Turbulent Scalar Fluxes in H_2 -Air Premixed Flames at Low and High Karlovitz Numbers

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The statistical behaviour and closure of sub-grid scalar fluxes in the context of turbulent premixed combustion has been assessed based on an *a priori* analysis of a detailed chemistry Direct Numerical Simulation (DNS) database consisting of three hydrogen air flames spanning the corrugated flamelets (CF), thin reaction zones (TRZ) and broken reaction zones (BRZ) regimes of premixed turbulent combustion. The sub-grid scalar fluxes have been extracted by explicit filtering of DNS data. It has been found that the conventional gradient hypothesis model is not appropriate for the closure of sub-grid scalar flux for any scalar in the context of a multi-species system. However, the predictions of the conventional gradient hypothesis exhibit a greater level of qualitative agreement with DNS data for the flame representing the BRZ regime. The aforementioned behaviour has been analysed in terms of the properties of the invariants of the anisotropy tensor in the Lumley triangle. The flames in the CF and TRZ regimes are characterised by a pronounced two dimensional anisotropy due to strong heat release whereas a three dimensional and more isotropic behaviour is observed for the flame located in the BRZ regime. Two sub-grid scalar flux models which are capable of predicting counter-gradient transport have been considered for *a priori* DNS assessment of multi-species systems and have been analysed in terms of both qualitative and quantitative agreements. By combining the latter two sgs flux closures a new modelling strategy is suggested where one model is responsible for properly predicting the conditional mean accurately and the other model is responsible for the correlations between model and unclosed term. Detailed physical explanations for the observed behaviour and an assessment of existing modelling assumptions have been provided. Finally, the classical Bray-Moss-Libby theory for the scalar flux closure has been extended to address multispecies transport in the context of Large Eddy Simulations (LES).

1. Introduction

The turbulent scalar flux closure plays a pivotal role in both Reynolds Averaged Navier-Stokes (RANS) and Large Eddy Simulations (LES). The closure of turbulent scalar flux in premixed flames is further complicated by the possibility of counter-gradient transport (CGT) and its theoretical justification was provided in [1–3]. The CGT in premixed turbulent flames has been confirmed in several previous experimental (e.g. [2,4–9]) and Direct Numerical Simulations (DNS) (e.g. [10–24]) analyses. Veynante et al. [11] have pointed out based on DNS data that the competition between scalar transport due to turbulent velocity fluctuations and flame normal acceleration determines the statistical behaviour of turbulent scalar flux in RANS modelling:

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when the effects of flame normal acceleration are stronger than the effects of turbulent velocity fluctuation a counter-gradient transport is observed and *vice versa*. A thorough account of experimental and DNS contributions to RANS modelling of turbulent scalar flux can be found in [24] and references therein. In comparison to RANS based scalar flux modelling (e.g. [1–8,10–12,15,16,18–20,24], the closure of turbulent sub-grid scale (SGS) scalar flux in the context of LES has received limited attention [9,13,14,17,21–23,25–28] and interested authors are directed to [21–23] for a detailed review of sub-grid scalar flux closures of premixed turbulent combustion. The present analysis will henceforth focus primarily on SGS scalar flux closure in the context of LES.

The majority of the computational and theoretical analyses discussed earlier were carried out subject to the assumptions of adiabaticity and equi-diffusivity. In this scenario the temperature and the mass fractions of the reactive species within premixed deflagration flames can be uniquely expressed as a function of the reaction progress variable c , whose transport equation is derived from that of a scalar variable. In the formulation for LES, filtering the convective term for the progress variable results in the unclosed SGS scalar flux $\tau_{i,c} = \bar{\rho}(\widetilde{u_i c} - \tilde{u}_i \tilde{c})$, where the overbar denotes a normal filtering and the tilde represents the Favre-filtered value of a general quantity Q given by $\tilde{Q} = \bar{\rho}Q/\bar{\rho}$. However, the above simplification of chemistry might lead to inexact predictions especially in the presence of differential diffusion of heat and mass. These differential diffusion effects are not necessarily limited to low Reynolds numbers [29,30] and interested readers are requested to refer to the extensive overview by Lipatnikov and Chomiak [31] and the recent investigations by Venkateswaran et al. [32] and Burluka et al. [33] for experimental verification of the sustenance of differential diffusion effects for high values of Reynolds number. Recent studies have also reported that effects of differential diffusion cannot be ignored in the context of LES [21,23]. With the advances in computing power, the modelling challenges arising from the differential diffusion of heat and mass may be addressed by solving the individual scalar transport equations in LES with detailed chemistry models [34,35]. In such an approach the SGS flux

$$\tau_{i,m} = \bar{\rho}(\widetilde{u_i Y_m} - \tilde{u}_i \tilde{Y}_m) \quad (1)$$

