Statistical Behaviours of Turbulent Scalar Fluxes in High Pressure Turbulent Premixed Combustion in the Context of Large Eddy Simulations

Christian Kasten¹, Felix B. Keil¹, Nilanjan Chakraborty², Markus Klein¹*

¹Bundeswehr University Munich,
Department of Aerospace Engineering
Werner-Heisenberg-Weg 39, 85577 Neubiberg, Germany
Email: markus.klein@unibw.de

²School of Engineering
University of Newcastle
Claremont Road, Newcastle
NE1 7RU, UK
Email: nilanjan.chakraborty@ncl.ac.uk

* Corresponding author
ABSTRACT

The effects of pressure on the physical behaviour of sub-grid scalar fluxes and their modelling have been analysed based on a-priori analysis of a Direct Numerical Simulation database of turbulent premixed Bunsen flames featuring different pressure levels. Due to an increasing ratio of the hydrodynamic to critical length scale for the occurrence of Darrieus-Landau instability, the flames become increasingly hydrodynamically unstable with increasing pressure. An increasing extent of counter-gradient transport with increasing pressure has been found, consistent with recent observations reported in the existing literature. The performance of a selection of well-known Large Eddy Simulation models for sub-grid turbulent scalar flux from literature has been assessed for a range of filter widths for different thermodynamic pressures. While some models show high correlations with the sub-grid scalar flux extracted from explicitly filtered Direct Numerical Simulation data, and a reasonable quantitative prediction of the sub-grid turbulent scalar flux magnitude for small values of filter width to flame thickness ratio, several models considerably underpredict the sub-grid scalar flux magnitudes for large filter widths. This effect becomes more pronounced for higher pressures as the length scale separation between the filter width and flame thickness increases with increasing pressure.

Keywords: Turbulent Scalar Fluxes, Direct Numerical Simulations, Large Eddy Simulation, High Pressure Bunsen Flame
1. INTRODUCTION

The optimization of combustion processes and the improvement of their efficiency are of pivotal importance for the minimization of the negative environmental side-effects of combustion, particularly greenhouse gas emissions. For these reasons, combustion systems are typically run at elevated pressures: aircraft gas turbine engines typically operate under an approximately constant pressure of 30 bar and gas turbine combustion pressures have been increasing at a near linear rate with time for long and are expected to increase further (Foster and Miller, 2010), while automotive engines exhibit pressures as large as 60 bar. Nevertheless, by far the largest part of the existing literature on numerical and theoretical analyses of turbulent premixed combustion has been conducted for ambient pressure conditions. For hydrocarbon fuels, laminar burning velocities and flame thicknesses decrease with increasing pressure (Fragner et al., 2015) and as a result of this, elevated pressure affects Reynolds, Damköhler and Karlovitz numbers, and chemical reaction rates (Bougrine et al. 2011). In particular, the reduction of the flame thickness promotes the hydrodynamic instability and in turn increases the turbulent flame speed (Kobayashi et al., 2002; Creta et al., 2016; Klein et al., 2018a,b,c; Chakraborty et al. 2019).

