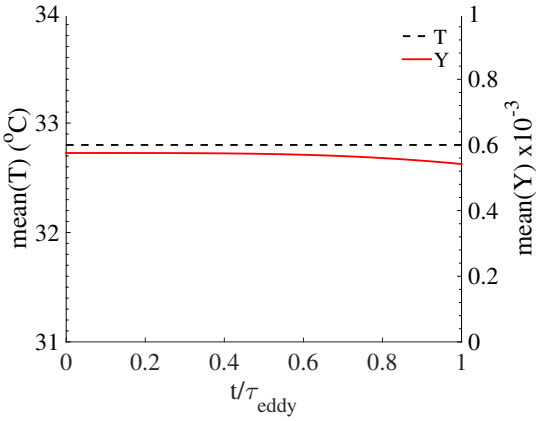


Figure 4: Time evolution of the domain average temperature and average vapor mass fraction.

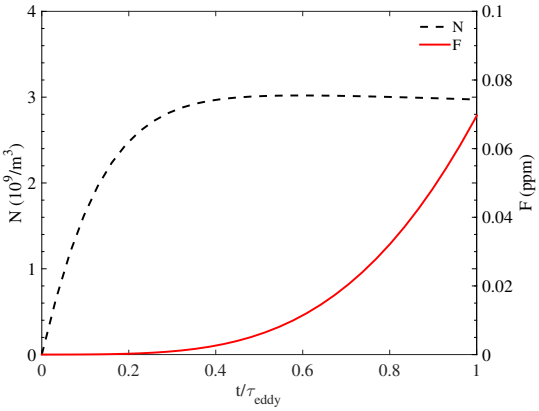


Figure 5: Time evolution of the domain averaged number density and volume fraction.

mean temperature in the domain remains constant throughout the simulation. Conversely, after evolving the system for one eddy turnover timescale, a slight decrease is observed in the mean vapor mass fraction due to the conversion into the liquid phase as shown in fig. 4.

At the beginning of the simulation, the domain-averaged number density increases linearly in time until $t \approx 0.3\tau_{\text{eddy}}$. After that, number density saturates because of nucleation-suppression by mixing, a process explained as follows. Due to the diffusion process, mixing drives mixture fraction towards its mean value, and the probability of finding a fluid parcel with high nucleation rate vanishes. On the other hand, volume fraction keeps on growing as shown in fig.5.

Unlike the scalar fields, the transport of droplets is characterized by a very large Schmidt number, i.e., droplets are convected along the pathlines, whereas the transport of the scalar fields (T and Y) includes diffusion processes. The resulting differential diffusion between droplets and the scalar fields causes a drift of the droplets in mixture fraction or composition space (Zhou *et al.*, 2014). This drift moves droplets from the location where they nucleate to the warmer regions of the domain where the growth rate of the droplets increases, and droplets grow in size. Nucleation is confined in the region $0 \leq Z \leq 0.3$, and so is number density as shown in fig. 6. The effect of drift is shown in fig. 7,

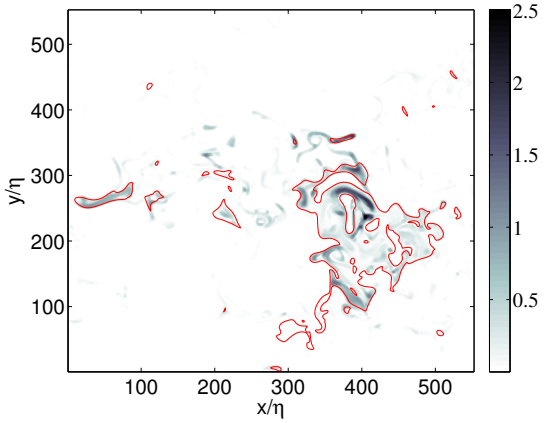


Figure 6: Number density ($10^{11}/\mathrm{cm}^3$) in a selected plane at $t = 0.3\,\tau_{\mathrm{eddy}}$. The red isocontours mark $Z = 0.3$.

