

Large eddy simulation of a turbulent diffusion flame with Conditional Source-term Estimation

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1. Motivation and objectives

Direct numerical simulation (DNS) of turbulent reacting flows at technically relevant high Reynolds numbers will be computationally unaffordable for the foreseeable future. Large eddy simulation (LES) represents an attractive alternative. The basic idea of LES is to resolve only the large scale motion of the flow while modeling the contribution of the small (i.e. sub-grid) scales, which tend to be more isotropic, hence, easier to model. Resolving only the large scales keeps the computational cost affordable. The LES set of equations is obtained by applying a spatial filter to the governing transport equations of mass, momentum and energy leading to

$$\frac{\partial \bar{\rho}}{\partial t} + \nabla (\bar{\rho} \tilde{\mathbf{u}}) = 0, \quad (1)$$

$$\frac{\partial \bar{\rho} \tilde{\mathbf{u}}}{\partial t} + \nabla (\bar{\rho} \tilde{\mathbf{u}} \tilde{\mathbf{u}}) = -\nabla \bar{p} + \nabla [\bar{\rho} (\tilde{\nu}_l + \tilde{\nu}_t) (\nabla \tilde{\mathbf{u}} + (\nabla \tilde{\mathbf{u}})^T)], \quad (2)$$

$$\frac{\partial \bar{\rho} \tilde{Y}_j}{\partial t} + \nabla (\bar{\rho} \tilde{\mathbf{u}} \tilde{Y}_j) = \nabla \left[\bar{\rho} (\tilde{D}_l + \tilde{D}_t)_j \nabla \tilde{Y}_j \right] + \bar{\Omega}_j, \quad (3)$$

$$\frac{\partial \bar{\rho} \tilde{h}}{\partial t} + \nabla (\bar{\rho} \tilde{\mathbf{u}} \tilde{h}) = \nabla \left[(\tilde{D}_l + \tilde{D}_t)_T \nabla \tilde{T} \right] + \sum_{j=1}^J (\bar{\Omega}_j h_{0,j}). \quad (4)$$

In order to account for the subgrid-scale (SGS) contributions to the fluxes of momentum, species, and enthalpy, the molecular viscosity $\tilde{\nu}_l$ and diffusivities \tilde{D}_l are augmented by an eddy viscosity $\tilde{\nu}_t$ and an eddy diffusivity \tilde{D}_t , respectively. Several SGS models for these turbulent transport coefficients have been suggested and successfully applied in many flow configurations (Lesieur & Métais, 1996). The dynamic models, which unlike the original Smagorinsky model (Smagorinsky, 1963) do not require a model constant to be set prior to the simulation, have become a well-established approach (Germano *et al.*, 1991). In the present LES the eddy viscosity and the eddy diffusivities are modeled using the dynamic model proposed by Moin *et al.* (1991) for compressible flows. In the case of reacting flows the spatially filtered chemical source terms occurring in the transport equations for reactive species and enthalpy represent an enormous additional challenge to LES. They represent the rate of change of species mass fraction Y_j due to chemical reaction and are a linear combination of the different reaction rates in which species j participates:

$$\bar{\Omega}_j = W_j \sum_{k=1}^K (\nu'_{jk} - \nu''_{jk}) \bar{\omega}_k. \quad (5)$$

Therein, K is the total number of reactions and $\nu_{jk'}$, and $\nu_{jk''}$ are the stoichiometric coefficients for species j in reaction k . The chemical reaction rates are in general given by Arrhenius-type equations

$$\dot{\omega}_k = A_k T^{n_k} \exp\left(-\frac{E_k}{RT}\right) \prod_{j=1}^J \left(\frac{\rho Y_j}{W_j}\right)^{\nu_{jk}}, \quad (6)$$

which are highly nonlinear functions of temperature T , density ρ , and the participating reactive species mass fractions Y_j . J is the total number of the reactive species, W_j denotes the molecular weight of species j , and ν_{jk} is the stoichiometric coefficient of species j in reaction k . A_k is the frequency factor, n_k is the pre-exponential temperature exponent, and E_k is the activation energy of reaction k , and R is the universal gas constant. Substituting the corresponding spatially filtered quantities $\bar{\rho}$, \bar{Y}_j and \bar{T} into Eq. (6) will generally lead to unacceptably erroneous predictions for the filtered reaction rates $\bar{\omega}_k$; thus, a model must be provided for $\bar{\omega}_k$.

The present LES considers a piloted methane-air jet flame with the Reynolds number $Re = 22400$. The feed stream consists of 25% (Vol.) methane and 75% (Vol.) air. The coflowing oxidizer stream is pure air, and the stoichiometric mixture fraction is $Z_{stoic} = 0.352$. The pilot stream surrounding the inner fuel jet resembles the hot product composition of a premixed methane-air flame with $Z_{pilot} = 0.27$. This flame, known as the ‘‘Sandia D-Flame’’, has been intensively investigated experimentally (Barlow, 1997; Barlow & Frank, 1998). A large amount of well documented experimental data for the individual species concentration fields is available. The LES of this flame attempts to assess the predictive capabilities of the Conditional Source-term Estimation method (CSE), which was recently proposed by Bushe & Steiner (1999), as well as the method’s robustness and computational cost. The CSE model has so far been tested only in an *a priori* test, where it gave promising results.