needs to be modelled for each reactive species conservation equation. Little is known about the behaviour of τ_{im} where m might be a reactant, a product, a radical or an intermediate species in a multi-species system. Therefore, the objective of this paper is to analyse the statistical behaviours of the SGS species fluxes in the context of LES by a-priori analysis of a detailed chemistry DNS database for hydrogen-air premixed flames representing three different regimes of turbulent combustion, where it can be expected that the nature of the scalar flux shows considerably different behaviour. Moreover, explicitly filtered DNS data has been used to assess if the SGS scalar flux models, which have been found to perform satisfactorily for τ_{ic} based on a-priori analysis of single step chemistry DNS data, also offer accurate predictions for τ_{im} in the context of a multi-species system. Based on this exercise, the modifications to the existing models have been proposed wherever necessary for the purpose of closure of SGS flux of especially intermediate species.

The rest of the paper is organised as follows. The DNS data used in this analysis will be introduced in the next section of this paper. This will be followed by the discussions on the statistical behaviours of SGS fluxes and their modelling in the next three sections. Main findings will be summarised and conclusions will be drawn at the

Table 1. List of inflow turbulence parameters

Case	u'/S_L	l_T/δ_{th}	Re_t	Da	Ka
A	0.7	14.0	227	20.0	0.75
B	5	14.0	1623	2.8	14.4
C	14	4.0	1298	0.29	126

final section of this paper.

2. Description of computational simulations

The DNS database used for this analysis [36] is taken from three-dimensional compressible, low Mach number simulations of freely-propagating statistically planar turbulent H_2 -air premixed flames with $\phi = 0.7$, employing a detailed chemical mechanism [37] with 9 steps and 19 chemical reactions. The unburned gas temperature T_0 is taken to be 300K, which yields an unstrained laminar burning velocity $S_L = 135.62 \text{ cm/s}$ under atmospheric pressure. The flame is initialised by a steady, 1D planar laminar flame profile. A pre-computed auxiliary divergence free, homogeneous, isotropic turbulence field generated using a pseudo-spectral method [38] following Passot-Pouquet spectrum [39] is injected through the inlet. The mean inlet velocity has been gradually modified to match turbulent flame speed as the simulation progresses. Nevertheless, it is difficult to keep the flame within the computational domain for a long run-time. Turbulence decays in axial direction and as the flame moves back and forth it experiences different turbulence intensities which gives rise to an unsteady behaviour and fluctuating flame area (respectively turbulent flame speed). A detailed discussion of alternative configurations for turbulent planar flame configurations with their advantages and disadvantages is given in Ref. [49].

A turbulent outflow boundary is specified using an improved Navier Stokes characteristic boundary conditions (NSCBC) technique [40] in the direction opposite to the turbulent inflow and the transverse boundaries are taken to be periodic. The inflow values of normalised root-mean-square turbulent velocity fluctuation u'/S_L , turbulent length scale to flame thickness ratio l_T/δ_{th} , Damköhler number $Da = l_T S_L / u' \delta_{th}$, Karlovitz number $Ka = (\rho_0 S_L \delta_{th} / \mu_0)^{0.5} (u'/S_L)^{1.5} (l/\delta_{th})^{-0.5}$ and turbulent Reynolds number $Re_t = \rho_0 u' l_T / \mu_0$ for all cases are presented in Table 1 where μ_0 is the unburned gas viscosity, $\delta_{th} = (T_{ad} - T_0) / \max |\nabla T|_L$ is the thermal flame thickness and the subscript L denotes unstrained laminar flame quantities. The turbulent length scale l_T is the most energetic scale of the Passot-Pouquet spectrum, which is often used for the purpose of convenience. The length scale l_T can be analytically related to an integral length scale L_{11} resulting in $L_{11} = 1.99 \text{ mm}$ (0.57 mm) for cases A,B (case C). An alternative definition of a turbulent length scale can be given as $L = k^{3/2}/\varepsilon$ and according to Pope [41] this length scale can be converted into L_{11} using the approximation $L_{11} = 0.43L$ in the limit of high Reynolds number. An evaluation of kinetic energy and dissipation on the fresh gas side provided the estimates $L_{11} = 2.5 \text{ mm}$, 1.0 mm , 0.25 mm for cases A-C demonstrating that the integral length scale is in fact considerably smaller than l_T . The width of the computational domain is 10mm (2mm) for cases A,B (case C). Hence the integral scale L_{11} can be estimated to be at the order of 25% of the computational domain or less which is a standard value to ensure decorrelation of field variables across the periodic boundaries.

