

Simulation and modeling of the behavior of conditional scalar moments in turbulent spray combustion

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A series of direct numerical simulations (DNS) have been conducted to examine the mixing and reaction statistics of spray combustion in forced isotropic turbulence. Particular attention has been given to statistics conditioned on mixture fraction to determine the modifications required for mixture-fraction based gas-phase combustion models. The goal is to apply these models in the presence of evaporating fuel droplets.

1. Introduction

Turbulent combustion in the gas phase is a complex phenomenon which has been the subject of a large body of research. However, even given an appropriate model for the treatment of turbulent gas-phase combustion, there are additional important physical processes which must be addressed if a practical predictive capability is to be derived.

A significant aspect of many combustion systems of practical interest is the presence and behavior of liquid fuel droplets. The strong influence that this condensed-phase species has upon practical combustion modeling is profound. Through the close interaction with the surrounding gas phase, the droplets influence the location, structure, and thermochemical yield of turbulent combustion zones within practical devices.

1.1. Mixture fraction in a multiphase system

When modeling turbulent nonpremixed combustion, it is common to employ a chemically conserved scalar, usually referred to as mixture fraction, as a coordinate for the computation of reactive scalar behavior. Examples of mixture fraction based models include the nonpremixed combustion versions of the steady and unsteady laminar flamelet methods of Peters (1984), Peters (1986) and coworkers, and the many variants of the conditional moment closure (CMC) method proposed by Klimenko (1990), Bilger (1993) and coworkers.

For pure gas-phase combustion, mixture fraction can be defined as an appropriate linear combination of reactive (and inert) species mass fractions such that it has no chemical source term. Since mixture fraction is defined as being conserved under chemical reaction, it is solely a measure of the fraction of mass present that originated from one of the two mixing streams. As such, its value is only subject to change due to mixing. This makes it an effective coordinate in which to solve conditionally averaged reactive scalar equations free from large mixing-induced fluctuations.

For liquid fuel combustion, however, mixture fraction is an ambiguous concept, and there are a number of alternatives for the definition. These alternatives result from 1) the

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question of whether to define it to be invariant under phase reaction or not, and 2) the spatial scale at which the definition is applied. If a mixture fraction ϕ is defined in terms of the gas-phase species alone on a spatial scale that is large compared to the mean free path length of the gas molecules but small compared to the interdroplet spacing, then its conservation equation is

$$\frac{\partial \phi}{\partial t} + u_i \frac{\partial \phi}{\partial x_i} = \frac{\partial}{\partial x_j} \left(\mathcal{D}_\phi \frac{\partial \phi}{\partial x_j} \right), \quad (1.1)$$

where u_i denotes local velocity and \mathcal{D}_ϕ the molecular diffusivity of the scalar. The boundary condition at the irregular gas-liquid interface is given by

$$\left[\rho \mathcal{D}_\phi \frac{\partial \phi}{\partial n} \right]_{surf} = \dot{m} (f_{surf} - f_{liq}), \quad (1.2)$$

where \dot{m} is the mass evaporation rate at that surface, n is the surface normal coordinate, and f_{surf} and f_{liq} are the values of mixture fraction at the surface and in the liquid-phase. Klimenko and Bilger (1998) provide plausible probability density functions (PDFs) and scalar dissipation rate profiles for this mixture fraction. These are derived from scaling arguments. Due to the overwhelming computational burden of resolving droplet surfaces in a reacting turbulent simulation, it is not feasible to directly examine the dynamics of ϕ in this study. Here, analysis was limited to examining fields with scales of variation which were commensurate with those of the turbulence simulations.

If a mixture fraction denoted as ξ is defined in terms of the gas-phase species alone on a spatial scale that is large compared to mean free path lengths of gaseous molecules and interdroplet spacing so that it is a spatial average $\xi \equiv \{\phi\}$ of that given above, then its conservation equation is

$$\frac{\partial \xi}{\partial t} + u_i \frac{\partial \xi}{\partial x_i} = \frac{\partial}{\partial x_j} \left(\mathcal{D}_\xi \frac{\partial \xi}{\partial x_j} \right) + \dot{s}_\xi, \quad (1.3)$$

where \dot{s}_ξ is an evaporative source term which represents the average flux of gaseous mass from the droplets, which exist on a spatial scale below the resolution of the equation.

At the spatial scale of the definition of ξ , it is, of course, possible to define a mixture fraction z which includes the mass associated with the droplets and the gas-phase alike,

$$z \equiv (1 - \alpha) \xi + \alpha, \quad (1.4)$$

where α is the mass fraction of the mixture which is associated with the liquid-phase. If α is assumed to be much smaller than unity, then $z \approx \xi + \alpha$ and the following is true,

$$\frac{\partial z}{\partial t} + u_i \frac{\partial z}{\partial x_i} = \frac{\partial}{\partial x_j} \left(\mathcal{D}_z \frac{\partial z}{\partial x_j} \right) + \dot{s}_z, \quad (1.5)$$

where the source term \dot{s}_z is due to the strong differential diffusion between the different components of z . Here an explicit evaporative term is avoided. Note that in the above, slip velocities between the two phases have been assumed to be negligible. This is consistent with small ‘‘flow following’’ droplets.