2. Modeling of the chemistry

In the context of turbulent non-premixed combustion, the mixture fraction Z plays an essential role. Z represents the local fraction of mass originating from the fuel stream. Many proposed models—e.g., the Equilibrium Chemistry Model (Cook & Riley, 1994), Laminar Flamelet Model (Peters, 1984; Cook & Riley, 1997), Conditional Moment Closure (CMC) (Klimenko, 1990; Bilger, 1993a)—rely on the mixture fraction Z and its probability density function (PDF) $P(Z)$. It is known that the individual reaction rates and the resulting chemical composition strongly depend on the state of mixedness described by Z and its PDF $P(Z)$. The probability density function $P(Z)$ can be obtained through a transport equation as suggested in the PDF-transport methods (Pope, 1985; Colucci *et al.*, 1998). Alternatively, the shape of the PDF can be presumed. In this case the presumed PDF can, for instance, be specified in terms of its first and second moment (\bar{Z} , \bar{Z}^2) given at every grid point (Frankel *et al.*, 1993). The method of ‘‘Conditional Source-term Estimation’’ (CSE), which is used in the present LES, is based on the CMC

approach. A detailed description of the CSE closure model can be found in previous work (Bushe & Steiner, 1999). In short, the CSE approach invokes the Conditional Moment Closure (CMC) hypothesis (Bilger, 1993a)

$$\langle \dot{\omega}_k | \zeta \rangle = f_k (\langle \rho | \zeta \rangle, \langle T | \zeta \rangle, \langle Y_j | \zeta \rangle), \quad (7)$$

where $\langle \dot{\omega}_k | \zeta \rangle$ denotes the average of the reaction rate $\dot{\omega}_k$ conditioned on the mixture fraction Z having some value ζ , or shortly, the “conditionally averaged reaction rate”. The conditionally averaged density, temperature, and mass fractions $\langle \rho | \zeta \rangle$, $\langle T | \zeta \rangle$ and $\langle Y_j | \zeta \rangle$ are input into the Arrhenius-type expression f_k on the RHS of Eq. (7). The CMC hypothesis (6) has proven to be a fairly accurate approximation applying data both from experiments and DNS (Bilger, 1993b; Bushe *et al.*, 1999). The original CMC method solves a transport equation for the conditional averages in mixture fraction space, which adds a further dimension to the problem and makes the computation exceedingly expensive. CSE proposes a computationally far less expensive alternative. In the CSE method the conditional averages are obtained by mapping the spatially filtered quantities from physical (LES) space into conditioning (i.e. Z) space. This mapping operation, e.g., for the mass fraction of the species j , is given by the integral equation

$$\int_0^1 \langle Y_j | \zeta \rangle \tilde{P}(\zeta; \mathbf{x}_m, t) d\zeta = \tilde{Y}_j(\mathbf{x}_m, t), \quad (8)$$

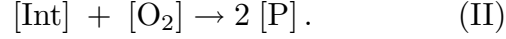
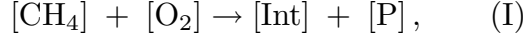
where $\tilde{P}(\zeta; \mathbf{x}_m, t)$ is the filtered probability density function (PDF) and $\tilde{Y}_j(\mathbf{x}_m, t)$ is the filtered mass fraction of the species j at some LES point \mathbf{x}_m at time t . Solving Eq. (8) on surfaces for which statistical homogeneity, hence invariance, for the conditional averages can be assumed (Bushe & Bilger, 1999), yields the conditionally averaged input quantities for Eq. (7) to give the reaction rates in conditioning space. Their unconditional spatially filtered reaction rates at some LES point \mathbf{x}_m at time t are then calculated using

$$\bar{\omega}_k(\mathbf{x}_m, t) = \bar{\rho}(\mathbf{x}_m, t) \int_0^1 \frac{\langle \dot{\omega}_k | \zeta \rangle}{\langle \rho | \zeta \rangle} \tilde{P}(\zeta; \mathbf{x}_m, t) d\zeta. \quad (9)$$

2.1 Reduced kinetic mechanism

The use of a very detailed chemical mechanism for methane-air diffusion flames involves a large number of reactive species and elementary reactions. While the CSE closure model is basically not restricted to strongly simplified kinetic mechanisms, it should be noted that every additional reaction requires at least one more transport equation, adding to the computational cost. Thus, a strongly reduced scheme, a two-step mechanism, was determined to be an appropriate choice for the present first test of CSE in a predictive LES, in that this reduced mechanism gives reasonably good predictions of methane-air flame properties while requiring minimal computational effort.

Based on steady-state and partial-equilibrium assumptions for various elementary steps in the complex methane-air reaction kinetics the detailed mechanism can be systematically reduced to the following two-step kinetic scheme (Williams, 1991):



The molar composition of the intermediate species [Int] and of product [P] is defined as

$$[\text{Int}] = \frac{4}{3} [\text{H}_2] + \frac{2}{3} [\text{CO}]$$

and

$$[\text{P}] = \frac{2}{3} [\text{H}_2\text{O}] + \frac{1}{3} [\text{CO}_2]$$

in moles $\cdot \text{cm}^{-3}$, respectively. The rates for reaction (I) and (II) are given by

$$\dot{\omega}_I = k_{11f} [\text{CH}_4][\text{H}], \quad (10)$$

$$\dot{\omega}_{II} = k_5 [\text{O}_2][\text{H}][\text{M}] \quad (11)$$

The steady-state assumption for the hydrogen radical [H] gives

$$[\text{H}] = (\text{K}_1\text{K}_2)^{\frac{1}{2}} \text{K}_3 \frac{[\text{O}_2]^{\frac{1}{2}} [\text{H}_2]^{\frac{3}{2}}}{[\text{H}_2\text{O}]} \Theta \left[1 - \exp\left(-\frac{\text{T}}{1000}\right)^5 \right], \quad (12)$$

where the quantity Θ is calculated following a proposal by Peters (1995):

$$\Theta = \exp \left[- \left(\frac{15}{4} \right)^{\frac{1}{2}} \frac{k_{11f} [\text{CH}_4]}{k_{1f} [\text{O}_2]} \right]. \quad (13)$$