Although several experimental (Kobayashi et al.; 2002, Kobayashi, 2002; Stopper et al. 2013; Fragner et al. 2015), theoretical (Pelce and Clavin, 1982, Matalon and Matkowsky, 1982, Creta and Matalon, 2011) and numerical (Schmitt et al., 2007; Bougrine et al. 2015) studies analysed the effects of elevated pressure on combustion dynamics and the related phenomenon of Darrieus-Landau (DL) instability, the possible modelling implications in the context of Large Eddy Simulation (LES) have rarely been addressed (e.g. Dinkelacker et al., 2011; Keppeler et al., 2014, Keppeler and Pfitzner, 2014; Lamioni et al., 2019). Among different LES modelling options for describing turbulence-chemistry interaction (Janicka and Sadiki, 2005; Pitsch,
2006), recent work by the present authors focused on algebraic (Rasool et al., 2020) and transport equation based (Keil et al., 2019a) Flame Surface Density (FSD) modelling for high pressure turbulent premixed Bunsen flames in the context of LES, whereas the modelling of the sub-grid scalar variance transport equation of reaction progress variable was discussed by Keil et al. (2020). Generally speaking, the main findings are that for a given filter width to flame thickness ratio, the physical behaviour and the modelling requirements are not significantly affected by pressure. However, as the flame thickness decreases with increasing pressure, the sub-grid model contribution and the modelling uncertainty increase considerably for flames under elevated pressures. Recent work by Lamioni et al. (2018) reported an enhancement of counter-gradient transport (CGT) for sub-grid turbulent scalar fluxes (TSF) for hydrodynamically unstable flames. This has wide-ranging modelling implications (Lamioni et al., 2018, 2019), as the CGT will not only be observed for the TSF but it occurs also for sub-grid transports of other quantities, such as SGS momentum (Klein et al. 2015), scalar variance of reaction progress variable (Keil et al., 2020) and FSD (Hawkes and Cant, 2001, Chakraborty and Cant, 2009a). The LES based modelling of sub-grid TSFs under atmospheric pressure conditions has been extensively studied in the past (see Gao et al. 2015, Klein et al. 2016, Klein et al. 2018d and references therein) but the modelling of these effects in the context of high pressure turbulent premixed combustion has not been analysed so far, to the best knowledge of the authors. The present work addresses this gap in the existing literature and hence, the main objectives of the present analysis are:

(1) to analyse the statistical behaviour of the TSF of reaction progress variable at different pressure levels.

(2) to assess the performance of selected, well-established closures of TSFs at elevated pressures.
The governing equations, the numerical methodology and the database are described in Section 2. The results will be presented and discussed in detail in the subsequent section. The main findings are summarised, and conclusions are drawn in the final section of this paper.

2. GOVERNING EQUATIONS AND MODEL EXPRESSIONS

For low Mach number, unity Lewis number, adiabatic combustion, the complexity of transport and chemistry can be simplified with the help of a reaction progress variable $c$, which assumes a value equal to zero in the unburned reactants and unity in the fully burned products. The Favre-filtered transport equation for the progress variable can be written as:

$$\frac{\partial (\bar{\rho} \bar{c})}{\partial t} + \frac{\partial (\bar{\rho} \bar{u}_j \bar{c})}{\partial x_j} = -\frac{\partial \bar{\rho} (\bar{u}_j \bar{c} - \bar{u}_j \bar{c})}{\partial x_j} + \frac{\dot{w}}{\tau_2} + \frac{\partial \left[ \rho D \left( \frac{\partial c}{\partial x_j} \right) \right]}{\partial x_j}$$

(1)

where $\rho$ is the gas density, $D$ is the mass diffusivity, $u_j$ is the $j^{th}$ component of velocity, $\dot{w}$ is the reaction rate of reaction progress variable $c$ and the tilde $\bar{q} = \rho \bar{q}/\bar{\rho}$ denotes the Favre filtering of a general variable $q$. The terms on the right-hand side of Eq. 1 are unclosed. The LES modelling of $(T_2 + T_3)$ for elevated pressure flames has been discussed in Rasool et al. (2020) and Keil et al. (2019a), and the present work focuses on the modelling of the turbulent transport term $T_1$, which depends on the closure of the sub-grid TSF. The sub-grid TSF is defined as:

$$\tau_j = \bar{\rho} [\bar{u}_j \bar{c} - \bar{u}_j \bar{c}]$$

(2)

The gradient hypothesis model (GHM) is the most often used model for closure of sub-grid TSF, which is given by:

$$\tau_i^{GHM} = -\frac{\mu_t}{S c_i} \frac{\partial \bar{c}}{\partial x_i}$$

(3)
\[
\mu_t = \bar{\rho}(C_s \Delta)^2 \sqrt{2 \tilde{S}_{ij} \tilde{S}_{ij}}, \quad \tilde{S}_{ij} = \frac{1}{2} \left( \frac{\partial \tilde{u}_i}{\partial x_j} + \frac{\partial \tilde{u}_j}{\partial x_i} \right), \quad C_s = 0.18, \quad Sc_t = 1.0
\]

