IMPROVED POLLUTANT PREDICTIONS IN LARGE-EDDY SIMULATIONS OF TURBULENT NON-PREMIXED COMBUSTION BY CONSIDERING SCALAR DISSIPATION RATE FLUCTUATIONS

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In this study a new formulation of the unsteady flamelet model is derived to account for the locally resolved distribution of the scalar dissipation rate obtained from large-eddy simulations (LES). Starting from the unsteady flamelet equations, a transformation leads to an Eulerian flamelet model, in which the scalar dissipation rate appears as function of time, space, and mixture fraction. In previous work, it has been shown that LES provides most of the fluctuations of the scalar dissipation rate. Therefore, the present model can be solved within an LES using a local fluctuating scalar dissipation rate. The model is applied to the Sandia flame D, which is a partially premixed, piloted jet diffusion flame. Previously, we have investigated this flame with an unsteady flamelet model, in which only conditionally averaged values for the scalar dissipation rate have been used. Compared to this simulation, accounting for scalar dissipation rate fluctuations leads to improved predictions of the flame structure. In particular, a region of heat release in the rich region of the flame, which is caused by the partial premixing of the fuel with air, does not occur if scalar dissipation rate fluctuations are considered, which is in agreement with the experimental data. This also leads to strongly improved predictions of the mass fractions of stable intermediate chemical species, such as CO and H₂.

Introduction

In recent years, large-eddy simulation (LES) of turbulent combustion has become a subject of intensive research and modeling efforts. The focus of most activities has been model development using *a priori* studies [1,2] and LES of simplified flow configurations using simple chemistry for comparison with direct numerical simulations [3–5]. The first application of LES for non-premixed combustion in a turbulent jet flame with detailed comparison with experimental data has recently been conducted by Pitsch and Steiner [6,7]. In this study the Lagrangian flamelet model (LFM), first introduced by Pitsch et al. [8] in Reynolds-averaged Navier-Stokes simulations (RANS), was used. In this model, unsteady flamelets are solved interactively with the LES solver. The unsteadiness is introduced by solving the flamelets with a scalar dissipation rate history, which a Lagrangian-like particle would experience if it were released at the nozzle and were transported downstream by convective transport. The time coordinate in the flamelet equations hence becomes a Lagrangian-like time. The scalar dissipation rate appearing as a parameter in the flamelet equations and also the axial velocity needed for the evaluation of the Lagrangian time are modeled by the conditional averages over cross-sectional planes perpendicular to the jet axis. The model has been applied to the so

called D flame of the Sandia non-premixed flame series, experimentally investigated by Barlow and Frank [9,10]. The results are compared to measured mean and conditionally averaged temperature and species mass fractions, showing in general very good agreement. However, in the rich part of the flame, CO and H₂ are overpredicted. This is in contrast to the general underprediction of CO in the rich part of diffusion flames that has been reported in the literature. Peters [11] shows a comparison of counter-flow diffusion flame calculations with experimental data and suggests the inadequate chemical mechanism for the CO underprediction. Mauss and Peters [12] discuss the experimentally found superequilibrium values of CO in turbulent jet diffusion flames close to extinction. They show that if local extinction events and a random distribution of residence times are considered in unsteady flamelet calculations, results are qualitatively similar to the experimental data.

The overprediction of CO in flame D has been attributed to the fact that the fuel composition is diluted with air and is hence partially premixed. This has been done to prevent formation of aromatic hydrocarbons, which would interfere with the experimental techniques. The partial premixing causes the occurrence of heat release in the rich premixed region in the predictions of the LES, which in turn leads to overpredictions of mass fractions of CO, H₂,

FIG. 1. Scalar dissipation rate distribution in a cross-section at $x/D = 15$. The isocontour of stoichiometric mixture fraction is given as black line.

and other species [7]. Interestingly, similar trends appear in many other modeling studies using, for instance, the conditional moment closure model [13] or transported probability density function (pdf) models [14,15], which have mainly been applied in RANS-type simulations.