The rate coefficients k_{1f}, k_5, k_{11f} as well as the equilibrium constants $\text{K}_1, \text{K}_2, \text{K}_3$ refer to the skeletal methane-air mechanism as listed in literature (Smooke, 1991); units in Eqs. (10)-(13) are moles, centimeters, grams, seconds, and Kelvins. The third body concentration in Eqs. (11) is

$$[\text{M}] = 6.5 [\text{CH}_4] + 6.5 [\text{H}_2\text{O}] + 1.5 [\text{CO}_2] + [\text{H}_2] + 0.75 [\text{CO}] + 0.4 [\text{O}_2] + 0.4 [\text{N}_2]. \quad (14)$$

The rates of change in the participating mass fractions due to the chemical reactions expressed in terms of $\dot{\omega}_I$ and $\dot{\omega}_{II}$ are

$$\dot{\Omega}_{\text{CH}_4} = -W_{\text{CH}_4} \dot{\omega}_I, \quad (15)$$

$$\dot{\Omega}_{\text{O}_2} = -W_{\text{O}_2} (\dot{\omega}_I + \dot{\omega}_{II}), \quad (16)$$

$$\dot{\Omega}_{\text{Int}} = W_{\text{Int}} (\dot{\omega}_I - \dot{\omega}_{II}), \quad (17)$$

$$\dot{\Omega}_{\text{P}} = W_{\text{P}} (\dot{\omega}_I + 2\dot{\omega}_{II}) \quad (18)$$

with W_{CH_4} , W_{O_2} , W_{Int} and W_{P} being the molecular weights of fuel, oxidizer, intermediate species, and product, respectively. The source term in the transport equation for enthalpy (4) reads

$$\dot{\Omega}_T = \sum_j \left(\dot{\Omega}_j h_{0,j} \right) = \dot{\omega}_I Q_I + \dot{\omega}_{II} Q_{II}, \quad \text{with } j = \text{CH}_4, \text{O}_2, \text{Int}, \text{P}, \quad (19)$$

where the reactive heat release due to reaction I and II is linearly composed of the enthalpies of formation of fuel, intermediate species, and product as follows:

$$\begin{aligned} Q_I &= h_{0,\text{CH}_4} - \overbrace{\frac{2}{3}h_{0,\text{CO}}}^{-h_{0,\text{Int}}} - \overbrace{\frac{2}{3}h_{0,\text{H}_2\text{O}} - \frac{2}{3}h_{0,\text{CO}_2}}^{-h_{0,\text{P}}}, \\ Q_{II} &= h_{0,\text{Int}} - 2h_{0,\text{P}}. \end{aligned}$$

The numerical values of Q_I and Q_{II} are $291 \cdot 10^3$ and $511 \cdot 10^3$ J · mol⁻¹, respectively.

The concentration of the radical [H], as is specified in Eqs. (12) and (13), is very sensitive to temperature and fuel concentration [CH₄]. The decrease of temperature together with the increase of the fuel mass fraction on the rich side of the diffusion flame gives [H] = 0, shutting off the chemistry so oxidizer can leak through the reaction zone to the fuel rich side as is the case at high strain rates. On the lean side, however, the reaction rate $\dot{\omega}_I$ drops to zero only if the fuel is burned completely – fuel cannot leak to the lean side. By allowing O₂ leakage but not CH₄ leakage in highly strained flames, this two-step mechanism qualitatively exhibits behavior found in experiment (Williams, 1991). Moreover, in the present case, the fuel stream is diluted with air. The shut-off of the reactions as the H radical concentration drops to zero on the rich side prevents the flame from penetrating too far into the partially premixed fuel zone.

2.2 CSE closure for two-step chemistry

Invoking the CMC closure hypothesis (7) with the present two-step chemistry yields for the conditional averages of rate (I) and (II)

$$\langle \dot{\omega}_I | \zeta \rangle = f_I (\langle T | \zeta \rangle, \langle Y_{\text{CH}_4} | \zeta \rangle, \langle Y_{\text{O}_2} | \zeta \rangle, \langle Y_{\text{Int}} | \zeta \rangle, \langle Y_{\text{P}} | \zeta \rangle), \quad (20)$$

$$\langle \dot{\omega}_{II} | \zeta \rangle = f_{II} (\langle T | \zeta \rangle, \langle Y_{\text{CH}_4} | \zeta \rangle, \langle Y_{\text{O}_2} | \zeta \rangle, \langle Y_{\text{Int}} | \zeta \rangle, \langle Y_{\text{P}} | \zeta \rangle), \quad (21)$$

where f_I and f_{II} include all the functional dependencies described in Eqs. (10)-(14). For the case of low Mach number for which thermodynamic pressure p_0 is constant, the conditional density can be expressed in terms of temperature and chemical composition using the equation of state

$$\langle \rho | \zeta \rangle = \frac{p_0}{\langle T | \zeta \rangle \sum_j \langle Y_j | \zeta \rangle \frac{R}{W_j}}, \quad \text{with } j = \text{CH}_4, \text{O}_2, \text{Int}, \text{P},$$

and does not appear in Eqs. (20) and (21). Rather than solving the integral equation given by Eq. (8) for every conditionally averaged quantity needed in f_I and f_{II} , the conditional averages for the species mass fractions and temperature can be rewritten as functions of two conditional progress variables. These two new variables, the fuel-progress variable C_1 and the oxidizer-progress variable C_2 , are defined in physical (LES) space as the difference of the actual fuel and oxidizer mass fractions to the corresponding chemically inert mixed states $\tilde{Y}_{\text{CH}_4}^*$ and $\tilde{Y}_{\text{O}_2}^*$, respectively:

$$\tilde{C}_1 = \tilde{Y}_{\text{CH}_4}^* - \tilde{Y}_{\text{CH}_4}, \quad (22)$$

$$\tilde{C}_2 = \tilde{Y}_{\text{O}_2}^* - \tilde{Y}_{\text{O}_2}. \quad (23)$$

The quantities $\tilde{Y}_{\text{CH}_4}^*$ and $\tilde{Y}_{\text{O}_2}^*$, which are associated with $\tilde{C}_1 = 0$ and $\tilde{C}_2 = 0$, respectively, represent mixing of fuel and oxidizer without any chemical reaction; they are linear functions of the mixture fraction \tilde{Z} only. In the mixing configuration of the present jet simulation they read