One can pose the question as to which definition of mixture fraction provides the most convenient set of equations for use in the context of mixture fraction based nonpremixed combustion models. Klimenko and Bilger (1998) advocate the simultaneous use of two mixture fractions ϕ and z in a doubly-conditioned CMC methodology, while Réveillon and Vervisch (1998) employed the ξ mixture fraction in a study of unconditional mean evaporation and mixing statistics.

The level of resolution of the simulations conducted during the Summer Program made it possible to examine the statistics for ξ and z , but not those of ϕ . Due to space constraints, this report is limited to the analysis of ξ mixture fraction and modeling based on this quantity. Note however, that it seems to be possible to treat the differential diffusion associated with \dot{s}_z in the z equation using the methodology of Smith, *et al* (1998) and Smith (1999).

1.2. Conditionally averaged statistics

The form of the instantaneous local equation (Eq. 1.3) for the coarse scale gas-phase mixture fraction ξ can be used to derive the corresponding PDF transport equation,

$$\frac{\partial P_\eta}{\partial t} + \frac{\partial}{\partial x_i} (\langle u_i | \eta \rangle P_\eta) = -\frac{\partial^2}{\partial \eta^2} (N_\eta P_\eta) - \frac{\partial}{\partial \eta} (\langle \dot{s}_\xi | \eta \rangle P_\eta) \quad (1.6)$$

where P_η is the mixture fraction PDF, and N_η denotes the conditional mean scalar dissipation rate which is given by

$$N_\eta \equiv \mathcal{D}_\xi \langle (\nabla \xi)^2 | \eta \rangle . \quad (1.7)$$

The shorthand $\langle \dots | \eta \rangle$ construction denotes the average of the argument taken over all samples where the condition $\xi = \eta$ is met. It can be shown that for the conditional mean mass fraction ($Q_\eta \equiv \langle Y | \eta \rangle$) of any reactive species, the corresponding conditional mean conservation equation can be written as

$$\frac{\partial Q_\eta}{\partial t} + \langle u_i | \eta \rangle \frac{\partial Q_\eta}{\partial x_i} = \langle \dot{\omega}_r | \eta \rangle + N_\eta \frac{\partial^2 Q_\eta}{\partial \eta^2} - \langle \dot{s}_\xi | \eta \rangle \frac{\partial Q_\eta}{\partial \eta} \quad (1.8)$$

where $\dot{\omega}_r$ is the net chemical production rate of mass fraction for the reactive species. It is clear from the equation above that the evaporative source term \dot{s}_ξ plays a role in a pseudo-convective process in mixture fraction space for both the PDF and conditional moment equations.

One of the principal aims of the study was to examine the shape and magnitude of the conditional mean source term $\langle \dot{s}_\xi | \eta \rangle$ as a function of mixture fraction for different spray combustion cases. It was hoped that sufficient knowledge of $\langle \dot{s}_\xi | \eta \rangle$ could be gained from the simulation to devise an adequate model for the term from known quantities. It was further hoped that this model would allow the above reactive scalar equation to be solved in order to predict the conditional mean evolution of a smoke-like species in the simulations.

2. Simulation conditions

Many different modes of spray combustion exist. Group combustion, where a diffusion flame envelopes large groups of evaporating drops rather than individual droplets, is the most common in gas turbine applications (Kuo (1986)). The DNS were designed to embody the basic physical features of group combustion in the simplest possible flow and mixing configuration.

The DNS involved the Lagrangian tracking of small inertial droplets in forced isotropic incompressible turbulence on a 64^3 grid with a constant Taylor Reynolds number of ~ 50 . In each simulation, the droplets were initially of uniform size and organized in a uniformly random distribution within a spherical cloud with a diameter equal to half the edge length of one cube of the periodic cubic domain. During the course of each simulation,

the droplets were redistributed throughout the domain, as a result of turbulent fluid motion, while evaporating to produce gaseous fuel in the turbulent fluid.

The droplets were defined to be sufficiently small so as to be in thermodynamic equilibrium with their surroundings so that their individual evaporation rates could be determined from

$$\frac{dm^k}{dt} = \pi \rho_{gas} \nu_{gas} D^k \frac{\mathcal{S}_h^k}{\mathcal{S}_c} \ln(1 + B^k) \quad , \quad (2.1)$$

where ν_{gas} is the gas kinematic viscosity, D^k is the droplet diameter, \mathcal{S}_h^k is the droplet Sherwood number, \mathcal{S}_c is the Schmidt number of the evaporating species in the gas-phase, and B^k is the local instantaneous Spalding transfer number of the droplet. The Sherwood number of the droplet can be approximated by

$$\mathcal{S}_h^k \approx 2 + 0.6 \left((\mathcal{R}_e^k)^{1/2} \mathcal{S}_c^{1/3} \right) \quad , \quad (2.2)$$

where \mathcal{R}_e^k is the droplet Reynolds number. The mass contributions from each of the droplets were summed at each step in the simulation to determine the mixture fraction source term \dot{s}_ξ which is required for the solution of Eq. 1.3.