A disadvantage of the LFM as it is applied in Pitsch and Steiner [7] is that only an averaged scalar dissipation rate is used. This is also the case for most other models, particularly in the context of RANS, where only the time-averaged scalar dissipation rate is known. It has been shown in Pitsch and Steiner [16] that the filtered scalar dissipation rate including the resolved and the subgrid part in a jet flame is a strongly fluctuating quantity appearing in large-scale organized structures. Fig. 1 shows the instantaneous distribution of this quantity in a cross-section at $x/D = 15$ from the LES of the Sandia flame D, which will be presented below. It is obvious that by following the isocontour of stoichiometric mixture fraction, given as a black line, a high scalar dissipation rate can be found in some regions, while the stoichiometric scalar dissipation rate is very low in other regions. This spatial structure is lost in the cross-sectional conditional averaging procedure applied in Ref. [7]. In recent work, we have investigated the influence of fluctuations of the scalar dissipation rate in non-premixed combustion [17,18], showing that these can have a strong impact and can lead to local flame extinction, even if the conditional mean scalar dissipation rate is well below the extinction limit.

In the present work, the LFM is reformulated to account for local inhomogeneities of the scalar dissipation rate and to demonstrate their influence. In contrast to the previous formulation, the new model is in Eulerian coordinates and is hence called the Eulerian flamelet model. In the following sections, the governing equations are first derived, and the

relationship to the flamelet equations as derived in earlier work by Peters [11,19,20] is discussed. The numerical implementation of the resulting equations is described, and finally, numerical results from the LES of a turbulent jet diffusion flame, the so-called Sandia flame D, are discussed and compared to the results obtained from LFM and experimental data.

Governing Equations

In this section, a new formulation of an unsteady flamelet model for LES of non-premixed combustion is developed. The underlying ideas are similar to the LFM, but a different formulation allows for a more detailed consideration of local effects. We will start with the unsteady flamelet equations as derived by Peters [11,19,20]. The derivation of these equations and the required assumptions are well known and very well described, for instance, in Peters [11]. The following derivation will only be discussed for the species mass fraction equation, since the derivation for other scalar equations, such as the temperature or enthalpy equation, can be performed analogously.

The unsteady flamelet equations can be written as

$$\rho \frac{\partial Y_i}{\partial \tau} - \rho \frac{\chi}{2} \frac{\partial^2 Y_i}{\partial Z^2} - \dot{m}_i = 0 \quad (1)$$

where the scalar dissipation rate χ has been introduced as

$$\chi = 2D (\nabla Z)^2 \quad (2)$$

Here, Y_i denotes the mass fraction of the chemical species i , τ is the time, ρ is the density, Z is the mixture fraction, and $\dot{m}_i$ is the chemical production term of species i . It should be noted that in equation 1, the time coordinate τ is the time defined in a coordinate system attached to the stoichiometric surface. This implies that the time derivative $\partial/\partial\tau$ is to be evaluated at constant mixture fraction Z . Hence, τ is a Lagrangian-like time coordinate. With respect to a point in space, which is independent of the stoichiometric surface, this new coordinate system moves with the velocity of a point moving on the stoichiometric surface. The unsteady flamelet model using equation 1 in a model for turbulent non-premixed combustion has therefore been called the Lagrangian flamelet model [7,21].

We now want to derive an Eulerian form of the flamelet equations. As the Lagrangian-like coordinate system moves with a point on the stoichiometric surface, for the Lagrangian-like time coordinate τ follows

$$\frac{\partial}{\partial \tau} = \frac{\partial}{\partial t} + \frac{\partial \mathbf{x}_Z}{\partial t} \cdot \nabla \quad (3)$$

where $\partial \mathbf{x}_Z / \partial t$ is the velocity of a point on the stoichiometric surface. The velocity of the scalar isocontours has been discussed in the derivation of the

LFM in Ref. [7] and can, according to Gibson [22], be given as

$$\frac{\partial \mathbf{x}_Z}{\partial t} = \mathbf{v} - \frac{\nabla \cdot (\rho D \nabla Z)}{\rho |\nabla Z|^2} \nabla Z \quad (4)$$

where the first part is due to convection and the second part due to diffusion of the mixture fraction. Since in the following we are considering a jet diffusion flame, the convection velocity we are primarily interested in is in the downstream direction. The second term on the right-hand side of equation 4 acts mainly in the radial direction, since the mixture fraction gradients in downstream direction are small compared to the radial gradients.