$$\tilde{Y}_{\text{CH}_4}^* = \tilde{Z} \tilde{Y}_{\text{CH}_4,1}, \quad (24)$$

$$\tilde{Y}_{\text{O}_2}^* = \tilde{Y}_{\text{O}_2,2} + \tilde{Z} (\tilde{Y}_{\text{O}_2,1} - \tilde{Y}_{\text{O}_2,2}), \quad (25)$$

where $Y_{\text{CH}_4,1}$ and $Y_{\text{O}_2,1}$ is the methane and oxygen mass fraction in the feed fuel stream, respectively, and $Y_{\text{O}_2,2}$ is the oxygen mass fraction in the oxidizer stream. Assuming no differential diffusion, i.e., unity Lewis number, the spatially filtered mixture fraction \tilde{Z} is defined as a linear combination of the individual filtered species mass fractions

$$\tilde{Z} = \frac{1}{Y_{\text{CH}_4,1} + Y_{\text{O}_2,2} - Y_{\text{O}_2,1}} \left(\tilde{Y}_{\text{CH}_4} - \frac{2W_{\text{CH}_4}}{W_{\text{O}_2}} \tilde{Y}_{\text{O}_2} - \frac{W_{\text{CH}_4}}{W_{\text{P}}} \tilde{Y}_{\text{P}} + Y_{\text{O}_2,2} \right). \quad (26)$$

The coefficients in Eq. (26) were determined so that the linear combination of the transport equations for \tilde{Y}_{CH_4} , \tilde{Y}_{O_2} and \tilde{Y}_{P} has no chemical source term and the boundary conditions— $\tilde{Z} = 0$ in the pure feed oxidizer stream and $\tilde{Z} = 1$ in the pure feed fuel stream—are satisfied.

The intermediate and product mass fractions can be expressed as linear combinations of \tilde{C}_1 , \tilde{C}_2 and \tilde{Z} :

$$\tilde{Y}_{\text{Int}} = \left(\frac{W_{\text{P}}}{W_{\text{CH}_4}} + 1 \right) \tilde{C}_1 - \left(\frac{2W_{\text{P}}}{W_{\text{O}_2}} - 1 \right) \tilde{C}_2 + \frac{W_{\text{P}}}{W_{\text{CH}_4}} \left(\frac{2W_{\text{CH}_4}}{W_{\text{O}_2}} - 1 \right) \tilde{Y}_{\text{O}_2}^*. \quad (27)$$

$$\tilde{Y}_{\text{P}} = \frac{2W_{\text{P}}}{W_{\text{O}_2}} \tilde{C}_2 - \frac{W_{\text{P}}}{W_{\text{CH}_4}} \tilde{C}_1 - \frac{W_{\text{P}}}{W_{\text{CH}_4}} \left(\frac{2W_{\text{CH}_4}}{W_{\text{O}_2}} - 1 \right) \tilde{Y}_{\text{O}_2}^*, \quad (28)$$

For the temperature the analogous linear functional dependence $\tilde{T} = \tilde{T}(\tilde{C}_1, \tilde{C}_2, \tilde{Z})$ is obtained by incorporating Eqs. (22)-(28) into the equation for the total enthalpy

$$c_p (\tilde{T} - T_0) + \sum_j (\tilde{Y}_j - \tilde{Y}_j^*) \frac{h_{0,j}}{W_j} = 0, \quad \text{with } j = \text{CH}_4, \text{O}_2, \text{Int}, \text{P}, \quad (29)$$

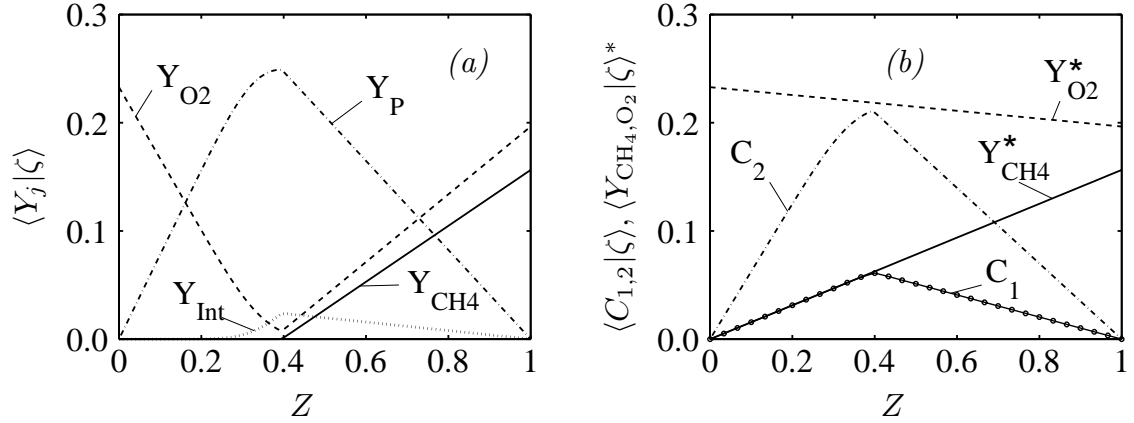


FIGURE 1. Typical structure of a strained diffusion flame for the reduced two-step mechanism: (a) conditional mass fractions $\langle Y_j | \zeta \rangle$ with $j = CH_4, O_2, Int, P$. (b) conditional fuel and oxidizer progress variables $\langle C_1 | \zeta \rangle$ and $\langle C_2 | \zeta \rangle$ corresponding to (a), and chemically inert mixed lines $\langle Y_{CH_4} | \zeta \rangle^*$ and $\langle Y_{O_2} | \zeta \rangle^*$