The Spalding transfer number was set to vary linearly with temperature, with values ranging between 3.4 and zero. The peak transfer number value was chosen to correspond with kerosene droplets evaporating within enveloping flames. The corresponding gas-phase stoichiometric mixture fraction ($\xi_s = 0.0625$) of kerosene was employed to specify the location of the peak transfer number in mixture fraction space. To account for individual droplets burning in fuel-lean regions, the transfer number B was set equal to the maximum value for all lean local mixture fractions ξ and was set to vary between zero and the maximum only on the rich side of stoichiometric.

Combustion was simulated using a *fast chemistry* approach so that all properties of the reacting mixture except the concentration of a minor pollutant were functions of the local instantaneous gaseous mixture fraction ξ , which was in turn governed by Eq. 1.3. The instantaneous local equation governing the evolution of the pollutant species mass fraction Y can be written as

$$\frac{\partial Y}{\partial t} + u_i \frac{\partial Y}{\partial x_i} = \frac{\partial}{\partial x_j} \left(\mathcal{D}_Y \frac{\partial Y}{\partial x_j} \right) + \dot{\omega}_r \quad , \quad (2.3)$$

where the $\dot{\omega}_r$ denotes the net chemical production rate, and \mathcal{D}_Y denotes the molecular diffusivity, which in practice was set equal to the diffusivity of mixture fraction ($\mathcal{D}_Y = \mathcal{D}_\xi$).

The purpose of including a non-equilibrium minor pollutant in each of the simulations was to provide a reactive scalar against which CMC model predictions could be assessed. The chemical behavior of the pollutant was defined so as to mimic soot in hydrocarbon flames. It was set to be produced under hot fuel-rich conditions and eliminated under hot fuel-lean conditions. The instantaneous local chemical production rate of the pollutant was given by

$$\dot{\omega}_r = k_1(\xi) - k_2(\xi) \frac{Y}{Y_{eq}} \quad , \quad (2.4)$$

where Y_{eq} is a constant that determines the approximate magnitude of the equilibrium pollutant concentration. The terms k_1 and k_2 are rate coefficient functions of mixture

Case	n_d	r_D	f_v	$\langle z \rangle / z_s$	r_τ	G
2	1.e5	0.035	1.e-3	1.0	0.071	52100
4	1.e5	0.027	5.e-4	0.5	0.045	40200
5	1.e4	0.027	5.e-4	0.5	0.045	4020
6	1.e4	0.035	1.e-3	1.0	0.071	5210

TABLE 1. Characteristic parameters for spray combustion simulations. See text for symbol definitions.

fraction defined by

$$k_1(\xi) \equiv \begin{cases} k, & \text{for } \xi_s < \xi < 5\xi_s \\ 0, & \text{otherwise} \end{cases} \quad (2.5)$$

$$k_2(\xi) \equiv \begin{cases} k, & \text{for } 0 < \xi < \xi_s \\ 0, & \text{otherwise} \end{cases} . \quad (2.6)$$

Here, k is an arbitrary rate constant with the value of the stoichiometric mixture fraction set to the same value specified earlier. The value of the non-dimensional rate constant k/Y_{eq} was selected to be small compared to the Kolmogorov timescale to ensure a temporally well resolved chemical evolution of the pollutant in the simulation.

The transfer of conserved properties between Lagrangian droplets and Eulerian quantities of the fluid phase was achieved using tri-linear interpolation in physical space with a consistent summation method over cell volumes.

2.1. Parametric variation between cases

Four different simulations were conducted during the course of the Summer Program to examine the effect of varying droplet size and overall fuel-air ratio on the mixing and reaction behavior. The main features of these simulations are listed in Table 1. The primary differences between the cases result from arbitrary variation in the initial number of droplets n_d and the ratio of initial droplet diameter to the Kolmogorov length scale r_D . Forced turbulent statistics were invariant for all cases.

The packing and interaction between droplets within the computational domain is of increasing concern as the liquid volume fraction f_v increases. The simulations considered here did not allow for droplet-droplet interaction. This limitation restricted the maximum allowable liquid volume fraction to the small values listed in Table 1.

The overall fuel mass fraction in the domain was significant when considering the overall chemical stoichiometry of the computational system. This mass fraction was defined as the fraction of mass in the entire domain (including outside the cluster) which originated in the fuel droplets and is equivalent to the mean combined-phase mixture fraction $\langle z \rangle$ (see Sec. 1.1). It can be seen from Table 1 that two of the simulated cases had an overall fuel-air ratio corresponding to stoichiometric, while the other two cases correspond to half the stoichiometric value.

