Direct numerical simulation of turbulent combustion: fundamental insights towards predictive models

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Abstract. The advancement of our basic understanding of turbulent combustion processes and the development of physics-based predictive tools for design and optimization of the next generation of combustion devices are strategic areas of research for the development of a secure, environmentally sound energy infrastructure. In direct numerical simulation (DNS) approaches, all scales of the reacting flow problem are resolved. However, because of the magnitude of this task, DNS of practical high Reynolds number turbulent hydrocarbon flames is out of reach of even terascale computing. For the foreseeable future, the approach to this complex multi-scale problem is to employ distinct but synergistic approaches to tackle smaller sub-ranges of the complete problem, which then require models for the small scale interactions. With full access to the spatially and temporally resolved fields, DNS can play a major role in the development of these models and in the development of fundamental understanding of the micro-physics of turbulence-chemistry interactions. Two examples, from simulations performed at terascale Office of Science computing facilities, are presented to illustrate the role of DNS in delivering new insights to advance the predictive capability of models. Results are presented from new three-dimensional DNS with detailed chemistry of turbulent non-premixed jet flames, revealing the differences between mixing of passive and reacting scalars, and determining an optimal lower dimensional representation of the full thermochemical state space.

1. Introduction
There are powerful motivators for the study of turbulent combustion. Combustion accounts for 85% of the energy production in the United States, and any change from this situation would occur on a multi-decade timescale due to large infrastructure investments. High oil prices and dependence on foreign oil, especially in the future as reserves dwindle, will create a great need for highly efficient combustion devices. There will also be increasing environmental impact of power generation activities due to growth in energy demand unless new, cleaner combustion technologies are developed to maturity. A deep understanding of the basic phenomena of reacting flows is essential to improve the performance, reliability, safety, and environmental impact of such technologies. The greatest impact of such an understanding can be achieved if it is translated into predictive models that can be used by designers through the use of Computational Fluid Dynamics (CFD).

The industrial use of CFD as a design tool is becoming increasingly widespread. Where a non-reacting flow is involved, methods are now sufficiently developed to produce routinely results that are physically realistic, even for complex flows. However, the situation is less satisfactory
where reactions and turbulence present. Local reaction rates and interactions between the thermo-chemistry and the turbulent flow field require representation via additional sub-grid scale modeling. In contrast to CFD approaches used in industry, where the turbulent fluctuations are at best partially resolved on the computational grid, direct numerical simulation (DNS) resolves all relevant continuum flow and flame scales and sub-grid scale models are not required. DNS is computationally a very demanding technique, and will remain intractable for industrial problems for the foreseeable future because of the need to resolve a large range of scales. However, in recent years, the rapid advance of computational capabilities has presented significant opportunities for increasingly realistic direct simulations of turbulent combustion flows, which are very well suited to assess the validity of underlying assumptions of modeling approaches. With full access to the spatially and temporally resolved fields, it is possible to extract, under well controlled conditions, detailed information about small scale flame behavior that cannot be measured by experiments. This information can then be used to develop modeling strategies for CFD approaches that do not resolve the entire range of scales.

In many practical combustors the fuel and air are not premixed. For example, this is the case in aircraft applications where fuel and oxidizer are segregated for safety reasons, and in direct injection internal combustion engines, for reasons related to efficiency gains. Several groups \[1\] have demonstrated reasonable success using CFD in modeling nonpremixed flames without strong finite-rate chemistry effects. However, there are still limitations and uncertainties in the ability of these models to describe important finite-rate combustion phenomena. Current modeling approaches would benefit greatly from more detailed characterization of these effects in turbulent nonpremixed combustion. DNS can play a major role in providing this information.

In this paper, new results from three-dimensional (3D) DNS of turbulent nonpremixed jet flames with detailed chemistry are presented. These extremely demanding computations have been enabled by Office of Science super-computing facilities. Calculations have been performed at the National Energy Research Scientific Computing Center (NERSC) using a large award of compute time under the INCITE program, at the Molecular Science Computing Facility (MCSF), and at the National Leadership Computing Facility (NLCF).

The remainder of the paper is organized as follows. In Section 2, the numerical methods used are described, and the parameters and set-up of the simulations are briefly presented in Section 3. Knowledge extraction from these large, complex, data-sets is a major challenge and requires specialized visualization techniques. Section 4 shows some flame visualizations generated by collaborators Kwan-Liu Ma and Hiroshi Akiba at U.C. Davis. The overall goals of the work are to provide fundamental insights on strong flame-turbulence interactions in nonpremixed flames and their impact on predictive models. Two examples of how this may be achieved are outlined in Sections 5 and 6.