where \( \mu_t \) is the eddy viscosity, \( C_s \) is the Smagorinsky constant, \( \Delta \) is the filter width, \( Sc_t \) is the turbulent Schmidt number and \( \tilde{S}_{ij} \) is the resolved strain rate. It is well-known that Eq. 3 fails in many scenarios. Particularly, in the context of turbulent premixed combustion, it is well established (Veynante et al. 1997) that the TSF can exhibit CGT behaviour if the effects of heat release overcome the strength of turbulent velocity fluctuations. Since then a large number of models has been suggested and analysed in the past (see Gao et al. 2015, Klein et al. 2016, Klein et al. 2018d and references therein) and only a representative subset of these will be discussed here.

Richard et al. (2007) proposed a model (RFM=Richard flux model) in the following manner:

\[
\tau_{i}^{RFM} = -\bar{\rho}C_L u'_i \Delta \frac{\partial \bar{c}}{\partial x_i} - \rho_0 S_L M_i (\bar{c} - \tilde{c}),
\]

\[
\tilde{M} = -\frac{\nabla \tilde{c}}{|\nabla \bar{c}|}, \quad u'_\Delta = \sqrt{\left( \bar{u}_i \bar{u}_j - \bar{u}_i \tilde{u}_j \right)/3}, \quad C_L = 0.12 \quad (4)
\]

The first term \( \bar{\rho}C_L u'_i \Delta \partial \bar{c}/\partial x_i \) on the right hand side of eq. (4) is responsible for gradient transport, whereas the second term \( -\rho_0 S_L M_i (\bar{c} - \tilde{c}) \) accounts for CGT. Boger (2000) and Weller et al. (1998) discussed about the possibility of an alternative model by replacing \( \rho_0 S_L M_i (\bar{c} - \tilde{c}) \) with \( \rho_0 S_L \Xi M_i (\bar{c} - \tilde{c}) \) where \( \Xi = |\nabla \bar{c}|/|\nabla \tilde{c}| \) is the wrinkling factor. Because of the moderate values of the wrinkling factor for the cases considered here, the model proposed by Weller et al. (1998) behaves similarly to the RFM model (in fact slightly worse) in terms of the magnitude of its prediction and correlation with sub-grid TSF extracted from DNS data, and hence is not discussed further. Clark et al. (1979) proposed a model expression for SGS momentum transport which in the context of sub-grid scalar flux closure reads:
\[ \tau_{i}^{\text{CGM}} = \frac{\rho \Delta^2}{12} \frac{\partial \bar{u}_i}{\partial x_k} \frac{\partial \bar{c}}{\partial x_k} \] (5)

It will henceforth be referred to as the CGM (Clarks gradient model) model in this paper. Huai et al. (2006) suggested an alternative model in the context of passive scalar transport denoted in the following Huai Flux Model (HFM):

\[ \tau_{i}^{\text{HFM}} = -\bar{\rho}(C_s \Delta)^2 \sqrt{2 \bar{S}_{ij} \bar{S}_{ij} \frac{\partial \bar{c}}{\partial x_i}} + \bar{\rho}D_{an} \Delta^2 \bar{S}_{ik} \frac{\partial \bar{c}}{\partial x_k} ; \quad D_{an} = 0.14 \] (6)

In Eq. 6, the first (second) term on right hand side represents the contribution of gradient (counter-gradient) type of transport. The CPR model (Clark plus Richard), given in Eq. 7, is based on the observation (Klein et al. 2018d) that the CGM model provides good correlations but deteriorates for large filter size, but the RFM model tends to be more successful in predicting the magnitude of the TSF even for large filter sizes while the correlation strength remains lower in particular for turbulent flames. The CPR model should not be considered as a new model but a modelling strategy. For this reason, no model constants have been introduced.

\[ \tau_{i}^{\text{CPR}} = \tau_{i}^{\text{CGM}} + \tau_{i}^{\text{RFM}} \] (7)