The ratio of the single droplet evaporative time scale to the Kolmogorov timescale r_τ is indicative of the amount of spatial movement that can occur during the lifetime of a fuel droplet. Large values of r_τ indicate that the droplet can evaporate over a protracted trajectory of motion through the surrounding fluid. The evaporative timescale ratio r_τ was determined using the evaporative rate equation (Eq. 2.1) and the stoichiometric reference condition, from which the well-known D^2 -law of single droplet evaporation can be recovered. Under that law, the square of the droplet diameter decreases linearly with

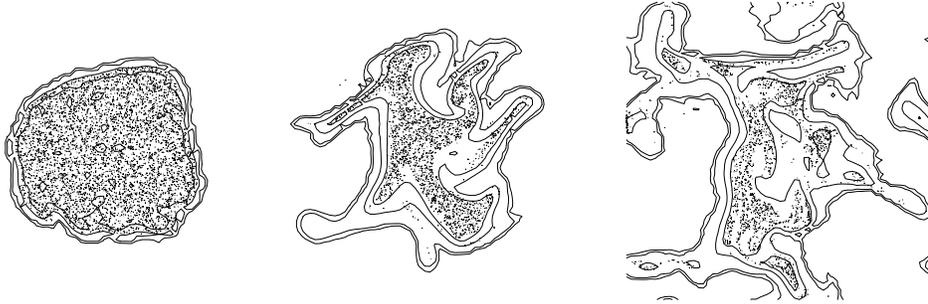


FIGURE 1. Three instances from a typical temporal evolution of a combustive spray cluster on a single slice through the domain over the course of a simulation in the periodic domain. Dots denote droplet location while lines denote isothermal contours.

time. Thus the single drop lifetime is directly proportional to the square of the initial droplet diameter when all other quantities are constant.

It is worth noting that the actual lifetime of the simulated droplet clusters exceeded the tabulated single droplet lifetimes to a degree which depended on the overall fuel-air ratio, and they were at least an order of magnitude greater in each case. This was because during group combustion, the efflux of fuel from other drops causes the conditions surrounding any given drop to be cooler than if it were in isolation.

Chiu and coworkers (see Kuo (1986)) have defined a group combustion number G which describes the ratio of droplet evaporation rate to the rate of mass transport from the droplet cluster. High values of G (e.g., $G > 10^2$) are associated with droplet clusters which are of such fuel type, droplet size, and spacing that allow for the evaporation of droplets within a non-combusting envelope that surrounds a core of non-evaporating droplets and is in turn surrounded at a standoff distance by the flame zone. Chiu calls this regime external sheath combustion. At the other extreme of G (e.g., $G < 10^{-2}$), droplets evaporate at a slower rate so that each individual droplet is enveloped in flame in the so-called single droplet combustion regime.

Following the definition of G , it is possible to simplify and nondimensionalize the definition to give the proportionalities $G \propto n_d r_D \propto f_v r_D^{-2}$. This indicates that for a fixed liquid volume fraction, the propensity for group combustion is proportional to the inverse square of the droplet diameter. From the values of G computed for Table 1, it is clear that all of the simulated cases fall well within the external sheath regime of group combustion.

Figure 1 provides a depiction of the temporal evolution of a typical simulation on a single slice through the domain at three instances over the course of two large-eddy turnover times. It is apparent that the initial spherical form of the cluster is quickly sheared into elongated shapes. Note that in the final (right-hand side) instance in the figure, the periodicity of the simulation is apparent as material is re-advected into the domain.

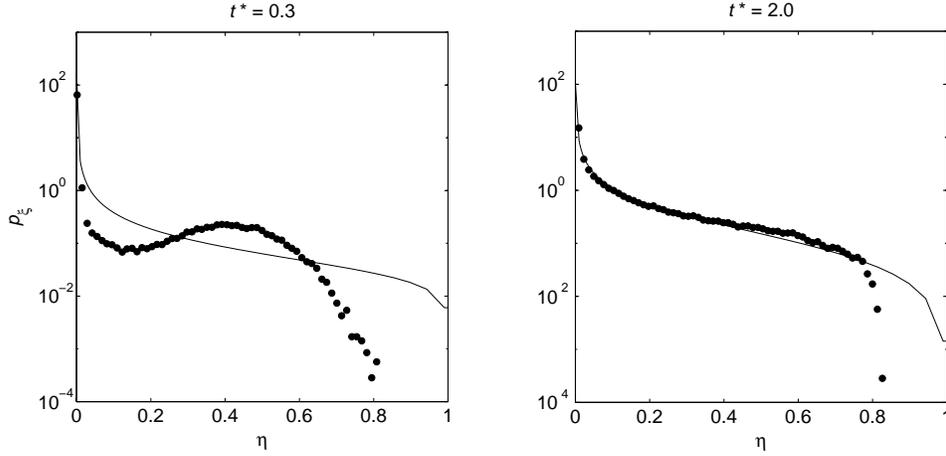


FIGURE 2. Case 6: Solid line = $p_{\xi}(\eta)$, the two-parameter $(\langle \xi \rangle, \langle \xi'^2 \rangle)$ presumed β -function PDF at times (normalized by the large-eddy turnover time) 0.3 and 2.0; \bullet = DNS data.