2. Numerical Methods
The simulations were performed using Sandia’s massively parallel DNS code, S3D, which has been developed with support from the Basic Energy Sciences program over the last 13 years. This code solves the full compressible reacting Navier-Stokes, total energy, species and mass continuity equations coupled with detailed chemistry. It is based on a high-order accurate, non-dissipative numerical scheme. It has been used extensively to investigate fundamental turbulence-chemistry interactions in combustion topics ranging from premixed flames \[2, 3\], autoignition \[4, 5\], to nonpremixed flames \[6, 7\]. Time advancement is achieved through a six-stage, fourth-order explicit Runge-Kutta (R-K) method \[8\], spatial differencing is achieved through eighth-order finite differences with tenth-order explicit finite-difference filters on a structured, Cartesian grid \[9\], and Navier-Stokes Characteristic Boundary Conditions (NSCBC) \[10, 11\] were used to prescribe the boundary conditions. The equations are solved on a conventional structured mesh, and scaleable parallelism is achieved through MPI and a domain-decomposition strategy.
Figure 1 shows the parallel speed-up of the code on several Office of Science platforms, including the IBM SP at NERSC known as Seaborg, the HP-Itanium-2 cluster at MSCF known as MPP2, the Cray-X1 at NLCF known as Phoenix, and a local Combustion Research Facility Opteron-Infiniband cluster known as Jet. In these tests the problem size per processor is fixed, and the total problem size is increased in proportion with the number of processors. It may be observed that the code is scaling very well on all platforms. On 4096 SP3 processors at NERSC, 70% parallel efficiency is observed. There is no reason to expect that this trend would not continue to tens of thousands processors and beyond.

![Figure 1. Parallel speed-up versus number of processors for several Office of Science platforms.](image)

3. Simulations
DNS of 3D turbulent nonpremixed CO/H\textsubscript{2}/N\textsubscript{2}-air jet flames with detailed chemistry have been performed. The physical configuration chosen corresponds to a temporally-evolving plane jet flame. In the temporal configuration, an inner turbulent fuel core flows within quiescent air, and these streams are separated by reacting mixing layers under the influence of significant mean shear. The configuration results in similar but not identical turbulent structures to those observed in a spatially evolving planar jet, with an observation window that moves with the mean jet velocity. This configuration was selected rather than the spatially-evolving jet because it allows for more significant flame-turbulence interaction within a given computational domain with wider separation in mixing scales than previously possible [12, 13], thereby potentially creating a more wrinkled flame surface through intense turbulent mixing.

The detailed initialization procedure is described in Sutherland [14]. The mean streamwise velocity fields are initialized using a smoothly varying profile with a specified mean convective velocity difference \( U \), jet height \( H \) and shear layer thickness \( \delta_u \). The velocity varies from \(-U/2\) at the boundaries in the transverse direction to \(U/2\) in the center of the jet. Superimposed upon this mean field is a small forcing of a solenoidal isotropic velocity field. The forcing has a turbulence intensity \( u' \) and an integral scale \( L_t \). The forcing is spatially confined to the jet region and is either centered on the core of the jet or in the shear layer, depending on the case.

The scalar fields are initialized by specifying an initial profile of mixture fraction in the domain, similar to the profile of velocity. The initial thickness of the mixture fraction variation is
given by $\delta_Z$. A flamelet table is used to specify the initial species mass fractions and temperature [14].

The flow evolves from the initial condition. Boundary conditions are periodic in the streamwise and spanwise directions and non-reflecting outflow conditions are employed in the transverse direction [10, 11]. The small initial turbulent forcing triggers instabilities in the jet shear layer, which eventually develop into a fully turbulent, shear-driven flow. The turbulence, mixing and reaction interact strongly during the middle of the simulations. Later, the jet breaks down, shear rates decrease, mixing rates relax, and the local thermochemical state begins to approach equilibrium.

Two runs have been performed to date. Table 1 summarizes the run parameters. In the table, $Re = UH/\nu$ is the Reynolds number based on the jet velocity and height, where $\nu$ is the oxidizer viscosity. The dimensions of the domain in the streamwise, transverse and spanwise directions are denoted $L_x$, $L_y$, and $L_z$ respectively. The Mach number, $M$, is defined as $U/2C$ [12], where $C$ is the speed of sound. The Mach number is sufficiently small such that compressible effects can be neglected, however not so small such that the computational cost becomes excessive due to the acoustic CFL criterion. A Damköhler number is defined as $Da = \chi_qH/U$, where $\chi_q$ is the steady extinction limit of the mixture as calculated by the laminar flamelet equations [15]. The fuel and oxidizer compositions are given in Table 2. The unreacted fuel and oxidizer temperatures are 700K in Case A and 600K in Case B.