Introducing equations 3 and 4 into equation 1 and neglecting the diffusive part in equation 4, which would result in a convective term in physical space by diffusion of the mixture fraction, leads to the flamelet equations in an Eulerian system

$$\rho \frac{\partial Y_i}{\partial t} + \rho \mathbf{v} \cdot \nabla Y_i - \rho \frac{\chi}{2} \frac{\partial^2 Y_i}{\partial Z^2} - \dot{m}_i = 0 \quad (5)$$

In this equation, both the velocity vector and the scalar dissipation rate are functions of space, time, and the mixture fraction. Therefore, the local distribution and temporal fluctuations of these quantities can be considered in the solution of equation 5.

The same equations could have been derived using a two-scale asymptotic analysis as recently suggested by Peters [23], where a short scale is defined in terms of the mixture fraction covering the range in close vicinity of the flame surface. A second coordinate is assumed to describe variations on the long scales only and essentially replaces the spatial coordinates. Since the long-scale coordinate is still Eulerian, the convection term remains during the derivation of the equation, and a transformation as given by equation is not needed.

Equation 5 has been derived to be valid locally and instantaneously. In a turbulent flow field, the velocity $\mathbf{v}$ and the scalar dissipation rate χ are fluctuating quantities. They also depend on time and on the mixture fraction. Mean values of the reactive scalars could in principle be obtained if statistical distributions of these parameters are assumed to be known. For the special case of steady flamelet solutions, Peters [11] has assumed that the pdf of χ is log-normal. Depending on the extinction value of χ , he was able to predict the probability of finding burning flamelets. Such an analysis is inapplicable for unsteady flamelets or in the presence of convective terms as in equation 5.

The flamelet equations derived here show close resemblance to the first-order conditional moment closure (CMC) model proposed by Klimenko [24] and Bilger [25]. The main difference between these models is that in CMC, the scalar dissipation rate is represented by its conditionally averaged value. In

flamelet modeling, these fluctuations would be taken into account by averaging over ensembles of flamelets with different but constant scalar dissipation rates and presuming a pdf of the scalar dissipation rate. As an alternative approach for unsteady flamelet models, it has been suggested in Ref. [26] to solve the unsteady flamelet equations with a fluctuating scalar dissipation rate leading to fluctuations in the dependent variables. For CMC, in contrast, the conditional fluctuations appear as additional unclosed correlations in the CMC equations. These correlations then must be modeled. Therefore, differences in both models appear in the way conditional fluctuations are taken into consideration. It will be argued in the following that the unresolved fluctuations of the scalar dissipation rate can be neglected for the present application. Therefore, the flamelet equations and the first-order CMC equations appear in very similar forms.

To apply the flamelet equations as a subgrid combustion model for LES, subgrid closure will be achieved as suggested in Ref. [8]: The scalar dissipation rate and the velocity in the flamelet equations are replaced by the instantaneous and local, conditionally filtered values of the scalar dissipation rate and the velocity. This implies that the influence of the subgrid fluctuations of the scalar dissipation rate and the velocity is small. This excludes the validity of the current model for situations in which local extinction phenomena are important, since local extinction events are assumed to be mainly caused by high scalar dissipation rate events of very low probability. Note, however, that the subgrid part of the scalar dissipation rate is certainly being accounted for in the model for the filtered scalar dissipation rate. Although conditionally filtered values for the scalar dissipation rate are used in equation 5, the resulting model follows the flamelet ideas. The reason for this is that large-eddy simulations have not only the advantage to yield the spatial structure of the scalar dissipation rate distribution as described in the discussion of Fig. 1, but also seem to predict the correct statistical behavior. Pitsch and Steiner [16] have compared the pdf of the filtered scalar dissipation rate at different locations in a jet diffusion flame with a lognormal distribution. Excellent agreement was found with the variance parameter of the log-normal pdf taken to be unity. The jet exit Reynolds number of the considered case is $Re = 22,400$, which approximately corresponds to a Taylor Reynolds number of $Re_\lambda = 150$. Yeung and Pope [27] have determined the variance parameter from direct numerical simulations and provide a Taylor Reynolds number dependent correlation up to $Re_\lambda = 94$, which yields a variance parameter of $\sigma = 0.95$. Extrapolating the correlation to $Re_\lambda = 150$ gives $\sigma = 1.09$. A value of $\sigma = 1$ has also been suggested by Effelsberg and Peters [28]. This implies that although the scalar dissipation rate is largest at the smallest scales, fluctuations of the scalar

dissipation rate are mainly governed by the large scales and therefore that the major part of the fluctuations of the scalar dissipation rate is resolved by LES. Here, the flamelet equations are solved with an instantaneous fluctuating scalar dissipation rate as suggested in Ref. [26].