3. Simulation results

All of the simulation cases were observed to share the common feature of a monotonic increase in the global mean mixture fraction $\langle \xi \rangle$ from zero to a final steady value dictated by the constant value of $\langle z \rangle$ in each case. The only additional significant difference between the cases was the rate at which this steady value was approached, with the cases with smaller initial droplet diameters having the shorter rise times. The droplets were, in any case, essentially completely evaporated within two eddy turnover times (or about ten Kolmogorov timescales) from the initial condition.

This observed $\langle \xi \rangle$ behavior is consistent with the set of earlier unconditional statistical behavior observed by Réveillon and Vervisch (1998). Other unconditional mean statistical data such as mixture fraction variance $\langle \xi'^2 \rangle$, and so on, also followed the findings of this earlier study. The observed behavior of $\langle \xi'^2 \rangle$ can be characterized as having an initial brief rise period which results from interactions between fluctuations in the evaporated field and the continuing evaporative process (see Réveillon and Vervisch 1988). This rise is then followed by a monotonic decay in variance towards zero in a manner which is not unlike the decay of scalar variance in a single-phase mixing system.

The remainder of this report will deal with the important conditional statistics of the simulations which were not reported upon before and yet are critical to subsequent modeling. For the sake of brevity, the following results and analysis is limited to simulation Case 6, but the features discussed are evident in all cases.

Figure 2 depicts the evolution of the PDF of ξ over the course of a simulation and provides a β -function presumed form PDF for comparison. Its controlling parameters $\langle \xi \rangle, \langle \xi'^2 \rangle$ have been taken directly from the DNS data. Typically, β -function presumed form PDFs are used extensively in mixture fraction based models to relate conditional and unconditional statistical properties in various model operations. Good agreement between actual mixture fraction PDF shapes and the presumed form is an implicit requirement for the application of the majority of these models, one which is usually well satisfied in single-phase systems.

Here, however, the β -function representation of the PDF is a poor approximation in the presence of a large mass of evaporating droplets. Note that in the final plot, the PDF

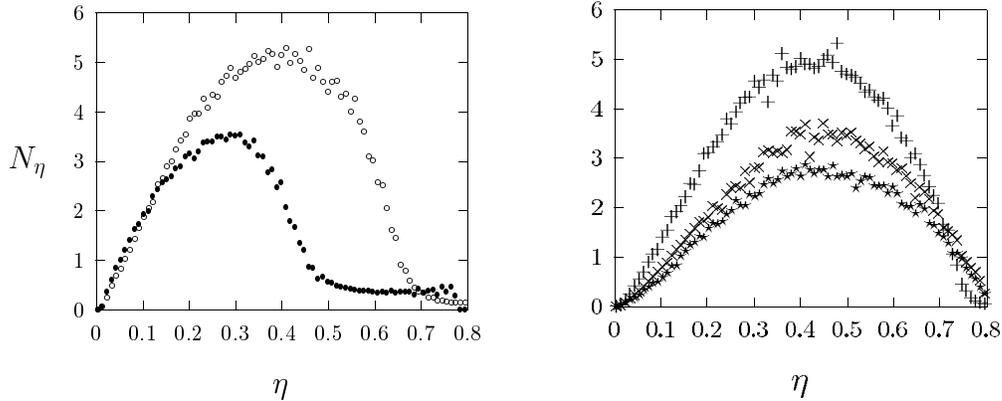


FIGURE 3. Conditional mean scalar dissipation rate (N_η) as a function of mixture fraction derived from DNS data at various times in the evolution of Case 6. Different symbols denote different times (normalized by the large-eddy turnover time): \bullet – 0.1, \circ – 0.2, $+$ – 0.3, \times – 0.7, \star – 1.1

agrees closely with the β -function since by that stage there is a total absence of droplets. The incorporation of additional spray-related parameters into a new presumed-form PDF is perhaps possible but was not attempted during the course of this study.

The related behavior of the conditional mean scalar dissipation rate N_η can be seen in Fig. 3. Unlike the case of mixture fraction in a single-phase system, it is evident from the left-hand plot that there is an initial ‘pumping up’ of the N_η profile before a monotonic decay is manifested in the right-hand plot. Further, the shape of the profile at stages where droplets are present is quite unlike that seen for purely passive scalar dissipation.

In mixture fraction based models for single-phase systems, N_η is required in order to solve Eq. 1.8 and is determined from the evolution of the presumed-form PDF by twice integrating the left-hand side of Eq. 1.6 between bounds (see Klimenko and Bilger 1998). The unusual behavior observed from these simulations renders this methodology doubtful for spray combustion applications. For the moment, however, we will assume that the statistics of ξ can be suitably modeled and focus on feasibility of modeling the combustion when in the presence of droplet evaporation.

4. Model predictions

In this section, we formulate and apply a variant CMC method to model the evolution of the smoke-like pollutant species (see Sec. 2) in the simulated spray combustion.