In Case A, the kinetic mechanism employed for CO/H$_2$ oxidation includes 12 species and 33 reactions [16]. In Case B, an improved starting mechanism was adopted [17] with the same numbers of species and reactions. Tests in steady premixed and non-premixed flames and unsteady two-dimensional DNS showed that H$_2$O was unimportant for the conditions considered and therefore it was removed from the mechanism. Also, several unimportant reactions involving HCO were removed. The resulting skeletal mechanism has 11 species and 21 reactions.

Table 1. Run parameters used in DNS cases.

| Case | A  | B  |
|------|----|----|
| Re   | 5850 | 7780 |
| Grid number (millions) | 40 | 100 |
| $L_x/H$ | 4.1 | 4.5 |
| $L_y/H$ | 6.2 | 6.7 |
| $L_z/H$ | 1.0 | 2.7 |
| $\delta_u/H$ | 0.34 | 0.12 |
| $\delta_Z/H$ | 0.34 | 0.5 |
| $L_{t,0}/H$ | 0.6 | 0.5 |
| $u'/U$ | 0.15 | 0.05 |
| $M$ | 0.1 | 0.25 |
| $Da$ | 0.04 | 0.014 |

Table 2. Streams composition used in DNS cases (% by volume).

| Case | A  | B  |
|------|----|----|
| Fuel |    |    |
| CO   | 28 | 45 |
| H$_2$ | 7 | 10 |
| N$_2$ | 65 | 45 |
| Oxidizer |    |    |
| O$_2$ | 21 | 21 |
| N$_2$ | 79 | 79 |

Case A was run on 480 processors of the MPP2 HP-Itanium-2 cluster at PNNL. Approximately 120,000 hours were required. A total of roughly 1TB of data was produced by this run.

Case B was started using 1728 Power 3 processors of the Seaborg IBM SP RS/6000 at NERSC. Approximately 300,000 hours were required to advance the simulation to one-third of completion. After the run was one third complete it was transferred to the Phoenix Cray X1 and
X1E, where it has used 192 or 240 processors, depending on queue availability. The completion of the run required an additional 140,000 hours. A total of 2.5 TB of data resulted from this run.

These cases will be complemented with new larger runs to be performed later this year using time from the INCITE grant at NERSC. It is expected that these calculations will require 2.5 million hours and generate 5TB of data.

4. Flame visualization
A single DNS simulation produces several terabytes of raw data that is vast in the spatial (hundreds of millions of grid points), temporal (hundreds of thousands of time steps) and variable (tens of variables) domains, creating a formidable challenge for subsequent analysis, visualization and interpretation. In addition to the size of the data-set, the difficulty of knowledge extraction is compounded by the sheer complexity of the turbulent flow-fields and of the phenomena being studied. To understand the dynamic mechanisms of turbulence-chemistry interactions in turbulent flames, simultaneous visualization of multiple scalars in a reacting flow is required. Kwan-Liu Ma of UC Davis has used interactive multi-variate exploration techniques taking into account particularly the cognitive aspects of visualization [18].

Figure 2 shows a time sequence of a three variable volume visualization from Case A. The mass fraction of the OH radical, which can be taken to represent the existence of a flame is shown in red. The stoichiometric mixture fraction isosurface, which represents the most likely flame location, is shown in blue, and the scalar dissipation rate, which represents mixing, is shown in yellow. It is observed that due to the effects of turbulence, the OH field is far from uniform on the mixture fraction isosurface, which would have been expected from fast chemistry arguments. High OH regions are found in regions where the mixture fraction isosurface is highly convoluted, and low values in the regions where the mixture fraction isosurface is stretched and not wrinkled. The scalar dissipation ($\chi$), which is closely related to strain rate, is essentially the rate of mixing between fuel and oxidizer, and is a very important quantity in nearly all models of nonpremixed combustion. Some mixing is desirable for efficient combustion, but very large mixing rates lead to heat and radical losses that are so great such that chemical reaction is no longer self sustaining. Figure 2 shows that the high $\chi$ regions occur in low-volume, pancake-like structures and are aligned according to principal strain directions of the shear flow. High $\chi$ and low OH regions coincide.

Figure 2. Simultaneous visualization of stoichiometric mixture fraction, scalar dissipation, and OH radical mass fraction variables from Case A at selected time steps.

Single variable volume visualizations from the larger Case B are shown in Figure 3. The scalar dissipation of mixture fraction is shown in Fig. 3(a), the vorticity magnitude in Fig. 3(b), and the molecular dissipation rate of OH and HO$_2$ in Figs. 3(c) and (d) respectively. Compared with Case A, the expanded computational domain and larger Reynolds number results in a more complex structure and more intense fine-scale turbulence. For a discussion of the significance of the dissipation rate variables, see the following section.
5. Mixing timescales
A substantial benefit of DNS calculations which incorporate detailed chemical kinetics, thermodynamics, and transport is that any aspect of the calculation may be interrogated in a high level of detail. This facilitates a priori analysis of the data to provide predictions of model performance and assess modeling assumptions. In this section the first example of how data from these simulations can help to advance predictive models is presented and concerns the modeling of molecular mixing timescales.