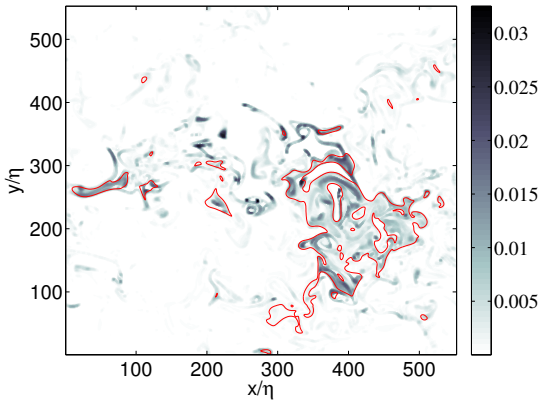
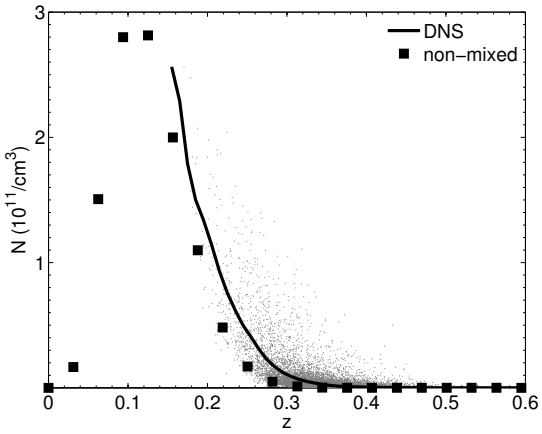


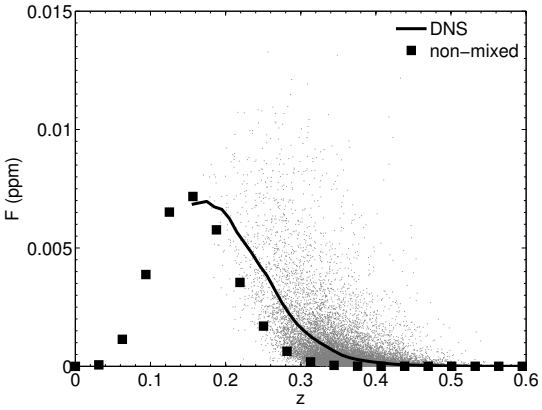
Figure 7: Volume fraction (ppm) in a selected plane at $t = 0.3\,\tau_{\mathrm{eddy}}$. The red isocontours mark $Z = 0.3$.

where the volume fraction of the liquid phase is shown. Although number density is mostly confined in the region of the flow where droplets nucleate, droplets that drift towards the warm and humid regions of the flow (larger values of mixture fraction) grow fastest creating peaks of volume fraction.

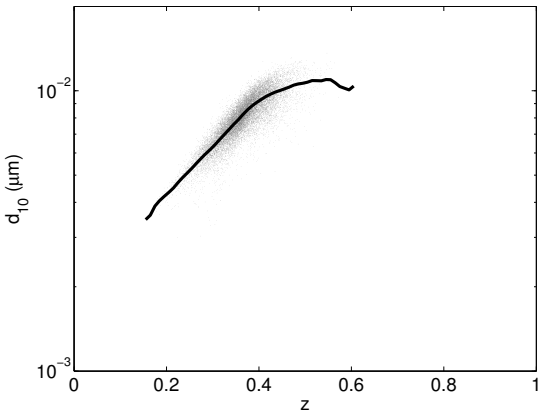
The effect of drift can be observed through the conditional statistics of aerosol quantities in mixture fraction space as shown in fig. 8. Prediction of the number density and volume fraction can be obtained by evolving the droplets without transport (as explained in the discussion for τ_n and τ_c). These results are compared with the conditional means computed from the DNS. As shown, the non-mixed estimates are a lower bound for number density and volume fraction, i.e., turbulent mixing results in droplets drifting from the region of maximum nucleation rate towards the warmer regions of the flow field. Although the location of peak conditional mean volume fraction is close to that of number density, peak volume fraction is shifted towards the hot stream side. Moreover, due to the mixing with the mean value, the variance of mixture fraction has dropped by a factor of two, and the smallest mixture fraction value increased to $Z = 0.15$.



(a) Number density



(b) Volume fraction



(c) Mean diameter

Figure 8: The scatter, the conditional mean, and the non-mixed estimate of (a) number density N, (b) volume fraction F, and (c) mean counter diameter ($d_{10} = \mu_1/\mu_0$) of droplets in mixture fraction space at $t = 0.3\,\tau_{\mathrm{eddy}}$.

CONCLUSION

The nucleation, growth, and coagulation of liquid droplets in three-dimensional homogeneous isotropic turbulence at $\mathrm{Re}_\lambda \approx 150$ was investigated. A DNS was employed for the simulation of the gaseous phase along with the QMOM for the aerosol phase. A Lagrangian particles scheme was used to transport the moments of PSD. The simulation showed that nucleation is suppressed by mix-

ing within 0.3 eddy turnover times. Droplets nucleate initially at the cold side of domain ($Z < 0.3$). Drift, driven by differential diffusion between droplets and the scalar fields, moves some droplets to warmer regimes of the domain where growth is larger. This creates peaks of volume fraction and average droplet size outside the nucleation zone.

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