The cases investigated in this study are representative of three regimes of com-

Figure 1. Instantaneous view of c isosurfaces for (a)-(c) cases A, B, C. The value of c increases from 0.1 to 0.9 from yellow to red.

bustion [42]: case A: corrugated flamelets ($Ka < 1$), case B: thin reaction zones ($1 < Ka < 100$) and case C: broken reaction zones regime ($Ka > 100$). The domain size is $20\text{mm} \times 10\text{mm} \times 10\text{mm}$ ($8\text{mm} \times 2\text{mm} \times 2\text{mm}$) in cases A and B (case C) and the domain has been discretised by a uniform Cartesian grid of $512 \times 256 \times 256$ ($1280 \times 320 \times 320$) cells. The long side of the computational domain is taken to align with the mean direction of flame propagation. The grid spacing was determined by the flame resolution, ensuring about 10 grid points across δ_{th} . It has been checked that halving the grid size made negligible difference to the major species field for laminar flame calculations. On the unburned side of the flame brush the ratio of $\Delta x/\eta$ has been determined to be 0.51, 2.07, 1.22 for cases A-C.

High order finite-difference and Runge-Kutta schemes have been used for spatial-differentiation and explicit time-advancement, respectively. The flame was initialized using steady laminar flame solutions. Simulations have been carried out for $1.0t_e$, $6.8t_e$ and $6.7t_e$ ($t_e = l_T/u'$) for cases A-C respectively, comparable to several previous analyses [43–46]. Often, eddy turnover time is defined in terms of the integral scale L_{11} which gives larger normalised run time values (i.e. normalised in terms of L_{11}/u') by a factor of 2.5. It is also worth mentioning that the temporal evolution of flame area has been monitored and the flame is taken to be statistically stationary when the sliding average of flame area no longer varies with time.

The DNS database has been explicitly filtered using a Gaussian filter kernel with filter widths ranging from $\Delta \approx 0.4\delta_{th}$ where the flame is almost resolved, up to $\Delta \approx 2.8\delta_{th}$ where the flame becomes fully unresolved.

3. Characterization of scalar flux

Figure 1 shows instantaneous views of c -isosurfaces (in the context of this database c is defined based on temperature, i.e. $c = (T - T_0)/(T_{ad} - T_0)$ where T, T_0, T_{ad} denote the local instantaneous, the unburned gas and the adiabatic flame temperature) for cases A, B, C where the value of c increases from 0.1 to 0.9 from yellow to red. The c -contours clearly show the increasing level of flame wrinkling as the turbulence intensity increases from cases A to C. In cases A and B the c -isosurfaces lie close together, whereas in case C they are both much further apart and the distance between neighbouring isosurfaces varies greatly. This distinction is indicative of the different combustion regimes. The turbulent consumption speeds normalized by the laminar burning velocity are found to be 3.25 for cases A and C and 5.0 for case B. Although case C has a considerably higher turbulence intensity than case B (see Table 1) the turbulent flame speed in case C is smaller. It is worth noting that the length scale ratio (l_T/δ_{th} resp. L_{11}/δ_{th}) for case C is significantly different from cases A and B. Thus, the energy content of the eddies interacting with the flame in case C is much smaller than in cases A and B and that is why the flame surface area generation in case C does not happen in the proportion of the increase in u'/S_L in comparison to the values in cases A and B. Furthermore energetic turbulent eddies penetrate into the reaction zone in case C resulting in local flame thickening and modification of chemical processes. A more detailed discussion of this aspect can be found in [47,48]. From Fig. 1 it is also clearly visible that turbulent length scales increase from the unburned to the burned gas side due to two effects. Firstly, due to increasing viscosity smaller, faster motions decay more rapidly, leaving behind the larger, motions. Secondly, dilatation effects tend to produce large scale structures and a detailed discussion in this regard can be found in [50]. Finally, it is mentioned that the inflow turbulence for cases A and B is identical and that the large scale wrinkled flame surface in case A possibly might be attributed to a flame instability as discussed in Klein et al. [49]. However, it is important to note, that the effects of instability will not influence at least the qualitative nature of the underlying physics for turbulent scalar flux modelling.

A traditional method to close the unknown SGS flux of species m , i.e. $\tau_{i,m} = \bar{\rho}(\widetilde{u_i Y_m} - \tilde{u}_i \tilde{Y}_m)$, employs the gradient hypothesis:

$$\tau_{i,m}^{GHM} = -\mu_t Sc_t^{-1} (\partial \tilde{Y}_m / \partial x_i) \quad (2)$$

with the turbulent viscosity μ_t and the turbulent Schmidt number Sc_t . In the case of multispecies transport, one needs to distinguish between sub-grid fluxes of reactant mass fractions (e.g. H_2, O_2) and product mass fraction (e.g. H_2O) because they exhibit different signs. For intermediate species or radicals the gradient can take both signs. The mass fraction for N_2 is usually obtained from the constraint $\sum Y_m = 1.0$ and a transport equation is generally not solved for N_2 . Thus, the subgrid scalar flux of N_2 is not discussed in this paper.