5.1. The need for mixing timescale models
Models for molecular mixing are required in many CFD approaches for the simulation of turbulent combustion. In particular, molecular mixing is the central modeling question in the PDF approach [19]. Within the context of this framework, many different strategies have been adopted, including Interaction by Exchange with the Mean (IEM) [20], Linear Mean-Square Estimation (LMSE) [21], Modified Curl (MC) [22], and Euclidean Minimum Spanning Trees (EMST) [23]. A common element in all of these approaches is a mixing timescale. Pope [24] has recently pointed out that model predictions are dependent on the choice of the timescale, and different choices are appropriate for different problems. Normally, the timescale

Figure 3. Clockwise from top left: (a) Scalar Dissipation (b) Vorticity (c) OH Dissipation Rate (d) HO\textsubscript{2} dissipation rate from Case B
is assumed to be the same for each different scalar, and the same order of magnitude as the large scale turbulence timescale. In flames, differential diffusion and the strong interplay with mixing and reaction might degrade these assumptions. It is difficult to directly assess these assumptions in *a-posteriori* tests, and measurements of reacting scalar mixing are not yet possible. DNS of reacting flows with detailed chemistry provides unique opportunities to evaluate such assumptions.

5.2. Definitions

Here, a mixing timescale is defined for a scalar $\phi$:

$$\tau_\phi = \frac{\phi''^2}{\chi_\phi},$$

(1)

where a dissipation rate $\chi_\phi$ is defined for the scalar as:

$$\chi_\phi = 2D_\phi \nabla \phi \cdot \nabla \phi,$$

(2)

and $D_\phi$ is the mixture-averaged diffusion coefficient for the scalar. A mechanical timescale, which represents the characteristic large-scale turbulence timescale is defined by:

$$\tau_u = \frac{k}{\epsilon},$$

(3)

where $k$ is the turbulence kinetic energy and $\epsilon$ is its dissipation rate. The average $\langle \ldots \rangle$ in these simulations is taken over the spanwise and streamwise directions, which are statistically homogeneous, and the symmetry in the cross-stream direction is also exploited. The ratio of the scalar to mechanical mixing timescale is defined as:

$$r_\phi = \frac{\int_0^{\delta_f} \tau_a dy}{\int_0^{\delta_f} \tau_\phi dy},$$

(4)

where $y$ is the distance in the transverse direction from the jet center-line, $\delta_f$ is the $y$ location at which $f < 0.05$, and $f$ denotes mixture fraction. Integration across the $y$ direction allows convenient presentation of the results as a single timescale ratio, and reduces statistical scatter.

5.3. Passive scalar mixing

Let $r_f$ denote the ratio of the mechanical timescale to that of the mixture fraction. Figure 4 shows the time variation of $r_f$, where time has been normalized a reference transient jet time $(U/H)$. The mixture fraction represents the local mass fraction of constituents from the fuel stream, and is a passive scalar in that its transport equation contains no reactive source term. A Lewis number of unity is assumed to calculate the diffusivity. The figure shows that the timescale ratio is order unity throughout the simulation, varying between values of 2.1 and 1.0. The present result is similar to values reported by experiments, simple chemistry DNS and used successfully in modelling [25]. The result confirms the assumption that $r_f$ should be order unity (due to the balance between production and dissipation) is valid for passive scalars with Schmidt numbers order unity, even in a reacting flow.
5.4. Effect of Schmidt number
Scalars with non-unity Schmidt numbers can potentially have different mixing timescales. Figure 5 shows the mixing timescale ratio for the H, H\textsubscript{2} and CO\textsubscript{2} mass fractions versus time, along with the passive scalar timescale. These molecules have different diffusivities - H is the most diffusive while CO\textsubscript{2} is the least. The figure clearly shows there is an effect of diffusivity on the mixing timescales. The most diffusive scalar H has a maximum timescale ratio of 6.2 while CO\textsubscript{2} has a maximum timescale ratio of 1.6. The difference between these values however is less than the difference in the diffusivities. The results indicate that differential diffusion effects may need to be incorporated within mixing models, at least at moderate Reynolds numbers. This conclusion could possibly be Reynolds number dependent, and a parametric study in Reynolds number will ultimately be required to determine any such dependence.