Numerical Implementation

Unlike the application of steady-state flamelet models, which is very straightforward, the numerical implementation of the present model needs some further discussion. The reason for this is that the flamelet equations derived here are time-dependent and three-dimensional in space and also depend on the mixture fraction. Also, the intended use of complex chemistry requires that equation 5 be solved for a large number of chemical species. It seems therefore that the current approach is prohibitive in LES, since for simulations of non-reactive flows LES is already known to be an expensive technique. However, an important aspect here is that in a spatially discretized form of equation 5, we solve in each computational cell not only for scalar values of Y_i , but for the function $Y_i(Z)$. Under the assumptions made earlier, this function is not likely to change rapidly in space. Strong changes of Y_i , however, are to be expected in the direction of Z . Here, the spatial discretization of equation 5 will be much coarser than the resolution of the remaining equations. Also variations of $Y_i(Z)$ in radial direction will be neglected.

The reasons for this can be described as follows. Starting from the flamelet equations, a large Damköhler number asymptotic analysis can be performed. The zeroth order of this analysis is the chemical equilibrium solution (or the homogeneous reactor in the unsteady case), which does not include the scalar dissipation rate. In this limit the function $Y_i(Z)$ is constant throughout the entire flow field. The only unsteady changes would be due to initial conditions. These have been shown previously only to be important for very slow reactions like those involved in thermal NO formation [8]. The first-order solutions of this analysis are the flamelet equations, where the scalar dissipation rate appears. Since the first order is a small correction to the zeroth order, only large changes in the scalar dissipation rate can cause changes in $Y_i(Z)$. Since large changes of the scalar dissipation rate can be expected only on large scales, as is discussed in the introduction, only these changes have to be spatially resolved.

Keeping this in mind, the reason why the variations in radial direction are completely neglected can also be understood. Considering an instantaneous radial profile of the mixture fraction, it is obvious that the mixture fraction strongly changes in this direction. Therefore, a particular value of the mixture fraction can only be found within a small region in

radial direction. If this region is smaller than the cell size that must be used in radial direction to resolved the large-scale fluctuations of the scalar dissipation rate, then this particular value of the mixture fraction appears in the radial direction only very locally in a range of the order of the size of one computational cell. Therefore, each cell at a particular time is uniquely represented by a certain mixture fraction range within the flamelet solution and a discretization is not necessary. It should be noted that the limit of very coarse spatial resolution would lead to the Lagrangian flamelet model, so that the improvement in the present model cannot be caused by too coarse spatial resolution.

The present simulations have been performed using a computational mesh in spherical coordinates with $192 \times 110 \times 48$ cells in downstream (s), radial (θ), and circumferential (ϕ) directions. The flamelet equations are spatially discretized in the downstream and circumferential directions only, using 48×8 cells.

In the time discretization of equation 5, the unsteady term, the diffusion in Z , and the chemical source term are solved implicitly. The convection term is explicit and treated as a source term in the implicit solution of the remaining terms.

To further reduce the computational effort, a reduced 20-step mechanism has been derived for the present simulation. The reduced scheme is based on the GRI-Mech 2.11 mechanism of Bowman et al. [29] and constructed such that it leads to the same results as the full mechanism for the present case.

Results and Discussion

In this section, numerical results of the presented model will be shown and compared with earlier results obtained by the LFM [7] and experimental data. Numerical simulations have been performed for a piloted methane/air jet diffusion flame (Sandia flame D), experimentally investigated by Barlow and Frank [9,10]. As mentioned earlier, the fuel has been diluted by 75 vol% of air to minimize the formation of polycyclic aromatic hydrocarbons and soot, which would interfere with the applied experimental techniques. The fuel nozzle is enclosed by a broad pilot nozzle and a coflow of air. The Reynolds number based on the fuel stream is $Re = 22,400$.

The time-averaged mixture fraction development along the centerline predicted by the Eulerian flamelet model and the LFM is given in Fig. 2. The overall agreement with the experimental data is very good. The differences between the two models are small. The stronger decrease of the mixture fraction in the far downstream part of the flow field can be attributed the lower heat release predicted by the new model, which leads to enhanced mixing and thereby to a faster decrease of the centerline value.