4.1. Variant model equations

The primary feature of the variant method is a ‘floating’ rich boundary condition, which is treated through an advantageous change in conditioning variable,

$$\zeta \equiv \frac{\eta}{\xi_R}, \quad (4.1)$$

where $\xi_R(t)$ represents the rich boundary which changes in time due to the counteracting effects of droplet evaporation and mixing. Pitsch (1998) has previously applied this type of transformation in relation to flamelet modeling. The change of variables transform

Eq. 1.8 to yield

$$\frac{\partial Q}{\partial t} = \frac{N_\zeta}{\xi_R^2} \frac{\partial^2 Q}{\partial \zeta^2} + \langle \dot{\omega}_r | \zeta \rangle + \left(\frac{\zeta}{\xi_R} \frac{d\xi_R}{dt} + \frac{\langle \dot{s}_\xi | \zeta \rangle}{\xi_R} \right) \frac{\partial Q}{\partial \zeta}, \quad (4.2)$$

where N_ζ corresponds to N_η under the Eq. 4.1 transformation. The initial and lean-boundary conditions for the conditional mean mass fraction of the pollutant are given by

$$Q(\zeta, t = 0) = 0 \quad \text{and} \quad Q(\zeta = 0, t) = 0,$$

respectively. To formulate the rich boundary condition, we draw upon results from an analogous simplified problem.

4.2. Laminar single-droplet analogy

A boundary condition for Eq. 4.2 at $\zeta = 1$ ($\eta = \xi_R$) can be found from consideration of a single fuel droplet evaporating in still air. The governing spherically-symmetric, steady, one-dimensional equations in the radial coordinate (r) for this system can be found in Williams (1985). For all species besides fuel, the boundary condition at the drop surface ($r = r_l$) is,

$$\left(\dot{m}Y - 4\pi r^2 \rho \mathcal{D} \frac{dY}{dr} \right)_{r=r_l} = 0. \quad (4.3a)$$

Defining the scalar dissipation rate $\chi \equiv 2\mathcal{D}(d\xi(r)/dr)^2$ for this laminar case and transforming Eq. 4.3a to the scalar ξ space yields

$$\left(\dot{m}Y(\xi) - \frac{\dot{m}^2}{4\pi\rho\mathcal{D}} \frac{1}{\ln(1-\xi)^2} \sqrt{\frac{\chi}{2\mathcal{D}}} \frac{dY}{d\xi} \right)_{\xi=\xi_R} = 0, \quad (4.3b)$$

where $\xi(r_l) \equiv \xi_R$. Equation 4.3b applies to any choice of ξ so long as its value is known at the droplet interface. Both $\dot{m}(\xi_R)$ and $\chi(\xi_R)$ must also be known in this methodology, but this is not especially difficult given that provisions must already have been made to determine the remainder of these conditional mean profiles.

Equation 4.3b is used to extrapolate the rich boundary condition for Eq. 4.2. Here we simply assume Eq. 4.3a is valid for the turbulent case and conditionally average Eq. 4.3b. A further change of variables given by Eq. 4.1 will yield

$$\left(\langle \dot{s}_\xi | \zeta \rangle Q - \frac{\langle \dot{s}_\xi | \zeta \rangle^2}{4\pi\rho\mathcal{D}} \frac{1}{\xi_R \ln(1-\xi_R)^2} \sqrt{\frac{N_\zeta}{\mathcal{D}}} \frac{\partial Q}{\partial \zeta} \right)_{\zeta=1} = 0. \quad (4.4)$$

This equation is to be enforced at the rich boundary of Eq. 4.2. Equation 4.4 assumes quasi-steadiness since ξ_R is a function of time.

4.3. Results and discussion

The effectiveness of the variant model embodied by Eq. 4.2 can be tested by comparing predictions for conditional mean smoke mass fraction with the evolution observed from the spray combustion simulations.

In this test, *a priori* information is required in the form of the observed N_η and $\langle \dot{s}_\xi | \eta \rangle$ profiles from the DNS. These observed quantities are mapped to ζ space via linear interpolation. The floating rich bound on mixture fraction (ξ_R) is defined to be that value where $\langle \dot{s}_\xi | \eta \rangle$ is a maximum. It is noted that, as with the earlier findings of Réveillon and Vervisch (1998), the profile of $\langle \dot{s}_\xi | \eta \rangle$ was found to exhibit a monotonic increase with mixture fraction at all times and in all cases. The time derivative of the