where T_0 is the reference temperature and the quantities with the asterisk again represent the chemically inert mixed state. Mapping the unconditional quantities $\tilde{Y}_j(\tilde{C}_1, \tilde{C}_2, \tilde{Z})$ and $\tilde{T}(\tilde{C}_1, \tilde{C}_2, \tilde{Z})$ from physical space into conditioning or mixture fraction space, using the integral relation as shown in Eq. (8), the corresponding conditional quantities are obtained as linear combinations of the conditional fuel-progress and oxidizer-progress variable $\langle C_1 | \zeta \rangle$ and $\langle C_2 | \zeta \rangle$, respectively:

$$\langle Y_{CH_4} | \zeta \rangle = \langle Y_{CH_4} | \zeta \rangle^* - \langle C_1 | \zeta \rangle, \quad (30)$$

$$\langle Y_{O_2} | \zeta \rangle = \langle Y_{O_2} | \zeta \rangle^* - \langle C_2 | \zeta \rangle, \quad (31)$$

$$\langle Y_{Int} | \zeta \rangle = \left(\frac{W_P}{W_{CH_4}} + 1 \right) \langle C_1 | \zeta \rangle - \left(\frac{2W_P}{W_{O_2}} - 1 \right) \langle C_2 | \zeta \rangle, \quad (32)$$

$$\langle Y_P | \zeta \rangle = 2 \frac{W_P}{W_{O_2}} \langle C_2 | \zeta \rangle - \frac{W_P}{W_{CH_4}} \langle C_1 | \zeta \rangle, \quad (33)$$

$$\langle T | \zeta \rangle = T_0 + \frac{Q_I - Q_{II}}{W_{CH_4}} \langle C_1 | \zeta \rangle + \frac{Q_{II}}{W_{O_2}} \langle C_2 | \zeta \rangle \quad (34)$$

with the molecular weights of the individual mass species being $W_{CH_4} = 16$, $W_{O_2} = 32$, $W_{Int} = 64/3$, $W_P = 80/3 \text{ g} \cdot \text{mol}^{-1}$. Fig. 1a shows the structure of a strained diffusion flame in conditioning space as it is typically obtained with the present two-step mechanism and stoichiometry. The corresponding fuel-progress variable $\langle C_1 | \zeta \rangle$ and oxidizer-progress variable $\langle C_2 | \zeta \rangle$ in mixture fraction (Z) space as well as the chemically inert straight mixed lines — $\langle Y_{CH_4} | \zeta \rangle^*$ in Eq. (30) and $\langle Y_{O_2} | \zeta \rangle^*$ in Eq. (31)— are shown in Fig. 1b.

Using Eqs. (30) and (31) an integral equation similar to Eq. (8) can be written for the two conditional variables $\langle C_1 | \zeta \rangle$ and $\langle C_2 | \zeta \rangle$

$$\tilde{Y}_{CH_4}^* - \tilde{Y}_{CH_4} = \int_0^1 \langle C_1 | \zeta \rangle \tilde{P}(\zeta; \mathbf{x}, t) d\zeta, \quad (35)$$

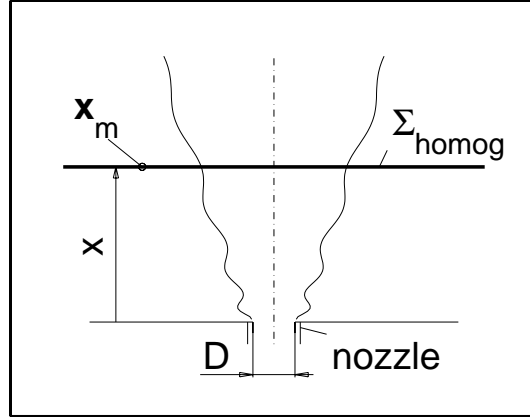


FIGURE 2. Planes Σ_{homog} across the jet's shear layer on which statistical homogeneity for the conditional averages is assumed.

$$\tilde{Y}_{\text{O}_2}^* - \tilde{Y}_{\text{O}_2} = \int_0^1 \langle C_2 | \zeta \rangle \tilde{P}(\zeta; \mathbf{x}, t) d\zeta, \quad (36)$$

where the shape of the filtered PDF at grid point \mathbf{x} and time t was assumed to be a β -function (Cook & Riley, 1994), which is specified by the instantaneous spatially filtered mean $\tilde{Z}(\mathbf{x}, t)$ and variance $\tilde{Z}''^2(\mathbf{x}, t)$

$$\tilde{P}(\zeta; \mathbf{x}, t) = \frac{\zeta^{a-1} (1-\zeta)^{b-1}}{\beta(a, b)}, \quad a = \tilde{Z} \left[\frac{\tilde{Z} (1 - \tilde{Z})}{\tilde{Z}''^2} - 1 \right], \quad b = a \left(\frac{1}{\tilde{Z}} - 1 \right).$$

Therein, the spatially filtered mixture fraction \tilde{Z} is expressed as a linear combination of the filtered species mass fractions as given in Eq. (26). The filtered variance \tilde{Z}''^2 is modeled using a dynamic procedure (Pierce & Moin, 1998). Thus, it is not necessary to solve an extra transport equation for either the filtered mean or the filtered variance of the mixture fraction. The conditional averages depend on the conditional scalar dissipation, which is a strong function of the downstream coordinate but has only a weak dependence on the radial, or crossflow, direction. Thus, statistical homogeneity of the conditional averages is assumed on planes of constant downstream distance from the nozzle (see Fig. 2).