FIG. 2. Mean and RMS of the mixture fraction along the centerline. Solid line, Eulerian flamelet model; dashed line, LFM; symbols, experimental data.

The differences in the predictions of the heat release will be discussed in more detail below. Also shown in Fig. 2 is the time-averaged root mean square (RMS) along the centerline. Again the agreement with the experimental data is quite good. Differences between the models are negligible. These trends were to be expected, since the combustion model influences the conserved scalar field only

through the density, which was already well predicted in the LFM simulations.

For the main reaction products, which are not shown here, the predictions of the Eulerian flamelet model are in very good agreement with the experiments. For CO_2 , there are barely any differences from the LFM. However, for H_2O , the new model seems to give some improvement on the fuel-rich side in the region closer to the nozzle. The differences will be explained below in the discussion of the conditional mean quantities.

Conditional mean quantities are given in Figs. 3 and 4 at $x/D = 15$ and 30, respectively. Temperature and NO mass fraction are given in the left-hand side of Figs. 3 and 4. As stable intermediates, CO and H_2 mass fractions are shown in the respective right-hand side of Figs. 3 and 4. Again, the results of the Eulerian flamelet model are compared to the LFM and experimental data.

In the region close to the nozzle, at $x/D = 15$, the maximum temperature can be observed to be slightly overpredicted by both models. This has been discussed by Pitsch and Steiner [7] in a comparison with the single-shot data from the experiments. It has been found that the temperature is well predicted provided that the flame is burning. The decreased conditionally averaged temperature found in

FIG. 3. Conditional mean averages of temperature, NO, CO, and H_2 mass fractions at $x/D = 15$. Solid line, Eulerian flamelet model; dashed line, LFM; symbols, experimental data.

FIG. 4. Conditional mean averages of temperature, NO, CO, and H_2 mass fractions at $x/D = 30$. Solid line, Eulerian flamelet model; dashed line, LFM; symbols, experimental data.

the experiments is only caused by relatively few local extinction events, which are neglected in both the Eulerian and the Lagrangian flamelet models. On the rich side, the temperature is slightly overpredicted by the LFM, but well predicted by the Eulerian flamelet model. Also, for the NO profile given in Fig. 3, the new model leads to a significant improvement, showing very good agreement with the experiments, whereas the predictions by the LFM overpredict the maximum NO concentration slightly. Since NO is well known to be strongly dependent on the temperature, it seems at first surprising that NO agrees well even though the temperature is overpredicted. The reason is that the high activation temperature of the NO formation causes NO to be formed essentially at the highest temperatures. As explained earlier, the local temperature seems to be well predicted, and the overprediction of the conditional mean temperature is only caused by a small volume fraction with significantly lower temperature, which in any case does not contribute to the formation of NO.

Similar to the temperature, but much more obvious, CO and H₂ mass fractions at $x/D = 15$ in the rich part of the flame are overestimated by the LFM. The predictions of the Eulerian flamelet model again predict the experimental data very well. The reason for these differences will be explained in the following discussion.

At $x/D = 30$, shown in Fig. 4, the temperature profile predicted by the LFM shows a distinct heat release region at approximately $Z = 0.6$. This heat release is caused by the high air content of the fuel stream. The interaction with the main diffusion flame, located at approximately $Z = 0.35$, causes consumption of fuel and oxidizer, which essentially forms a rich premixed reaction zone. Within this region, because of the high local equivalence ratio, rich stable intermediates, such as CO and H₂, are formed in large concentrations as shown in the same Fig. 4. However, the rich premixed heat release region cannot be observed in the experimental data. Hence, the temperature in this region is slightly overpredicted, while the CO and H₂ mass fractions are strongly overpredicted.

The main difference between both combustion models discussed here is that in the LFM, the scalar dissipation rate is conditionally averaged over cross-sections, whereas the Eulerian flamelet model is able to take the local fluctuations of the scalar dissipation rate and the unsteady response of the mixing process and the chemistry into account. The consideration of these fluctuations in the present simulations using the Eulerian flamelet model obviously changes the turbulence-chemistry interaction in such a way that chemical reactions in the rich premixed part cannot occur, which is consistent with the experimental findings. Therefore, all quantities predicted by the Eulerian flamelet model are in