3. NUMERICAL METHODOLOGY AND DATABASE

The DNS code Senga (Jenkins and Cant, 1999) has been used to generate the present database. In Senga, the compressible form of the conservation equations of mass, momentum, energy and reaction progress variables is solved numerically, by using a high order finite difference (i.e. 10th order central difference for internal grid points with gradually reducing to a 2nd order one-sided scheme at non periodic boundaries) method. The time-integration is performed using a 3rd order low-storage Runge-Kutta scheme. A generic single-step Arrhenius type irreversible chemical mechanism has been considered for this work.
because of the extreme demands of flame resolution at elevated pressures. This assumption is not expected to affect the conclusions of this paper, as this work focuses only on the fluid-dynamical aspects of the pressure dependence. In the context of a simple chemical mechanism, the pre-exponential factor and kinematic viscosity have been altered to achieve the well-known behaviour of methane-air flames, where the unstrained laminar burning velocity $S_L$ scales with pressure $P$ as: $S_L \sim P^{-0.5}$ (Turns, 2011), dynamic viscosity $\mu$ does not change with pressure but gas density $\rho$ increases with pressure as $\rho \propto P$. This implies that the thermal flame thickness $\delta_{th} = (T_{ad} - T_0)/\max|\nabla T|_L$ (where $T, T_0$ and $T_{ad}$ are the instantaneous dimensional, unburned gas and adiabatic flame temperatures respectively) scales as: $\delta_{th} \sim \mu/(\rho Sc S_L) \sim P^{-0.5}$ where $Sc$ is the Schmidt number.

Three different Bunsen flames, cases A-C, at three different normalised pressure $P/P_0$ levels are considered for this analysis, where the reference pressure $P_0$ is taken to be 1.0 bar. In this work, a small value of normalised turbulent root-mean-square (rms) velocity fluctuation $u'/S_L$ has been used in order to be able to analyse the effects of DL instability at elevated pressure. The simulation domain is for all cases taken to be $2d_n \times 2d_n \times 2d_n$, with the nozzle diameter $d_n$, which corresponds to a cube of $50 \delta_{th} \times 50 \delta_{th} \times 50 \delta_{th}$ [112 $\delta_{th} \times 112 \delta_{th} \times 112 \delta_{th}$] (159 $\delta_{th} \times 159 \delta_{th} \times 159 \delta_{th}$) for case A [case B] (case C), which is discretised using a uniform Cartesian grid of $250\times250\times250$ [560$\times$560$\times$560] (795$\times$795$\times$795) points. This ensures resolution of both the Kolmogorov length scale and the flame thickness. All boundaries of the computational domain except the inflow are taken to be partially non-reflecting (NSCBC) outflows (Poinsot and Lele, 1992). Inflow data has been generated using a modified version of the method suggested by Klein et al. (2003), tailored for a compressible flow solver with a small time step (see Klein et al. 2018b for details).
Table 1. Inlet flow parameters for the considered cases

<table>
<thead>
<tr>
<th>Case</th>
<th>(P/P_0)</th>
<th>(Re_{d_n})</th>
<th>(Re_t)</th>
<th>(U_B/S_L)</th>
<th>(u'/S_L)</th>
<th>(l/d_n)</th>
<th>(l/\delta_{th})</th>
<th>(Ka)</th>
<th>(Da)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>1.0</td>
<td>399</td>
<td>13.30</td>
<td>6.0</td>
<td>1.0</td>
<td>1/5</td>
<td>5.20</td>
<td>0.45</td>
<td>5.00</td>
</tr>
<tr>
<td>B</td>
<td>5.0</td>
<td>892</td>
<td>29.26</td>
<td>6.0</td>
<td>1.0</td>
<td>1/5</td>
<td>11.40</td>
<td>0.30</td>
<td>11.40</td>
</tr>
<tr>
<td>C</td>
<td>10.0</td>
<td>1262</td>
<td>41.22</td>
<td>6.0</td>
<td>1.0</td>
<td>1/5</td>
<td>16.13</td>
<td>0.25</td>
<td>16.13</td>
</tr>
</tbody>
</table>