For an arbitrary reaction progress variable c or any other suitably scaled mass fraction, it can be shown (following the steps in [51] but replacing the averaging with a filtering operation), that the probability density function (PDF) of the progress variable c is approximated by a bimodal distribution in the limit of thin flames, and under these conditions the turbulent scalar flux takes the following form: $\tau_{i,c} \approx \bar{\rho} \tilde{c} (1 - \tilde{c}) [\overline{(u_i)_P} - \overline{(u_i)_R}]$, where $\overline{(u_i)_P}$ and $\overline{(u_i)_R}$ are the conditionally filtered ve-

Figure 2. (a) Species mass fractions and temperature for the H_2 air flame taken from a laminar flame calculation. (b) Cosine of angle Θ between $\bar{\rho}(\tilde{u}_i \tilde{Y}_m - \tilde{u}_i \tilde{Y}_m)$ and $-\partial \tilde{c} / \partial x_i$ conditional on $\tilde{c}$ for different species m , calculated from case B with filter width $\Delta \approx 2.8 \delta_{th}$. Same color coding is used for the two figures.

locities in products and reactants, respectively. The density of the products is smaller than the density of the reactants and therefore, considering the mass conservation through a steady, quasi-laminar planar flame, this can lead to $\overline{(u_i)_P} > \overline{(u_i)_R}$ and hence $\tau_{i,m} > 0$ for product mass fractions, in contradiction with the gradient hypothesis closure (see eq. 2). It can further be shown that, for a reaction *regress* variable $b = 1 - c$ (which decreases monotonically from 1 in the fresh gas to 0 in the fully burned products), the flux can be expressed as $\tau_{i,b} \approx \bar{\rho} \tilde{b} (1 - \tilde{b}) [\overline{(u_i)_R} - \overline{(u_i)_P}]$ and hence, by analogy, $\tau_{i,b} < 0$ for reactant species, again in contradiction with the gradient hypothesis closure. This also indicates that fluxes of product and reactant mass fractions will have the opposite signs. This behaviour is demonstrated in Figure 2, showing the cosine of the angle Θ between $\tau_{i,m}$ and $-\partial \tilde{c} / \partial x_i$ conditionally averaged on Favre averaged progress variable $\tilde{c}$. for different species m calculated from DNS for case B. The opposite alignment of species H, O, OH, H_2O and H_2 , O_2 , HO_2 , H_2O_2 relative to the filtered progress variable gradient is clearly seen and it becomes also clear that a single progress variable might not be sufficient to represent the behaviour of the turbulent scalar flux for each individual mass fraction. Although this behaviour is not surprising, modelling implications are yet to be analyzed in the existing literature.

It should be noted, that in the context of LES, $\tilde{c}$ is a three-dimensional variable even for statistically planar turbulent premixed flames, which is in contrast to RANS where the Favre-averaged reaction progress variable is a unique function of the mean direction of flame propagation. For the purpose of conditional averaging upon $\tilde{c}$, the quantities of interest are ensemble averaged for entries in the predefined bins of $\tilde{c}$. Conditional averaging (based on $\tilde{c}$) is a standard analysis technique used for assessing combustion models in the context of LES (e.g. Boger et al. [13] and many other papers given in the references), which takes samples only from the flame brush and ensemble averages on each isosurface is given equal weightage. It is not equivalent to averaging conditional on x because $\tilde{c}$ isosurfaces are wrinkled surfaces having different x -coordinate at different points.

Figure 3 shows the cosine of the angle Θ between $\tau_{i,m}$ and $\partial \tilde{Y}_m / \partial x_i$ conditional on $\tilde{c}$ for different species m . For most of the species, negative values of $\cos(\Theta)$ are observed within the bulk of the flame thickness, indicating CGT, i.e. dominance of heat release over turbulence effects. Exceptions are H_2O_2 and HO_2 , which exist primarily on the fresh gas side (i.e. $\tilde{c} < 0.5$) where heat release effects are not pronounced. Consequently, both species show an increasing trend of gradient flux towards the fresh gas side, where the effects of heat release are relatively weak. Comparison among different cases reveals

Figure 3. Cosine of Θ between $\widetilde{\rho}(u_i \tilde{Y}_m - \tilde{u}_i \tilde{Y}_m)$ and $-\partial \tilde{Y}_m / \partial x_i$ conditional on $\tilde{c}$ for different species m , calculated from cases A, B, C, $\Delta \approx 2.8\delta_{th}$. The color code follows that in Fig. 2.

the results as expected: the effects of CGT are attenuated from cases A to C as the turbulence intensity increases. Only the results with the largest filter width are shown in Figs. 2 and 3; smaller filter widths yielded qualitatively similar behaviour, while the level of CGT decreases with decreasing the filter width. This is consistent with the observations from single step chemistry reported earlier [13,21–23].