5.5. Effect of reaction-mixing coupling
The strong interplay between reaction and molecular mixing in nonpremixed flames can also affect mixing timescales. Figure 6 shows the mixing timescale ratios for HO\textsubscript{2}, H\textsubscript{2}O\textsubscript{2}, and O and OH. Initially, these timescales are ordered according to the diffusivity, but during the middle of the simulation, the timescales of O and OH increase and those of HO\textsubscript{2} and H\textsubscript{2}O\textsubscript{2} decrease, and the trend of diffusivity no longer holds. This is a result of an interesting interplay between reaction and molecular mixing. Figure 7(a) shows the HO\textsubscript{2} mass fraction, and Figure 7(b) shows the OH mass fraction on a color scale for the time 10 \textit{H/U}. In both figures, white contours of the scalar dissipation \( \chi \), a measure of the passive scalar molecular mixing rate, are overlaid. It may be observed that OH levels are lower in regions of high \( \chi \), while HO\textsubscript{2} levels are higher. In these high \( \chi \) regions, conditions approach extinction and OH and O radicals are chemically destroyed while the stable intermediates HO\textsubscript{2} and H\textsubscript{2}O\textsubscript{2} are produced. This has a direct effect on the dissipation fields. Figure 8(a) shows the fields \( \chi_{HO2} \), \( \chi_{OH} \) and \( \chi \) for Case A. As a result of the strong production of HO\textsubscript{2} in high \( \chi \) regions the \( \chi_{HO2} \) and \( \chi \) fields are coincident, while in these regions high \( \chi_{OH} \) dissipation does not occur. These high \( \chi \) regions are most prevalent in the middle of the simulation, and together with the coupling of reaction and molecular mixing lead to longer mixing timescales in the case of OH and shorter in the case of HO\textsubscript{2}. Later, dissipation rates relax and O and OH return, leading to an increase of their timescales. These findings underline the importance of considering the interplay of diffusion and reaction, particularly when strong finite chemistry effects are involved. Preliminary results of the same fields from Case B are shown in Figure 8(b). Although this case has not yet been analyzed in detail, a qualitatively similar picture is emerging.

5.6. Implications
In summary, the data show for the first time how detailed transport and chemistry effects can influence the mixing of reactive scalars. It may be advantageous to incorporate these effects within molecular mixing models. It is worth noting that at present it is impossible to obtain this type of information any other way than by using the type of highly resolved simulation performed here. Experimental measurements of the scalar dissipation in flames are very difficult and even point-wise measurements have only recently become possible [26]. Full access to the 3D spatial and temporally resolved dissipation fields without the interference of noise, and of multiple reacting scalars occurring in even thinner layers than those corresponding to mixture fraction is at present unthinkable experimentally.

6. Assessment of Thermochemical Modeling Approaches
The second example of how these simulations can be used to develop models concerns the possible representation of the full thermochemical state space in a lower dimensional space.
Figure 4. Mechanical to scalar timescale ratio for mixture fraction versus time.

Figure 5. Mechanical to scalar timescale ratio for H, H\textsubscript{2} and CO\textsubscript{2} mass fractions and mixture fraction versus time.

Figure 6. Mechanical to scalar timescale ratio for H, H\textsubscript{2} and CO\textsubscript{2} mass fractions and mixture fraction versus time.
In general, the thermochemical state of a single phase system may be uniquely specified by the temperature ($T$), pressure ($P$), and $N_s - 1$ mass fractions ($Y_i$). In other words, we require $N_s + 1$ independent parameters to uniquely specify any thermochemical property of the system. The task from a modeling perspective is then to choose a much smaller set of parameters which represent the thermochemical state of the system to within a desired degree of accuracy. Obviously, greater accuracy requires more variables to be transported on the computational mesh. We refer to a model which represents the thermochemical state by fewer than $N_s + 1$ parameters as a reaction model. Analysis of the DNS data allows a priori assessment of the potential accuracy of classes of models.

6.1. Analysis Procedure

Let us denote the set of variables $\{T, p, Y_i\}$ as Natural reaction variables. In general, we wish to obtain a set of state variables, $\phi$, as functions of the natural reaction variables. State variables
are any thermochemical variable which can be derived from the full set of natural reaction variables. A model attempts to parameterize the state variables by a set of reaction variables that is much smaller than the full set of natural reaction variables.