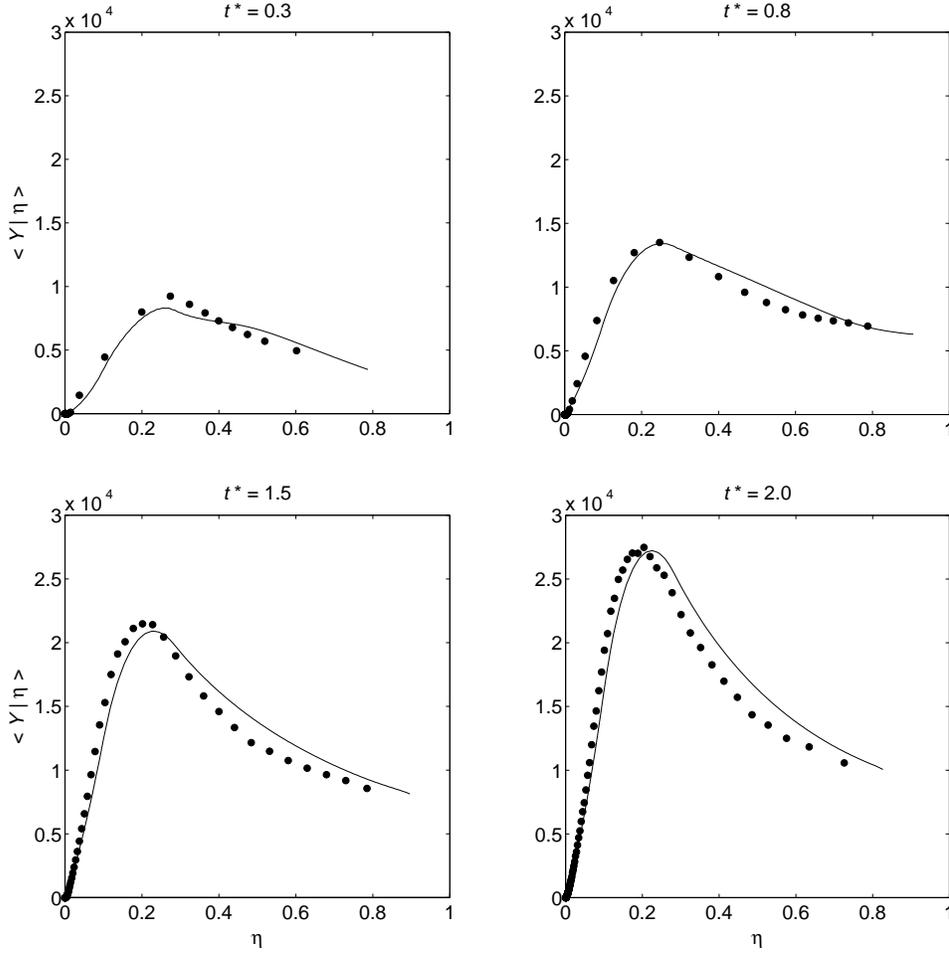


FIGURE 4. Case 6: Solid line = *a priori* modeling solutions of $\langle Y|\eta \rangle$ from Eq. 4.2 at times (normalized by the large-eddy turnover time) 0.3, 0.8, 1.5, and 2.0; \bullet = DNS data.

value of the rich bound ($d\xi_R/dt$) was calculated from a cubic spline fit to the resultant DNS data for $\xi_R(t)$. Cubic splines were also used to interpolate the *a priori* N_η and $\langle \dot{s}_\xi | \eta \rangle$ data in time where temporally-local DNS data was not available in the time record.

Predicted and observed conditional mean results for smoke formation in simulation Case 6 (see Table 1) are shown in Fig. 4. The CMC modeling predictions, which use *a priori* mixing information, are clearly good estimates of the DNS experimental data for all the times of interest. Although not plotted due to space restrictions, the respective model predictions compare equally favorably for all of the other simulation cases. This outcome is encouraging for future model development, which must necessarily focus on finding submodels to obviate the need for the *a priori* information employed in this test.

In order to use Eq. 4.2 in an *a posteriori* role, additional models are required for the interrelated quantities ξ_R , $\langle \dot{s}_\xi | \eta \rangle$, and N_η . In theory, it is possible to solve reactive conditional moment equations like Eqs. 1.8 and 4.2 for droplet mass fractions and number densities and in so doing compute $\langle \dot{s}_\xi | \eta \rangle$ directly. The differential diffusion inherent in

these equations could be treated in the manner of Smith (1998, 1999). The value of ξ_R and its time derivative would also then be available from the expression,

$$\frac{d\xi_R}{dt} = \left[\frac{1}{P_\eta} \frac{\partial}{\partial \eta} (N_\eta P_\eta) + \langle \dot{s}_\xi | \eta \rangle \right]_{\eta=\xi_R} .$$

Further simplification may be possible if it eventuates that the PDF slope term on the right-hand side of the above equation is zero as it is in single-phase mixing cases, so that,

$$\frac{d\xi_R}{dt} = [\langle \dot{s}_\xi | \eta \rangle]_{\eta=\xi_R} . \quad (4.5)$$

Alternatively, a simple empirical model for $\langle \dot{s}_\xi | \eta \rangle$ could be employed as was done by Réveillon and Vervisch (1998) for a different purpose. For the DNS cases considered here, a least-squares curve-fit of $\langle \dot{s}_\xi | \eta \rangle \sim \eta^n$ (for $\eta \in [0, \xi_R]$) reveals that $\langle \dot{s}_\xi | \eta \rangle$ is approximately linear ($n \approx 1.1$ averaged over time). Réveillon and Vervisch (1998) found that n varied more widely according to the regime of spray combustion. This empiricism is perhaps best avoided by using the conditional moment modeling approach for $\langle \dot{s}_\xi | \eta \rangle$ in tandem with the chemically reactive scalars.