The gradient hypothesis closure relies on an isotropic flow situation. The anisotropy tensor in the context of LES can be defined as $b_{ij} = \tau_{ij}^{SGS} / \bar{\rho} k^{sgs} - 2/3 \delta_{ij}$ where $k^{sgs} = \tau_{kk}^{sgs} / 2\bar{\rho}$ is the trace of the SGS stress tensor. The properties of the isotropy tensor can be illustrated by looking at the Lumley triangle shown in Fig. 4 for cases B,C at $\Delta = 2.8\delta_{th}$. The boundaries of the triangle can be described in terms of the invariants $II_b = -b_{kk}^2/2$ and $III_b = -b_{kk}^3/3$ and are given by the expressions

Figure 4. Lumley triangle for cases B,C for $\Delta = 2.8\delta_{th}$. Red dots represent an average conditional on $\tilde{c}$. The points for $\tilde{c} = 0$ and $\tilde{c} = 1$ are shown in green and blue respectively.

$III_b = \pm 2(|II_b|/3)^{3/2}$. Case A looks similar to case B and thus is not shown for brevity. If a coordinate system is chosen such that the anisotropy tensor is diagonal, the axisymmetric expansion is given by two equal components of the stress tensor with $b_{22} = b_{33} < 0$. Due to the trace free condition this implies that $b_{11} > 0$. Further, this indicates $\tau_{22}, \tau_{33} < 2/3\bar{\rho}k^{gsgs}$ and $\tau_{11} > 2/3\bar{\rho}k^{gsgs}$. Thus, the lower right branch in Fig. 4 represents exactly the flow state of a statistically planar flame where mean direction of flame propagation is aligned with x_1 -direction. For further details see the discussion in [52]. Figure 4 shows a strong two-dimensional anisotropy for case B due to the strong heat release effects and this behaviour is slightly more pronounced for Case A (not shown) where the invariant III_b attains values larger than 0.3 whereas III_b remains smaller than 0.3 for case B. In all cases the flow has not recovered its isotropic state on the burned gas side ($\tilde{c} = 1.0$, blue dot). In comparison, Case C is located in the broken reaction zones regime where turbulent eddies enter and considerably disturb the reaction zone. As a consequence dilatational effects are relatively weak for case C and the anisotropy is not any longer purely two-dimensional. Note that the strength of CGT in Fig. 3 is directly correlated to the anisotropy, in particular along the axisymmetric expansion shown in Fig. 4.

4. Modelling of turbulent scalar flux

A conventional closure for the turbulent scalar flux is the gradient hypothesis model $\tau_{i,m}^{GHM}$ as written in eq. 2. It has been pointed out in Refs. [21,22] that scale-similarity type models are relatively successful in closing the unknown turbulent scalar flux, and a variety of such models has been discussed by [23]. Writing the convolution filter as a Taylor series it can be shown that the well known Clark model [53] is of scale similarity type (see Ref. [23]). The Clark model for individual scalar variables can be written as:

$$\tau_{i,m}^{CGM} = \Delta^2 / 12 \bar{\rho} \partial \tilde{u}_i / \partial x_k \partial \tilde{Y}_m / \partial x_k \quad (3)$$

and will henceforth be denoted as CGM (Clark's gradient model). In addition, the Flux Richard model (FRM) suggested by [27] has been selected as an alternative model especially developed for turbulent premixed flames:

$$\tau_{i,m}^{FRM} = -\bar{\rho} C_L u'_\Delta \Delta \partial \tilde{Y}_m / \partial x_i - \rho_0 S_L M_i (\bar{Y}_m - \tilde{Y}_m) \quad ; M_i = -\partial \tilde{c} / \partial x_i / |\nabla \tilde{c}| \quad (4)$$

where $u'_\Delta = ((\widetilde{u_i u_i} - \tilde{u}_i \tilde{u}_i)/3)^{1/2}$ is the subgrid scale velocity fluctuation. The FRM consists of a gradient type contribution where the model parameter is given by $C_L = 0.12$, and a CGT contribution represented by the last term of eq. 4. Variants of this model type have been analysed in detail [21,22] and only one of the more successful representatives will be discussed here. The last term in eq. 4 is typically derived by considering the mass conservation in a steady laminar flame $\rho u = \rho_0 S_L$ which gives $\bar{\rho} \tilde{u} \tilde{c} - \bar{\rho} \tilde{u} \tilde{c} \approx \rho_0 S_L (\bar{c} - \tilde{c})$. This expression is then generalized to the three dimensional case by writing $\bar{\rho} \tilde{u}_i \tilde{c} - \bar{\rho} \tilde{u}_i \tilde{c} \approx -\rho_0 S_L M_i (\bar{c} - \tilde{c})$ where the flame normal vector is defined by $M_i = -\partial \tilde{c} / \partial x_i / |\nabla \tilde{c}|$. It seems that eq. 4 is the most appropriate generalization for multi-species transport. Note, that the flame normal vector is still defined in terms of $c = c_T$ rather than Y_m because using the species mass fraction based flame normal vector would result in a wrong sign of the flux for reactant species. In the following, the Pearson correlation coefficient is used to measure the degree of linear dependence