Consider a set of reaction variables \( \eta \), by which we wish to parameterize the thermochemical state, \( \phi \), of the system. We may project a DNS data set into \( \eta \)-space and define a mean surface that the DNS data occupies by \( \langle \phi | \eta \rangle \), the average value of the state variables conditioned on a given value of the reaction variables. We may then determine the higher-order moments to quantify the degree to which the DNS departs from the mean surface in \( \eta \)-space. Specifically, the standard deviation of \( \phi \) from its mean in \( \eta \)-space may be expressed as

\[
\sigma_{\phi_i} = \sqrt{\left\langle \left( \phi_i - \langle \phi_i | \eta \rangle \right)^2 \right\rangle | \eta \rangle},
\]

where \( \phi_i | \eta \) represents all values of the \( i^{th} \) state variable which correspond to the given values \( \eta \), and \( \langle \rangle \) indicates an average. Note that while this measure is local in \( \eta \)-space, it is non-local in physical space. It is also non-unique in physical space, i.e., there may be many points in physical space which have the same \( \eta \). Thus, \( \sigma_{\phi_i} \) is a statistical measure of the ability of \( \eta \) to parameterize \( \phi \). If \( \eta \) uniquely parameterizes \( \phi \), then \( \sigma_{\phi_i} \) defined by Equation (5) is identically zero. This is the case if \( \eta = (Y_1, Y_2, \ldots, Y_{N_s-1}, T, P) \). If, however, a set of \( \eta \) were chosen such that the mapping were not unique, then Equation (5) would be nonzero for at least some values of \( \eta \). Thus, for an arbitrary choice of \( \eta \), Equation (5) provides a measure of how well \( \phi \) may be parameterized by \( \eta \).

We will also consider the normalized standard deviation, given as

\[
\epsilon_{\phi_i} \equiv \frac{\sigma_{\phi_i}}{\langle \phi_i | \eta \rangle}.
\]

This may be interpreted as the average fractional error that is incurred by representing the DNS data by its conditional mean in \( \eta \)-space.

As described in [14], the measure \( \sigma_{\phi_i} \) obtained from Equation (5) provides a quantitative measure of the best possible performance a given model parameterized by \( \eta \) can achieve relative to the DNS data. Of course, this does not guarantee that a particular choice of a model parameterized by \( \eta \) will perform ideally; it only provides a quantitative measure of the best possible performance for such a model. This provides valuable insight into how well a model can perform, and can guide model development by suggesting the best parameters to provide minimal error in representation of the thermochemical state variables. The measure \( \epsilon_{\phi_i} \) provides a convenient error estimate for an ideal model parameterized by \( \eta \).

### 6.2. Single-Parameter Models

We first consider a class of models parameterized by the mixture fraction, \( f \). In nonpremixed combustion, stoichiometry is of primary importance. Thus, the mixture fraction is a logical choice for a reaction variable.

Figure 9(a) shows the DNS data projected into mixture fraction space (points), as well as the conditional mean temperature (solid line) and \( \sigma_T \) as defined by Equation (5) (dashed line). Figure 9(b) shows the normalized standard deviation, \( \epsilon_T \), as given by Equation (6). At the stoichiometric mixture fraction, \( f_{st} = 0.52 \), a single-parameter model based on \( f \) could predict temperature within an average of 10% error at best.

From a modeling perspective, the best-possible performance that a single-parameter model based on mixture fraction alone could achieve is correctly predicting \( \langle T | f \rangle \). Thus, the best possible accuracy that could be attained in predicting the temperature field for the DNS data in question is described by Figure 9(b).

One may consider two alternatives to obtain increased accuracy:
(i) Select a different parameter that may collapse the data better within the class of single-parameter models.

(ii) Add more parameters (reaction variables) to introduce more thermochemical degrees of freedom and thereby increase accuracy.

In the next section, we consider the second option.

6.3. Two-Parameter Models

One way to improve accuracy in the ability to represent the thermochemical state variables is by adding more reaction variables. In this section we choose the mixture fraction \( f \) as the first parameter and consider two choices for the second parameter:

(i) Parameterization by the mixture fraction and dissipation rate, \( \eta = (f, \chi_o) \). The parameter \( \chi_o \) is determined in terms of the true dissipation rate \( \chi = 2D \nabla f \cdot \nabla f \), as \([15, 27]\)

\[
\chi_o = \frac{\chi}{\exp \left[ -2 \left( \text{erf}^{-1} \left( 2f - 1 \right) \right)^2 \right]},
\]

where \( \text{erf}^{-1} \) denotes the inverse error function.

(ii) Parameterization by the mixture fraction and a progress variable, chosen based on CO for this study, \( \eta = (f, \eta_{CO}) \). Here \( \eta_{CO} \) is defined as \([14]\)

\[
\eta_{CO} = \frac{Y_{CO} - \beta_{CO}}{\alpha_{CO} - \beta_{CO}},
\]

where \( \alpha_{CO} = \max(Y_{CO} | f) \) and \( \beta_{CO} = \min(Y_{CO} | f) \).

The state variables from the DNS may be projected into the two-dimensional space parameterized by the reaction variables. For convenience, we only present results at stoichiometric mixture fraction, \( f_{st} \).