All inlet flow parameters are provided in Table 1 and are defined as follows. The Reynolds number \(Re_{d_n} = U_B d_n / \nu_u\) is based on the bulk inlet velocity \(U_B\), nozzle diameter \(d_n\), and the kinematic viscosity in the unburned gas \(\nu_u\), whereas the turbulent Reynolds number is \(Re_t = u' l / \nu_u\). The normalised inlet velocity is \(U_B / S_L\). The longitudinal integral length scale to nozzle diameter ratio \(l/d_n\) and longitudinal integral length scale to thermal flame thickness ratio \(l / \delta_{th}\) are provided as well in Table 1. Finally, the Karlovitz and Damköhler numbers are given by \(Ka = (u'/S_L)^{3/2}(l/\delta_{th})^{-1/2}\) and \(Da = l S_L / \delta_{th} u'\). The heat release parameter \(\tau = (T_{ad} - T_0) / T_0\) and the Zel’dovich number \(\beta = T_{ac}(T_{ad} - T_0) / T_{ad}^2\) are taken to be 4.5 and 6.0 respectively where \(T_{ac}\) is the activation temperature. Standard values of Prandtl number \((Pr = 0.7)\) and ratio of specific heats \((\gamma_g = 1.4)\) have been used. The Lewis number of all the species are taken to be unity (i.e. \(Le = Sc / Pr = 1.0\)) for the current analysis. The cases considered here are shown on the regime diagram by Peters (2000) in Fig. 1.

It can be seen from Table 1 and Fig. 1, that cases A,B,C have the same inlet values of \(u'/S_L\), \(U_B/S_L\) and \(l/d_n\) but \(Re_t\), \(Da\) and \(Ka\) values are different, and thus they fall on different locations on the combustion regime diagram. All flames are located on the boundary of the wrinkled and the corrugated flamelets regimes according to the regime diagram by Peters (2000).
The reacting flow field has been initialised by an unstrained premixed laminar flame solution specified as a function of radius from the nozzle centre. As in jet like flows the mean velocity profile after the nozzle exit has been approximated by a hyperbolic-tangent like distribution. All statistics have been extracted after 2 through-pass times (i.e. $L/U_B$ where $L = 2d_n$ is the length of the simulation domain). For a-priori analysis, the DNS data has been explicitly filtered using a Gaussian filter kernel for a wide range of filter width from $\Delta/\delta_{th} = 0.8$ where the flame is still partially resolved up to $\Delta/\delta_{th} = 12$ where the flame is fully unresolved.

![Figure 1: The inlet conditions of the Bunsen burner flames on the combustion regime diagram.](image)

### 3. RESULTS & DISCUSSION

Figure 2 (a) shows instantaneous views of isosurfaces of reaction progress variable $c$ for cases A-C. All the Bunsen flames considered here have the same value of $u'/S_L$ and $l/d_n$ but as pressure increases the flames become increasingly wrinkled because the ratio of hydrodynamic length scale to critical wavelength (denoted henceforth $\Lambda_{crit}$) for DL instability increases (see Klein et al. 2018a,b,c for a detailed discussion of this effect for the present database).
signature of DL instability in case C is visible from the sharply negative curved cusps with positively curved large wavelength bulges in between. For the same reason the curvature PDFs, shown for reaction progress variable $c = 0.8$ which is close to the location of maximum reaction rate, become increasingly negatively skewed with increasing pressure (see Fig. 2 (b)).

(a) ![](image) (b) ![](image)

**Figure 2:** a) Instantaneous views of isosurfaces of reaction progress variable $c$. Colour changes from dark to light red from the unburned gas to the burned gas. b) Curvature PDFs for the considered cases for reaction progress variable $c = 0.8$. The curvature $\kappa_m$ is normalized with the thermal flame thickness $\delta_{th}$ of the corresponding flame.

The statistical behaviour of the unclosed terms $T_1 - T_3$ of the transport equation for $\tilde{c}$ will be discussed in the next section and this will be followed by a detailed characterisation of the TSF and its LES modelling.