Figure 5. Correlation coefficients between modelled and actual values of $\bar{\rho}(\tilde{u}_i \tilde{Y}_m - \tilde{u}_i \tilde{Y}_m)$ in the range $0.1 \leq \bar{c} \leq 0.9$ for models $\tau_{i,m}^{CGM}$ (■), $\tau_{i,m}^{GHM}$ (■), $\tau_{i,m}^{FRM}$ (■) and $\tau_{i,m}^{CPR}$ (■) for cases A,B,C (top to bottom) for filter width $\Delta \approx 0.4\delta_{th}$ (left column) and $\Delta \approx 2.8\delta_{th}$ (right column). Correlation coefficients are averaged over all three components. The negative of the correlation coefficient is shown for the GHM model.

between the scalar flux evaluated from DNS and the different model expressions given in eqs. 2, 3, 4. In addition, the CPR model (i.e. Clark plus Richard) denotes the sum of the CGM model and the FRM model and will be discussed later. The regions corresponding to $\bar{c} < 0.1$ and $\bar{c} > 0.9$ have been ignored since the correlation coefficients have little physical significance in these regions due to the small magnitudes of the turbulent scalar fluxes in the fully burned and unburned regions.

The left and right columns of Figure 5 show the correlation coefficients (averaged over all three components) for all cases for filter width $\Delta \approx 0.4\delta_{th}$ and $2.8\delta_{th}$, respectively. Note that the negative of the correlation coefficient is shown for the GHM model in all figures. In particular, for Cases A and B, the GHM model has a very strong negative correlation with the fluxes from DNS which is due to the strong anisotropy, caused by dilatational effects, depicted in Fig. 4. Going from case A to C, however, the negative correlation becomes weaker when turbulent velocity fluctuations become stronger relative to heat release. Similar qualitative behaviour has been reported earlier for single step chemistry based simulations. However, the planar flames analysed by [21–23] have been started from an initially isotropic turbulent velocity field which subsequently decayed. Consequently, the anisotropy observed in these flames is smaller compared to the inflow-outflow configuration considered herein, which explains the stronger negative correlation observed in the present study. In accordance with earlier findings, the CGM model gives relatively high positive correlations for all cases, but the correlation strength decreases with increasing filter size. This can be explained based on two observations: (a) the CGM model is finite-difference gradient based and the numerical errors become larger for larger filter size, and (b) the CGM model is based on a Taylor series expansion of the convolution filter, which again is a more accurate assumption for smaller filter size. The reduction of the correlation strength for $\Delta = 2.8\delta_{th}$ is most clearly seen for the intermediate species HO_2 and H_2O_2 .

The FRM model benefits from the large amount of CGT in cases A and B for the flow configuration considered in this work. Consequently, it shows reasonably high

positive correlations for all reactive species. Case C has the highest turbulence intensity and consequently the GHM model now exhibits either a small positive correlation for $\Delta \approx 0.4\delta_{th}$ or a weak negative correlation for $\Delta \approx 2.8\delta_{th}$ for some species. The CGM model yields reasonable positive correlations for both filter widths, whereas the FRM model performance deteriorates in the broken reaction zones regime and fails for small filter widths.

It is also of interest to analyse the magnitude of the modelled scalar flux term relative to the corresponding DNS value. Exemplarily Fig. 6 shows the variation of the scalar flux of H_2 in direction of mean flame propagation for cases B and C (case A behaves similar to case B) and $\Delta/\delta_{th} = 0.4, 2.8$ together with the different model expressions averaged conditional on $\tilde{c}$. The behaviour of a product species would be similar but with an opposite sign. Intermediate species show a somewhat more complex behaviour but are not discussed further for the sake of brevity. The characteristic features of the model behaviours can be summarised from Fig. 6: The GHM model fails to predict the correct sign of the flux for cases A,B but it works slightly better for small filter size and high turbulence intensity (e.g. case C). The CGM model predicts qualitatively the correct behaviour of the flux (except for Case C, $\Delta/\delta_{th} = 2.8$) but underpredicts the flux magnitude especially for large filter width. The FRM model performs reasonably in direction of mean flame propagation from a qualitative perspective but mostly under-predicts the flux magnitude. Including the wrinkling factor $\Xi = |\nabla \tilde{c}|/|\nabla \bar{c}|$ in the CGT part of the FRM model (i.e. $-\bar{\rho}C_L u'_\Delta \Delta \partial \tilde{Y}_m / \partial x_i - \rho_0 S_L \Xi M_i (\bar{Y}_m - \tilde{Y}_m)$ see [28]) increases the negative flux magnitude to a small, but not sufficient, extent for cases A and B but will not be beneficial for case C. The remarkable performance of the FRM model for case C, $\Delta/\delta_{th} = 2.8$ might be fortuitous. This is particularly surprising because Fig.5 depicted very low correlations for the same parameters. A detailed investigation reveals that in general correlations are mostly uniformly distributed over all three flux components, except for the FRM model in case C, $\Delta/\delta_{th} = 2.8$, where the correlation coefficient in direction of mean flame propagation is very high, consistent with the good prediction of the conditional average depicted in Fig. 6, but the correlation coefficients in the direction normal to mean flame propagation are nearly zero.