Figure 10 shows the temperature at stoichiometric conditions \( f_{st} = 0.52 \) as a function of \( \chi_o \) (Figure 10(a)) and \( \eta_{CO} \) (Figure 10(b)). Also shown is the conditional mean temperature (solid line) and \( \sigma_T \) (dashed line). Figure 10 demonstrates several important points. First, by adding a second parameter (either \( \chi_o \) or \( \eta_{CO} \)), the parameterization error is reduced. Secondly, the choice of the second parameter is quite important.

For the \((f, \chi_o)\) parameterization, a comparison of Figure 9(a) (at \( f_{st} \)) and Figure 10(a) shows that adding \( \chi_o \) as a second parameter reduces the error uniformly across all \( \chi_o \). However, choosing \( \eta_{CO} \) as the second parameter yields a maximum error at \( f_{st} \) of less than 2%. By comparison, choosing \( \chi_o \) as the second parameter yields at best an error of 6%; higher than the maximum error for the \((f, \eta_{CO})\) parameterization. This demonstrates that the second parameterization is a better choice to collapse the data.

In a model parameterized by \((f, \chi_o)\), the transport equations for these reaction variables do not include any chemical source terms, due to their conservative property. However, in the case of the \((f, \eta_{CO})\) parameterization, the reaction rate of CO appears in the \( \eta_{CO} \) transport equation of and must also be parameterized. Figure 11 shows the parameterization of the CO reaction rate \( \omega_{CO} \) as a function of \( \chi_o \) (Figure 11(a)) and \( \eta_{CO} \) (Figure 11(b)) at \( f_{st} \). (Figure 11(a) is included only as a point of reference.)

Referring to Figure 11(b), it is apparent that, although \((f, \eta_{CO})\) parameterizes many state variables with remarkable accuracy, it does not represent its source term as well. Unfortunately, \textit{a priori} analysis of the data cannot determine what effect a 10-25% error in \( \omega_{CO} \) will have on the ultimate model-predicted results. To determine the sensitivity of a model to errors in \( \omega_{CO} \), we must conduct \textit{a posteriori} analyses, which incorporate the feedback of errors in \( \omega_{CO} \) into the solution. Then, comparing those results to the results obtained from the full DNS, we could make a determination of the ultimate usefulness of a model based on \( \eta_{CO} \).
Figure 9. (a) Left: Temperature as a function of mixture fraction. Solid line shows conditional mean value; dashed line shows conditional standard deviation. (b) Right: Normalized conditional standard deviation, $\epsilon_T$.

Figure 10. Temperature as a function of $\chi$ ((a), left) and $\eta_{CO}$ ((b), right) at $f_{st}$. Solid line shows conditional mean value; dashed line shows conditional standard deviation.

Figure 11. CO reaction rate as a function of $\chi$ ((a), left) and $\eta_{CO}$ ((b), right) at $f_{st}$. Solid line conditional mean value; dashed line shows conditional standard deviation.
6.4. Implications
The present results show that a parameterization using mixture fraction alone results in fairly large errors in predicting state variables. For example, the average error in temperature is nearly 200 K at stoichiometric conditions. The two-parameter representations investigated both provide better collapse of the data. The new \((f, \eta_{CO})\) parameterization collapses the data better than the traditional \((f, \chi)\) parameterization. Further work is needed to assess how well this improved collapse will translate into improved models.

7. Conclusions
The S3D code is a mature massively parallel DNS solver that has been demonstrated here to scale very well to thousands of processors. This capability, together with Office of Science supercomputing platforms are enabling new combustion science to advance the development of predictive models.

In this paper, new results from computationally demanding three dimensional DNS of turbulent nonpremixed flames with detailed chemistry were presented. The large data size and complexity result in new challenges in knowledge extraction, which are being met by the application of innovative multi-scalar visualization techniques. Two examples were presented showing how the DNS data can be used to help assess and develop modeling approaches. In the first, it was shown how detailed transport and chemistry effects can impact the mixing rates of reacting scalars. In the second, it was shown how it is possible to explore different possible reduced parameterizations of the thermochemical state, and in particular that a two-variable parameterization employing mixture fraction and a progress variable based on the CO mass fraction provides an excellent state representation.

Work in this area is continuing, with further large simulations planned later this year using time from the INCITE grant at NERSC. Additional simulations are also planned at the NLCF. Ultimately, the simulation data will be shared with the turbulent combustion modelling community at large in order to maximize their impact.

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References
[1] Proceedings of Seventh International Workshop on the Measurement and Computation of Turbulent Nonpremixed Flames, 2004. http://www.ca.sandia.gov/TNF/7thWorkshop/TNF7.html.
[2] E. R. Hawkes and J.H. Chen. Direct numerical simulation of hydrogen-enriched lean premixed methane-air flames. Combust. Flame, 3(138):242–258, 2004.
[3] J. H. Chen and H. G. Im. Stretch effects on the burning velocity of turbulent premixed hydrogen/air flames. Proc. Combust. Inst., 28:211–218, 2000.

