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Formation, growth, and transport of soot in a three-dimensional turbulent non-premixed jet flame

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ABSTRACT

The formation, growth, and transport of soot is investigated via large scale numerical simulation in a three-dimensional turbulent non-premixed *n*-heptane/air jet flame at a jet Reynolds number of 15,000. For the first time, a detailed chemical mechanism, which includes the soot precursor naphthalene and a high-order method of moments are employed in a three-dimensional simulation of a turbulent sooting flame. The results are used to discuss the interaction of turbulence, chemistry, and the formation of soot. Compared to temperature and other species controlled by oxidation chemistry, naphthalene is found to be affected more significantly by the scalar dissipation rate. While the mixture fraction and temperature fields show fairly smooth spatial and temporal variations, the sensitivity of naphthalene to turbulent mixing causes large inhomogeneities in the precursor fields, which in turn generate even stronger intermittency in the soot fields. A strong correlation is apparent between soot number density and the concentration of naphthalene. On the contrary, while soot mass fraction is usually large where naphthalene is present, pockets of fluid with large soot mass are also frequent in regions with very low naphthalene mass fraction values. From the analysis of Lagrangian statistics, it is shown that soot nucleates and grows mainly in a layer close to the flame and spreads on the rich side of the flame due to the fluctuating mixing field, resulting in more than half of the total soot mass being located at mixture fractions larger than 0.6. Only a small fraction of soot is transported towards the flame and is completely oxidized in the vicinity of the stoichiometric surface. These results show the leading order effects of turbulent mixing in controlling the dynamics of soot in turbulent flames. Finally, given the difficulties in obtaining quantitative data in experiments of turbulent sooting flames, this simulation provides valuable data to guide the development of models for Large Eddy Simulation and Reynolds Average Navier Stokes approaches.

1. Introduction

Soot emissions are an undesirable byproduct of rich combustion in technical combustion devices such as internal combustion engines, jet engines, and industrial burners [1]. Since soot poses important health and environmental hazards [2,3], there exists significant interest in improving our understanding of the details of its formation and growth with the aim of reducing harmful emissions. Recently, emphasis has been placed on the characterization of toxicity, particulate size distribution, and surface fraction of soot emissions from newly introduced combustion devices [4].

Studies addressing the formation and growth of soot particulate in flames have mostly focused on laminar configurations, for which well-controlled flow conditions enable detailed measurements of soot mass, number density, size distribution, and morphology. These studies account for most of the current understanding of soot processes, i.e., nucleation, growth, and oxidation [5]. Due to important complications in methods and diagnostics brought by the unsteady, three-dimensional turbulent flow field, soot formation in turbulent flames has received significantly less attention compared to laminar flames. As highlighted by recent works on turbulent sooting flames [6–13], there are reasons to believe that soot formation and growth are strongly affected by turbulence; therefore, Direct Numerical Simulation (DNS) studies such as the one presented in this work are key to furthering our understanding of soot formation in practical combustion devices which operate in the turbulent flow regime.

Turbulent combustion DNS coupled with models for soot formation have been employed successfully to investigate the evolution of soot in turbulent flames. Examples include the study of soot formation in turbulent ethylene–air counterflow diffusion flames

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[7], soot-flame interaction and soot transport in two- and three-dimensional temporally evolving ethylene–air jet flames [8,9], and flame weakening and extinction leading to soot leakage in a two-dimensional convection-driven, turbulent wall-flame [10]. Bisetti et al. [12] analyzed the formation and early evolution of soot in a two-dimensional, turbulent *n*-heptane/air flame. Their work [12] distinguishes itself from previous studies [7–9] by considering finite rate chemistry describing the formation of polycyclic aromatic hydrocarbons (PAH). Moreover, a detailed soot model based on elementary physical processes and rates [14] is used rather than a semi-empirical approach [15,16].

In Ref. [12], a planar, two-dimensional, temporally evolving *n*-heptane/air flame subject to isotropic, decaying turbulence is considered. The main findings of the study pertain to the role of turbulent mixing in affecting (a) soot formation and growth rates and (b) soot displacement across the flame. Firstly, it was found that the local concentration of soot precursors (PAH) is very sensitive to scalar dissipation rate. Therefore, the statistics of turbulent mixing at the small scales have an impact on soot formation and growth rates. In regions of high dissipation rate, PAH concentrations decrease by several orders of magnitude, and soot growth is suppressed. Conversely, in regions of low dissipation rate, growth is promoted. It was noted that the appearance of "islands" of soot in regions of low scalar dissipation rate, such as the central portion of large rollers in shear flows, is in qualitative agreement with experimental evidence in sooting turbulent flames [17,18]. These conclusions are in agreement with experimental observations in laminar counterflow flames [19], where it is shown that the concentration of soot precursors decreases severalfold as the global strain increases, even though the flame is far from extinction. Secondly, it was observed [12] that soot drifts in mixture fraction space due to its high Schmidt number and significantly lower mass diffusivity compared to gas-phase scalars. The drift of soot in mixture fraction space has important implications for the time-evolution of soot number density, volume fraction, and morphology as soot parcels meander across the turbulent flame encountering regions of temperature and composition that promote (or suppress) soot growth. The statistics of soot drift are connected tightly to the formation and destruction of thin layers of dissipation rate.