Based on this observation a new model, i.e. the CPR model, has been tested consisting of the sum of the FRM and CGM models with the idea that the CGM model part will result in good correlations for all flux components and that the FRM contribution will properly predict the conditional mean. Interestingly this combined model did show the best overall performance of all models considered so far, with the exception of very small filter sizes where the pure CGM model performs more or less equally well (see Figs. 5,6). However, it is likely that such small filter sizes are not relevant in the context of LES modelling. Nevertheless, the model is still far to be fully satisfactory.

5. Bray-Moss-Libby theory for multispecies transport

One problem in the FRM model (eq. 4), which has not been discussed so far, is the closure of $\tilde{Y}_m$ which is unknown because a transport equation for its Favre averaged counterpart $\tilde{Y}_m$ is solved. This problem is sometimes ignored or alternatively solved using BML theory (see e.g. Ref. [51]), where filtered reaction progress variable $\tilde{c}$ and

Figure 6. Variation of turbulent scalar flux component in direction of mean flame propagation $\bar{\rho}(\widetilde{u_n Y_{H_2}} - \widetilde{\rho u_n Y_{H_2}})$ conditional on $\tilde{c}$ for $\Delta/\delta_{th} = 0.4$ (left column) and $\Delta/\delta_{th} = 2.8$ (right column) for cases B,C.

$\bar{\rho}$ can be expressed in terms of $\tilde{c}$ as

$$\bar{c} = (1 + \tau)\tilde{c}/(1 + \tau\tilde{c}) \quad \bar{\rho} = \rho_0/(1 + \tau\tilde{c}) \quad (5)$$

where $\tau = (T_{ad} - T_0)/T_0$ is the heat release factor. Inserting eq. 5 in the FRM model gives the closed, one dimensional expression

$$\rho_0 S_L (\bar{c} - \tilde{c}) = \bar{\rho} \tau S_L \tilde{c} (1 - \tilde{c}). \quad (6)$$

A (again one-dimensional) formula in direct analogy to eq. 6 would be

$$\rho_0 S_L (\bar{Y}_m - \widetilde{Y}_m) = \bar{\rho} \tau S_L (\widetilde{Y}_m - Y_{m,R})(Y_{m,P} - \widetilde{Y}_m)/(Y_{m,P} - Y_{m,R}), \quad (7)$$

where $Y_{m,R}$ and $Y_{m,P}$ denote mass fractions of species Y_m on the reactant and product side. Although $Y_{m,P}$ could be different in turbulent flames where differential diffusion plays an important role, we refer to the values taken from a laminar flame calculation for the sake of simplicity. However, eq. 7 gives, except for H_2, O_2, H_2O unsatisfactory results and is not recommended. A better approach is to use a mass fraction based reaction progress variable (i.e. $c = (Y_{m,R} - Y_m)/(Y_{m,R} - Y_{m,P})$) which in conjunction with eq. 5 yields

$$\bar{Y}_{m,BML*} = (\widetilde{Y}_m + \tau\tilde{c}Y_{m,P})/(1 + \tau\tilde{c}) \quad (8)$$

This formula makes implicit use of the filtered density expression (eq. 5) which can be problematic because not every reaction progress variable c should be used to express $\bar{\rho}$. The recommended expression therefore is given by

$$\bar{Y}_{m,BML} = [\bar{\rho}\widetilde{Y}_m(Y_{m,R} - Y_{m,P}) + Y_{m,R} \cdot Y_{m,P}(\rho_R - \rho_P)]/[\rho_R Y_{m,R} - \rho_P Y_{m,P}] \quad (9)$$

This equation is derived solely on the assumption of a bimodal PDF for the distribution of mass fraction Y_m . For intermediate species like H_2O_2 or HO_2 which do not exist in the fresh gas or the fully burned products (refer to Fig. 2) this assumption is entirely wrong. The PDF of these species is better described by a single Dirac function which has its peak at the location where Y_m takes its maximum value, denoted $Y_{m,max}$. This gives rise to the following expression (denoted *SDP* for single Dirac peak):

$$\bar{Y}_{m,SDP} = \bar{\rho}\widetilde{Y}_m/\rho_{max} \quad (10)$$

Chakraborty and Cant [54] have pointed out that the BML formalism is not correct for small filter sizes. This is because the sub-grid reaction progress variable pdf is not necessarily bimodal in nature. In addition to this $\bar{c}$ in eq. 5 does not revert to $\tilde{c}$ in the limit of zero filter size. Therefore the following modification is strongly recommended in order to ensure the correct asymptotic behaviour for small filter size.