Writing the integral Eqs. (35) and (36) at every LES sample point \mathbf{x}_m : $m = 1, \dots, M$, located on each of these planes establishes a linear system equations for $\langle C_1 | \zeta \rangle$ and $\langle C_2 | \zeta \rangle$. Inserting the solutions of this linear system into the RHS of the Eqs. (30)-(34) provides the conditionally averaged temperature and species mass fractions needed for the closure hypothesis in Eqs. (20) and (21) on each plane. The integration of these conditional averages with the PDF over ζ gives the unconditional spatially filtered values for the reaction rates $\bar{\omega}_I$ and $\bar{\omega}_{II}$ at every

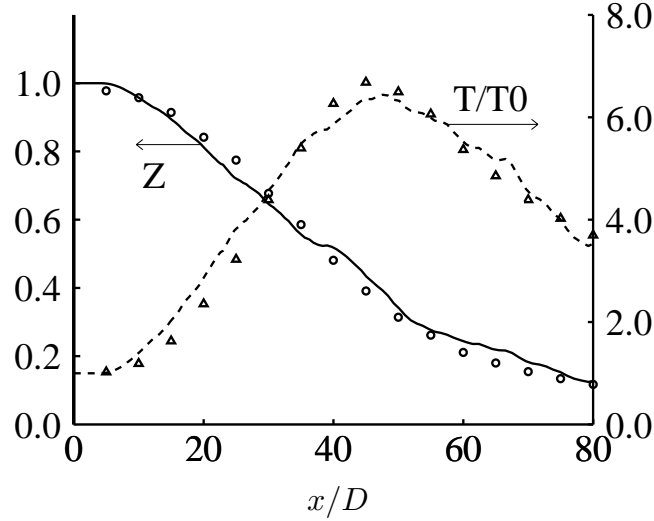


FIGURE 3. Mixture fraction Z and temperature T/T_0 normalized with the reference temperature $T_0 = 291\text{K}$ at the centerline of the jet versus downstream distance in nozzle diameters D ; — : LES result for mixture fraction; ---- : LES result for temperature; symbols denote the experimental data.

point \mathbf{x}_m :

$$\begin{aligned}\bar{\omega}_I(\mathbf{x}_m, t) &= \bar{\rho}(\mathbf{x}_m, t) \int_0^1 \frac{\langle \dot{\omega}_I | \zeta \rangle}{\langle \rho | \zeta \rangle} \tilde{P}(\zeta; \mathbf{x}_m, t) d\zeta, \\ \bar{\omega}_{II}(\mathbf{x}_m, t) &= \bar{\rho}(\mathbf{x}_m, t) \int_0^1 \frac{\langle \dot{\omega}_{II} | \zeta \rangle}{\langle \rho | \zeta \rangle} \tilde{P}(\zeta; \mathbf{x}_m, t) d\zeta.\end{aligned}$$

Expressing the thermodynamic state in terms of two progress variables reduces the computational cost considerably because only two integral equations (35) and (36) have to be inverted. The LHS of these equations is obtained from the filtered fields of the mass fractions of fuel and oxidizer. Only two scalar transport equations (3) have to be solved for \tilde{Y}_{CH_4} and \tilde{Y}_{O_2} , whose source terms $\tilde{\Omega}_{\text{CH}_4}$ and $\tilde{\Omega}_{\text{O}_2}$ are related to $\bar{\omega}_I$ and $\bar{\omega}_{II}$ by Eqs. (15) and (16), respectively. The spatially filtered instantaneous values of the remaining scalars involved in the two-step mechanism, i.e., intermediate species \tilde{Y}_{Int} , product \tilde{Y}_{P} , and temperature \tilde{T} , are calculated by substituting the corresponding conditionally averaged quantities provided by Eqs. (32)-(34) into the integrals

$$\begin{aligned}\tilde{Y}_{\text{Int}}(\mathbf{x}_m, t) &= \int_0^1 \langle Y_{\text{Int}} | \zeta \rangle \tilde{P}(\zeta; \mathbf{x}_m, t) d\zeta, \\ \tilde{Y}_{\text{P}}(\mathbf{x}_m, t) &= \int_0^1 \langle Y_{\text{P}} | \zeta \rangle \tilde{P}(\zeta; \mathbf{x}_m, t) d\zeta, \\ \tilde{T}(\mathbf{x}_m, t) &= \int_0^1 \langle T | \zeta \rangle \tilde{P}(\zeta; \mathbf{x}_m, t) d\zeta.\end{aligned}$$

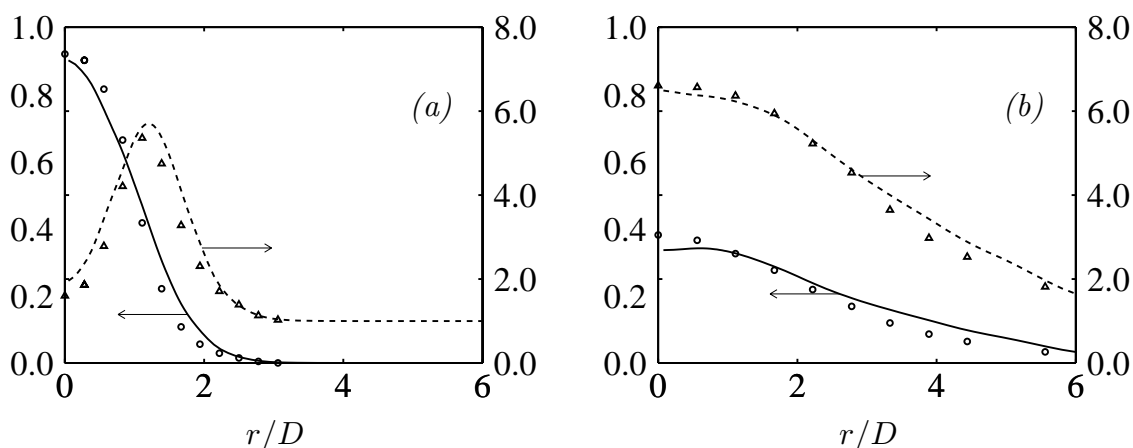


FIGURE 4. Mixture fraction Z and temperature T/T_0 normalized with the reference temperature $T_0 = 291\text{K}$ versus radial direction in jet diameters D at two different downstream locations: (a) at $x = 15$, (b) at $x = 45$ diameters downstream the nozzle; — : LES results for Z ; ---- : LES results for T/T_0 ; symbols denote the measurements.