[4] R. Sankaran, H. G. Im, E. R. Hawkes, and J. H. Chen. The effects of nonuniform temperature distribution on the ignition of a lean homogeneous hydrogen-air mixture. Proc. Combust. Inst., 30:875–882, 2005.

[5] T. Echekki and J. H. Chen. Direct numerical simulation of autoignition in non-homogeneous hydrogen-air mixtures. Combust. Flame, 134:169–191, 2002.

[6] S. Mahalingam, J.H. Chen, and L. Vervisch. Finite-rate chemistry and transient effects in simulations of turbulent non-premixed flames. Combust. Flame, 102:285–297, 1995.

[7] J. C. Sutherland, P. J. Smith, and J. H. Chen. Quantification of differential diffusion in nonpremixed systems. Combust. Theory Modelling, 115:487–514, 2005.

[8] C. A. Kennedy, M. H. Carpenter, and R. M. Lewis. Low-storage, explicit runge-kutta schemes for the compressible Navier-Stokes equations. Appl. Num. Math., 14(4):397–433, 1994.

[9] C. A. Kennedy and M. H. Carpenter. Several new numerical methods for compressible shear-layer simulations. Appl. Num. Math., 14(4):397–433, 1994.

[10] T. J. Poinset and S. K. Lele. Boundary conditions for direct simulations of compressible viscous flows. J. Comp. Phys., 101(1):104–139, 1992.

[11] J. C. Sutherland and C. A. Kennedy. Improved boundary conditions for viscous, reacting, compressible flows. J. Comp. Phys., 191(2):502–524, 2003.

[12] C. Pantano. Direct simulation of nonpremixed flame extinction in a methane-air jet with reduced chemistry. J. Fluid Mech., 514, 2004. 231–270.

[13] P. Sripakagorn, S. Mitalai, G. Kosály, and H. Pitsch. Extinction and reignition in a diffusion flame (a direct numerical simulation study). J. Fluid Mech., 2004. submitted.

[14] J. C. Sutherland. Evaluation of Mixing and Reaction Models for Large-Eddy Simulation of Nonpremixed Combustion Using Direct Numerical Simulation. PhD thesis, University of Utah, Salt Lake City, UT, May 2004.

[15] N. Peters. Turbulent Combustion. Cambridge University Press, New York, 2000.

[16] R. A. Yetter, F. L. Dryer, and H. Rabitz. Flow reactor studies of carbon monoxide - hydrogen - oxygen kinetics. Comb. Sci. Tech., 79:97–128, 1991.

[17] J. Li, Z. Zhao, A. Kazakov, and F. L. Dryer. Int. J. Chem. Kinetics, 2005. to appear.

[18] H. Akiba, K. L. Ma, J. H. Chen, and E. R. Hawkes. Simultaneous visualization of simulated combustion data. In IEEE Visualization 2005 Conference, Minneapolis, MN, October 23-28, 2005. submitted.

[19] S. B. Pope. PDF methods for turbulent reacting flows. Prog. Energy Combust. Sci., 11:119–195, 1984.

[20] J. Villermaux and J. C. Devillon. In Proc. Second Int. Symp. On Chemical Reaction Engineering, New York, 1972. Elsevier.

[21] C. Dopazo and E. E. O’Brien. An approach to the autoignition of a turbulent mixture. Acta Astronaut., 1:1239–1264, 1974.

[22] J. Janicka, W. Kolbe, and W. Kollmann. Closure of the transport equation for the probability density function of turbulent scalar fields. J. Non-Equilib. Thermodyn., 4:47–66, 1977.

[23] S. Subramaniam and S. B. Pope. A mixing model for turbulent reactive flows based on Euclidean minimum spanning trees. Combust. Flame, 1998. to appear.

[24] S. B. Pope. Mixing models. In Proceedings of Sixth International Workshop on the Measurement and Computation of Turbulent Nonpremixed Flames, 2002.

[25] C. Pantano, S. Sarkar, and F. A. Williams. Mixing of a conserved scalar in a turbulent reacting shear layer. J. Fluid Mech., 481:291–328, 2003.

[26] A. N. Karpetis and R. S. Barlow. Measurements of flame orientation and scalar dissipation in turbulent partially premixed methane flames. Proc. Combust. Inst., 30:665–672, 2005.

[27] N. Peters. Laminar diffusion flamelet models in non-premixed turbulent combustion. Prog. Energy Combust. Sci., 10:319–339, 1984.