$$\begin{aligned} \bar{Y}_{m,BML,corr} &= \bar{Y}_{m,BML} (1 - \exp(-\Lambda\Delta/\delta_{th})) + \widetilde{Y}_m \exp(-\Lambda\Delta/\delta_{th}), \\ \bar{Y}_{m,SDP,corr} &= \bar{Y}_{m,SDP} (1 - \exp(-\Lambda\Delta/\delta_{th})) + \widetilde{Y}_m \exp(-\Lambda\Delta/\delta_{th}), \quad \Lambda = 3.0. \end{aligned} \quad (11)$$

The behaviour of Eqs. 8–11 is best illustrated for filtered laminar flame data, i.e. the filtered species profiles used for initialisation of the turbulent flame configuration under investigation (see Fig. 7 (a-d)). It can be seen that for major species like H_2O the difference between the predictions of eqs. 8 and 9 is rather small. Furthermore, it becomes clear that BML formulas are not appropriate for small filter size ($\Delta/\delta_{th} = 1$) and that the modification given by eq. 11 is required even for moderate to large filter size. For OH eq. 9 provides a better model for the SGS scalar flux compared to eq. 8, which is due to the non-monotonic behaviour of Y_{OH} which shows a maximum in the middle of the flame before approaching a smaller non zero value towards the burned gas side (see Fig. 2). This makes it not very appropriate for expressing $\bar{\rho}$ in terms of $c_{Y_{OH}}$. Finally, Fig. 7d shows that the assumption of a bimodal distribution of the species PDF is not appropriate for modelling the scalar flux of intermediate species (e.g. H_2O_2 where mass fraction is zero on the fresh gas side and in fully burned products), whereas the assumption of a single Dirac peak, resulting in eq. 10, provides a reasonable description of the unknown SGS scalar flux. The turbulent case C at $\Delta/\delta_{th} = 2.8$ is exemplarily taken for demonstrating the model prediction for two species which are difficult to model. The behaviour for other cases and filter width is qualitatively similar and not shown for the sake of brevity. Again eq. 9 is a reasonable model for $\bar{Y}_{OH}$, (see Fig. 7f), but does not work suitably for $\bar{Y}_{H_2O_2}$ where eq. 10 is more suitable (see Fig. 7e). This can be understood by comparing the model assumptions of eqs. 8–10 with the characteristics of the mass fraction profiles depicted in Fig. 2.

Figure 7. Variation of filtered scalar flux calculated from laminar flame data together with different model expressions. (a) $m = H_2O, \Delta/\delta_{th} = 1$, (b) $m = H_2O, \Delta/\delta_{th} = 6$, (c) $m = OH, \Delta/\delta_{th} = 6$, (d) $m = H_2O_2, \Delta/\delta_{th} = 6$. Variation of flux terms for turbulent Case C for $\Delta/\delta_{th} = 2.8$ (e) $m = H_2O_2$ (f) $m = OH$. Subscripts BML BML^* $BML, Corr$ $SDP, Corr$ refer to eqs. 9, 8 as well as 9 and 10 each combined with 11. $SDP, Corr$ is only applied to intermediate species and therefore not shown in subfigure (f).

6. Conclusions

The turbulent SGS scalar fluxes of individual species have been *a-priori* analysed using a detailed chemistry based DNS database of three planar turbulent premixed H_2 -air flames, representing three different regimes of combustion. The flames in the corrugated flamelets (case A) and thin reaction zones (case B) regime show a pronounced two-dimensional anisotropy due to strong dilatation effects. The effect of dilatation rate diminishes with increasing Karlovitz number (i.e. from case A to C) and this is clearly reflected in the Lumley triangle for the different flames. All species show predominantly counter-gradient type of transport for cases A and B and to some extent even for case C with exception of the intermediate species H_2O_2 and HO_2 , which exhibit a reduced degree of CGT towards the unburned gas side. The statistical behaviour of the scalar flux depends on the competition between heat release and turbulence as a consequence of the decreasing extent of CGT from case A to C.

Furthermore, the sign of the flux depends on the question whether a product or reactant species is considered. The performances of three SGS scalar flux models have been analysed based on this database. The GHM model shows a strong negative correlation for all major species in cases A and B and for all filter widths. The correlation increases for case C but overall the correlation strength remains very small. The CGM model correlates very well for small filter width but its performance deteriorates to some extent for large filter widths. Finally the FRM, correlates well for all major species for Cases A and B, if it is suitably adopted to multi-species transport. However, this model fails if the extent of CGT remains small. This holds in particular for the scalar flux components in direction normal to mean flame propagation. It was found that a combination of the FRM and CGM model provides the best modelling compromise for the range of filter sizes and Karlovitz numbers considered here.

In all cases modelling of SGS scalar fluxes of non-major species is much more challenging and modifications to BML theory have been suggested. Finally, all these results qualitatively confirm earlier findings based on simple chemistry based analyses.

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