3. Accomplishments

The computational code used for the present LES is based on a code originally developed for DNS of non-reacting, round, free jets at low Mach number (Boersma, 1998). The LES version of that code was obtained by implementing dynamic subgrid-scale models for the unresolved turbulent transport and the CSE closure model for the chemical reaction terms. The LES was performed on a computational grid in spherical coordinates with $192 \times 84 \times 48$ points in stream-wise, cross-stream, and tangential direction of the jet. The profile of the time-averaged mixture fraction and temperature—the latter normalized with the inflow temperature of the coflowing air stream, $T_0 = 291\text{K}$ —on the centerline and radial distribution at various locations downstream the nozzle are shown in Figs. 3 and 4a,b, respectively. The agreement with the experimental data is reasonable, implying that the heat release is predicted fairly accurately by the model.

Figures 5a-d show the radial profiles of time-averaged species mass fractions at two locations downstream the nozzle. The agreement of the LES results with the experiment is good. There is, however, an underprediction of intermediate species and an overprediction of product. This is a result of discrepancies in the prediction of conditional averages; as shown in Figs. 6a-d the conditional averages computed according to the procedure described in Eqs. (30)-(36) also exhibit the aforementioned disagreement for the intermediate species and product. Some part of these discrepancies can be attributed to the extreme simplicity of the chemical kinetic mechanism which was applied in the present LES as derived in literature (Williams, 1991) without adjusting any of the mechanism's parameters. Although this two-step mechanism mimics qualitatively the structure of methane-air diffusion flames very well, it is conceivable that simplifying the very complex methane-air chemistry to just two reactions will give quantitatively less accurate predictions than the full

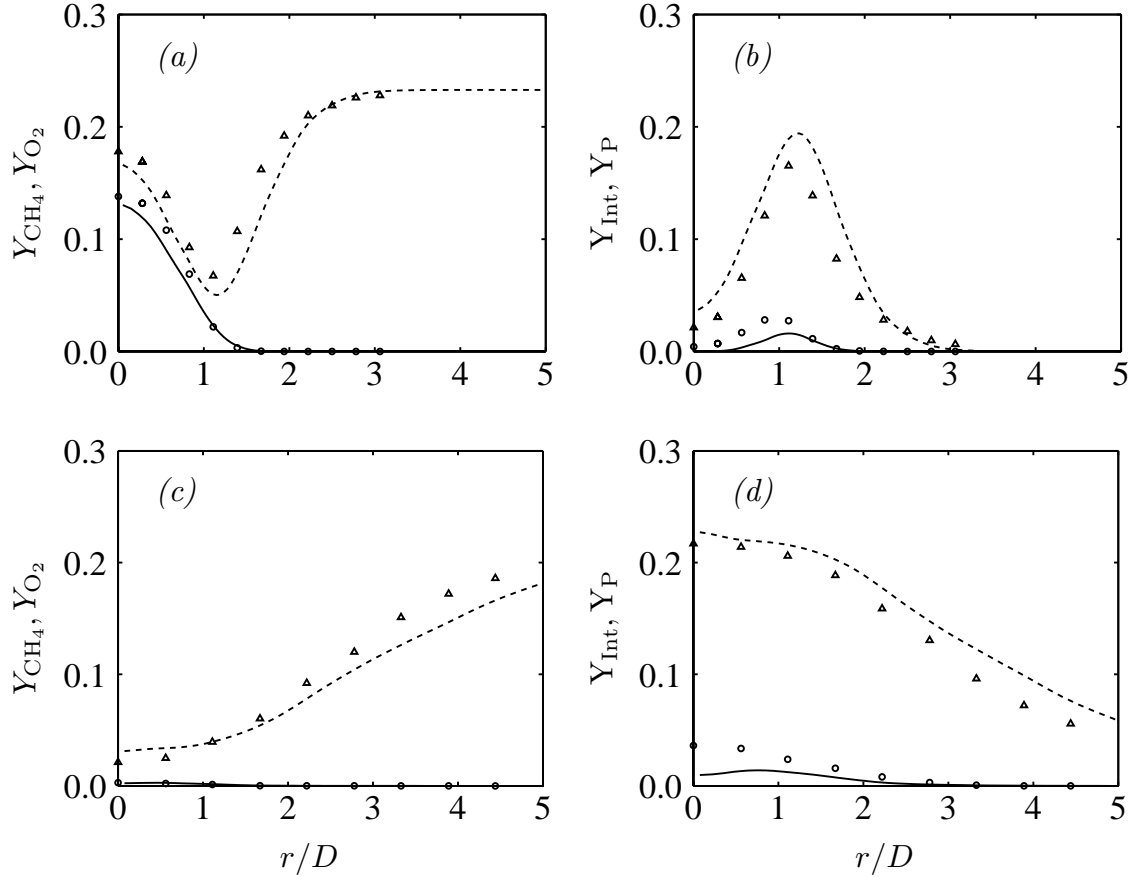
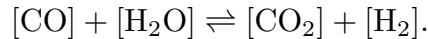


FIGURE 5. Species mass fractions of fuel Y_{CH_4} , oxidizer Y_{O_2} , intermediate Y_{Int} and product Y_{P} versus radial direction in jet diameters D at different downstream locations: (a), (b) at $x = 15$, (c), (d) at $x = 45$ diameters downstream the nozzle; — : LES results for Y_{CH_4} [(a),(c)] and Y_{Int} [(b),(d)]; ---- : LES results for Y_{O_2} [(a),(c)] and Y_{P} [(b),(d)], respectively; symbols denote the measurements.

detailed mechanism. In deriving the two-step mechanism one of the major reduction steps involves the partial equilibrium assumption for the water-gas shift