Although significant, the results discussed in Ref. [12] were mostly qualitative and supported only a preliminary assessment of the mechanisms responsible for turbulence-chemistry-soot interaction. In particular, two-dimensional flow configurations do not justify in-depth, quantitative investigations into flow statistics given the unphysical nature of two-dimensional turbulence. For example, it is well known that the small scale statistics are qualitatively and quantitatively different in two- and three-dimensional turbulence [20–22]. Given the important role played by small scale mixing in controlling soot evolution, the results in Ref. [12] require further analysis in a three-dimensional flow.

In the present work, the methods and models in Ref. [12] are applied to the simulation of soot formation in a three-dimensional, temporally evolving turbulent jet flame with the aim of investigating soot statistics quantitatively. Important questions on the interaction of turbulent mixing, chemistry of soot precursors, and soot formation and growth processes are addressed. Differently from our previous work, Lagrangian statistics, rather than statistics based on Eulerian fields, are employed to characterize soot dynamics. Lagrangian statistics are available as a byproduct of the Lagrangian particle method adopted for the transport of soot variables in the context of the method of moments [23]. The Lagrangian statistics prove invaluable to explaining key trends in the overall evolution of the soot mass fraction and number density.

2. Physical models and numerical methods

2.1. Models

The gas phase hydrodynamics are modeled with the reactive, unsteady Navier–Stokes equations in the low Mach number limit [24]. The transport of species mass fractions is described using the Hirschfelder and Curtiss approximation to the diffusive fluxes [25] together with a velocity-correction approach [26] to enforce mass conservation. Soret and Dufour effects are neglected. The species mass fraction and temperature equations read

$$\frac{\partial \rho Y_i}{\partial t} + \nabla \cdot (\rho \mathbf{u} Y_i) = -\nabla \cdot (\rho Y_i \mathbf{V}_i) + \dot{\omega}_{Y_i} \tag{1}$$

and

$$c_p \left[\frac{\partial \rho T}{\partial t} + \nabla \cdot (\rho \mathbf{u} T) \right] = \nabla \cdot (\lambda \nabla T) - \sum_{i=1}^{M} c_{p,i} (\rho Y_i \mathbf{V}_i) \cdot \nabla T + \dot{\omega}_T. \tag{2}$$

The mass-based species velocity $\mathbf{V}_i$ is defined as

$$\mathbf{V}_i = -D_i \frac{\nabla X_i}{X_i}, \tag{3}$$

where X_i and D_i are the mole fraction and mixture-average diffusion coefficient of species i, respectively. The thermal conductivity is λ; c_p is the mixture specific heat at constant pressure; and $c_{p,i}$ are the specific heats for each species. The terms $\dot{\omega}_{Y_i}$ and $\dot{\omega}_T$ indicate source terms for species mass fractions and temperature, respectively.

Combustion is modeled using a reduced mechanism for the oxidation of *n*-heptane comprising 47 species and 290 reactions. The reduced mechanism is based on the detailed mechanism developed by Blanquart et al. [27] for the high-temperature combustion of engine relevant fuels with emphasis on the prediction of soot precursors. The main pathways to the formation of naphthalene (A2) are retained in the reduction procedure in order to describe soot formation and growth. The details on the mechanism reduction and various validation cases relevant to *n*-heptane oxidation and benzene formation are available in Bisetti et al. [12].

Soot particles and aggregates are described by their volume (V) and surface area (S) [28], and a bivariate moment method is adopted to describe the evolution of soot particulate. The bivariate soot moment $M_{x,y}$ (in units of concentration) is defined as

$$M_{x,y} = \sum_j V_j^x S_j^y N_j, \tag{4}$$

where x and y are the moment orders for volume and surface, and V_j, S_j, and N_j are the volume, surface area, and number density of soot aggregates belonging to size class j. Moments evolve according to

$$\frac{\partial M_{x,y}}{\partial t} + \nabla \cdot (\mathbf{u} M_{x,y}) = \dot{M}_{x,y}, \tag{5}$$

where $\dot{M}_{x,y}$ is a source term describing aerosol internal processes. Soot transport is characterized by a high Schmidt number (low diffusivity) and diffusive mass fluxes are therefore neglected. As shown by Lignell et al. [29] and Bisetti et al. [12], thermophoretic effects [30] are also negligible in sooting turbulent nonpremixed jet flames. Given the small size of the flame, the short physical time simulated, and the limited soot loading, the effects of gas-phase and soot radiation are small [31] and are neglected in the model.

As for all moment methods, the rate of change of a given moment depends on additional moments, and a closure scheme is necessary. In this work, the Hybrid Method of Moments (HMOM) closure [32] is used with seven statistical moments for the soot population: $M_{0,0}, M_{0,1}, M_{0,2}$, $M_{1,0}, M_{1,1}, M_{2,0}$, and N_0. The moment N_0, representing the number density of very small particles, is also