The need to provide accurate N_η data for the solution of the CMC equations is a more difficult problem. This problem is made particularly onerous by the absence of an easily parameterized form for the mixture fraction PDF. Réveillon and Vervisch (1998) make use of the β -function presumed form despite its poor agreement with mixture fraction profiles in spray combustion as observed in this study (see Fig. 2). In fact, the inapplicability of simple presumed form PDFs in spray combustion throws doubt on the validity of their use of their one droplet model (ODM) (Réveillon and Vervisch, 1998).

It may be appropriate as a rough approximation to use the β -function presumed form in conjunction with the unconditional mean and variance equations for mixture fraction (as described by Réveillon and Vervisch) for the single purpose of determining an estimate of N_η . This would be done in the usual manner (Klimenko and Bilger 1998) via the double integration of the left-hand side of Eq. 1.6. While this process would likely give reasonable “bulk” magnitudes for the N_η profile, one should be aware that correct profile shapes would not result and even negative N_η profile sections may emerge from the process.

In the absence of an effective presumed form PDF, the only viable alternative is to employ a stochastic methodology for predicting the evolution of the mixture fraction in the PDF and the CMC equations (Klimenko and Smith 2000). Such a method would involve a significant shift away from traditional CMC and unsteady flamelet methodologies but could prove fruitful even beyond the confines of spray combustion.

5. Final remarks

Direct numerical simulations of the group combustion of spray clusters in isotropic turbulence have revealed some important previously unreported observations. Not unexpectedly, it has been found that when in the presence of evaporative sources, mixture fractions defined in terms of gas-phase mass exhibit statistics which are quite distinct from those of single-phase systems.

Perhaps most importantly, the shape and behavior of the mixture fraction PDF is found to deviate significantly from the forms typically seen in pure gas-phase combustion. This observation poses significant complications for the application of gas-phase mixture fraction based combustion models in a liquid fuel environment. This finding calls into

question models proposed at earlier Summer Programs which do not acknowledge these deviations.

Using *a priori* mixing information from the simulations, a new variant of the conditional moment closure and unsteady flamelet family of models was found to provide very accurate predictions of the conditional mean chemical formation of a smoke-like pollutant in the spray combustion systems studied.

Future work will focus on better understanding and parameterizing the gas-phase based mixture fraction PDF forms in spray combustion through further and a more extensive DNS program. This is the current stumbling block to the immediate application of the widely successful mixture fraction based models to spray combustion.

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REFERENCES

- BILGER, R. W. 1993 Conditional Moment Methods for Turbulent Reacting Flow. *Phys. Fluids*, **5**, 436-444.
- KLIMENKO, A. YU. 1990 Multicomponent Diffusion of Various Admixtures in Turbulent Flow. *Fluid Dyn.* **25**, 327-334.
- KLIMENKO, A. YU. & BILGER, R. W. 1998 Conditional Moment Closure for Turbulent Combustion, submitted to *Progress in Energy and Combustion Sci.*
- KLIMENKO, A. YU. & SMITH, N. S. A. 2000 Private communication.
- KUO, K. K. 1986 *Principles of Combustion*. John Wiley and Sons Inc., New York.
- PETERS, N. 1984 Laminar diffusion flamelet models in nonpremixed turbulent combustion. *Progress in Energy & Combustion Sci.* **10**, 319-339.
- PETERS, N. 1986 Laminar Flamelet Concepts in Turbulent Combustion. *Twenty-First Symposium (International) on Combustion*, the Combustion Institute, Pittsburgh, 1231-1250.
- PITSCH, H. 1998 *Modellierung der Zündung und Schadstoffbildung bei der dieselmotorischen Verbrennung mit Hilfe interaktiven Flamelet-Modells*. PhD thesis, RWTH Aachen.
- RÉVEILLON, J. & VERVISCH, L. 1998 Accounting for spray vaporization in turbulent combustion modeling. *Proceedings of the Summer Program*, Center for Turbulence Research, NASA Ames/Stanford Univ. 25-38.
- SMITH, N. S. A., RUETSCH, G. R., OEFELEIN, J. C., & FERZIGER, J. 1998 Simulation and modeling of reacting particles in turbulent nonpremixed combustion. *Proceedings of the Summer Program*, Center for Turbulence Research, NASA Ames/Stanford Univ. 39-60.
- SMITH, N. S. A. 1999 On the differential diffusion of passive scalars in homogeneous turbulence: Implications for combustion modeling. *Proceedings of the 1999 Australian Symposium on Combustion*, University of Newcastle, Australia.
- WILLIAMS, F. A. 1985 *Combustion Theory*. Benjamin-Cummings, New York.