much better agreement with the experimental data. This is reflected in the temperature profile, which does not reveal the rich heat release region as well as in the conditionally averaged mass fractions of the stable intermediates. Both CO and H₂ mass fractions show excellent agreement with the experiments. For NO, this effect does not have a large influence, since the mass fraction in the rich part is more governed by transport of NO from the formation region at the maximum temperature, which is only weakly influenced by the rich heat release region, even in the predictions by LFM. However, as shown in Fig. 4, at $Z \sim 0.6$, the LFM predictions reveal an NO consumption region caused by the heat release in this region. Even though NO also seems underpredicted by the results of the Eulerian flamelet model, these do not show NO consumption, which is consistent with the experiments. A similar discrepancy as seen in the comparison of NO obtained from the Eulerian flamelet model, and the experimental data have been found in a comparison of predictions of a laminar counterflow diffusion flame with experiments using the same fuel [30], indicating that this underprediction might be caused by the chemical reaction scheme rather than the combustion model.

The observed trends continue for the farther downstream positions. Here, the heat release in the rich region is even more pronounced in the temperature predictions by the LFM simulation. Interestingly, this can also be observed in a much weaker form in the predictions of the Eulerian flamelet model and in the experimental data. This can be very clearly seen in the right-hand side of Fig. 5, where the conditional averages of methane and molecular oxygen are shown for $x/D = 45$. Obviously the oxygen is depleted not only in the reaction zone of the diffusion flame at $Z = 0.35$, but over a much wider region, ranging from stoichiometric conditions to approximately $Z \sim 0.6$. Also the consumption of fuel starts at higher values of the mixture fraction compared to $x/D = 30$, which is shown in the left-hand side of Fig. 5.

The influence of the rich heat release on CO and H₂ at $x/D = 45$ is still quite strong. The results of the Eulerian flamelet model again provide significantly improved predictions compared to the LFM.

Conclusions

In the present paper, an Eulerian formulation of the unsteady flamelet model for LES of non-premixed combustion is presented, which in contrast to the Lagrangian flamelet model, as formulated in an earlier study, accounts for local fluctuations of the scalar dissipation rate.

The model has been applied to the Sandia flame D, and the predictions are compared to results obtained by the LFM and to experimental data. It is

FIG. 5. Conditional mean averages of CH_4 and O_2 mass fractions at $x/D = 30$ and $x/D = 45$. Solid line, Eulerian flamelet model; dashed line, LFM; symbols, experimental data.

demonstrated that inaccuracies in the LFM predictions in the rich part of the flame are mainly caused by an incorrect prediction of the heat release in this region. It is shown that the results using the newly formulated model provide a significant improvement over the LFM results and are in excellent agreement with the experimental data.

The main conclusions of this study are that the most important reason for the improvement over the earlier model formulation is the consideration of the locally resolved fluctuations of the modeled scalar dissipation rate and the unsteady response of the interaction of molecular mixing and chemistry.

It seems noteworthy that these fluctuations of the scalar dissipation rate can only be observed in LES-type calculations, which implies that the level of accuracy obtained in the present study cannot be achieved using Reynolds-averaged methods.

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COMMENTS

Stephen B. Pope, Cornell University, USA. You draw the strong conclusion that pollutants in flame D can only be calculated by LES. Is this conclusion consistent with the accurate PDF calculations of Lindstedt et al. (Ref. [14] in paper), Xu and Pope (Ref. [15] in paper), and Tang et al. [1]?

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1. Tang, Q., Xu, J., and Pope, S. B., *Proc. Combust. Inst.* 28:133 (2000).

Author's Reply. The results from transported PDF calculations reported in Xu and Pope (Ref. [15] in paper), Tang et al. (Ref. [1] in comment), and Lindstedt et al. (Ref.

[14] in paper) all show overpredictions in both H_2 and CO on the rich side by values up to 30%, which is roughly what we found in our earlier simulations using the Lagrangian Flamelet model. As shown in the paper, the present simulations lead to a strong improvement for H_2 and CO by accounting for local scalar dissipation rate fluctuations. However, recent unpublished results by Lindstedt (private communication) also show very accurate predictions for these species on the rich side. It will be interesting to explore the differences in the modeling compared with their earlier simulations reported (Ref. [14] in paper). Some portion of the scalar dissipation rate fluctuations might be inherently included in some of the models used to describe mixing in transported PDF methods. If this is the case, I agree with you that these methods should also be able to predict CO for the present case correctly.