Applied to the present flame this partial-equilibrium evidently causes an underprediction of the concentrations of $[\text{CO}]$, the principal intermediate species. Using a more detailed chemical kinetic mechanism would help to overcome this problem. It should be noted, however, that computational cost is an important issue in the choice of a chemical mechanism to be employed in CSE. As already pointed out above, every further reaction accounted requires at least one more scalar transport equation in the resolved physical space plus one more mapping procedure (Eq. (35)) for the corresponding conditional average in mixture fraction space. The main objective of the present LES was to assess the feasibility and predictive capabilities of the CSE method in a self-sustained LES. The two-step mechanism was chosen

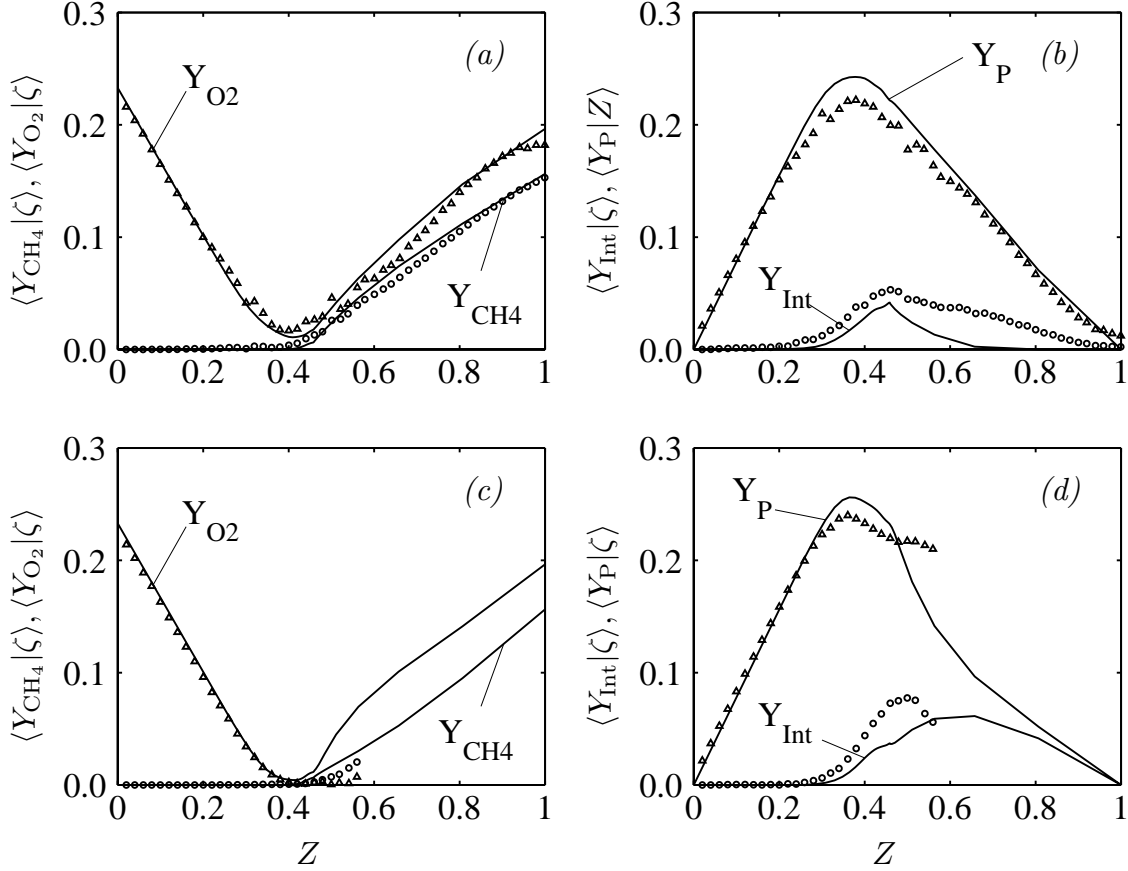


FIGURE 6. Conditional averages of species mass fractions of fuel $\langle Y_{\text{CH}_4} | Z \rangle$, oxidizer $\langle Y_{\text{O}_2} | Z \rangle$, intermediate $\langle Y_{\text{Int}} | Z \rangle$ and product $\langle Y_{\text{P}} | Z \rangle$: (a), (b) at $x = 15$ diameters downstream the nozzle; (c), (d) at $x = 45$ diameters downstream the nozzle; — : CSE results; symbols denote the measurements.

as the least computationally expensive methane-air mechanism which can be applied to the particular configuration given in the present jet flame. Applying very detailed chemistry was determined to be beyond the scope of this study. A close examination of the experimental data on the rich side of the flame unveils something else that should not be overlooked: the partial premixedness of the feed fuel stream. The experimental data of the conditional averages of the intermediate species and product are shown in Fig. 6b and exhibit a slightly negative curvature on the fuel side of the diffusion flame around mixture fraction $Z = 0.6$. Assuming rather a physical background than experimental uncertainties as a possible reason for this curvature, it can be concluded that there is some reactive formation of intermediate species and product in this partially premixed fuel region. It is conceivable that the occurrence of a premixed reaction zone might contribute to the disagreement of the model's predictions with experiment on the partially premixed rich side as well.

4. Future work

The test simulation of a piloted jet reported here has shown the method to be

a tool with satisfactory predictive capabilities. The results of this LES have also shown that the simple two-step kinetic methane-air mechanism clearly has its limits in producing quantitatively accurate predictions for all the species involved. In particular, the disagreement of the intermediate species with the experiment suggests the application of more detailed chemistry for future work. Full or large skeletal kinetic mechanisms have to be ruled out as they would make the LES with CSE closure computationally too expensive. A compromise between very accurate chemistry and computational cost has to be made. Thus, a four-step, or eventually, a five-step mechanism will be considered in the very next step of future work. Later on a second conditioning variable should be introduced into the model. Using a quantity which is closely related to the scalar dissipation rate should make it possible to account for extinction and ignition phenomena.

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