**Statistical behaviour of the transport of Favre filtered reaction progress variable**

The variations of the mean values of $T_i \times (\delta_{th}/\rho_0 S_L)$ with $i = 1,2,3$, conditional upon $\tilde{c}$ for cases A-C are shown in Fig. 3 for $\Delta/\delta_{th} = 0.8, 2.4, 4.0$. All conditional plots in this work are shown in the range $0.01 \leq \tilde{c} \leq 0.99$. It can be seen from Fig. 3 that the mean values of the chemical reaction rate $T_2$ remain positive for all cases and all filter widths. By contrast the mean values of the filtered molecular diffusion term $T_3$ and the turbulent diffusion term (i.e. divergence of the TSF) $T_1$ assume positive values towards the fresh gas side and negative values towards the burned gas side. The mean value of the turbulent diffusion term $T_1$ is
considerably smaller than that of $T_3$ for small filter widths (e.g. $\Delta/\delta_{th} = 0.8$), but the mean values of these terms are of similar magnitude for $\Delta/\delta_{th} = 2.4$. The mean behaviour of $T_i \times (\delta_{th}/\rho_0 S_L)$ with $i = 1,2,3$ remains both qualitatively and quantitatively similar for a given filter width to flame thickness ratio $\Delta/\delta_{th}$. However, as the pressure increases the flame thickness decreases. For example, the flame thickness in case C is about one third (i.e. $10^{-0.5}$) of the flame thickness in case A. As a consequence, the computational time increases by a factor of $10^{0.5-4} = 100$ assuming that the time step is proportional to the grid spacing. For engineering applications, it might be more likely that the LES grid spacing (and therefore LES filter width) will be dictated by the geometry and the energy carrying flow structures that can be adequately represented, but not by the flame thickness.

Figure 3: Profiles of the mean values of the terms of the transport equation of $\tilde{c}$ conditional upon $\tilde{c}$ for cases A, B and C for filter widths $\Delta/\delta_{th} = 0.8, 2.4, 4.0$. 
Hence, it might be more likely that the resolution will be chosen more or less independent of pressure and for this reason Fig. 4 shows the mean values of $T_i \times (\delta_{th}/\rho_0S_L)$ with $i = 1,2,3$ conditional upon $\tilde{c}$ but this time the filter width $\Delta$ has been normalised with the nozzle diameter $d_n$, such that $\Delta/d_n = 0.15$ for case A (case C) corresponds to $\Delta/\delta_{th} \approx 4.0$ ($\Delta/\delta_{th} \approx 12$). Figure 4 shows that the qualitative and quantitative behaviours of the mean values of $T_1 - T_3$ now change with increasing pressure. It is worth noting that the mean value of $T_1$ is now considerably larger than that of $T_3$ for the largest filter width in case C.

![Figure 4](image-url)

Figure 4: Budgets of the transport equation of $\tilde{c}$ for cases A, B and C for filter widths $\Delta/d_n = 0.03, 0.09, 0.15$.

As mentioned earlier, the hydrodynamic instability has an associated critical wavelength $\Lambda_{crit}$ and it will be instructive to consider also the ratio of filter width $\Delta$ to $\Lambda_{crit}$. The well-known expression of Matalon and Matkowsky (1982) has been used to provide the ratio $\Delta/\Lambda_{crit}$ in Table 2. The value of $\Delta/d_n$ changes with pressure for a given value of $\Delta/\delta_{th}$. For case A (1
bar) $\Delta/\delta_{th} = 0.8, 2.4$ and $4.0$ correspond roughly to $\Delta/d_n = 0.03, 0.09, 0.15$, respectively.

For cases B (5 bar) and C (10 bar) the value of $\Delta/\Lambda_{crit}$ corresponding to $\Delta/d_n$ has to be multiplied with $p^{0.5}$ i.e. with 2.24 and 3.16 respectively. Therefore, one gets $\Delta/\Lambda_{crit} \leq 1$ even for the largest filter width in case C.

Table 2. Ratio of filter width to critical wavelength for hydrodynamic instability $\Delta/\Lambda_{crit}$ for the corresponding values of filter width to flame thickness $\Delta/\delta_{th}$ for cases A, B, C according to Matalon and Matkowsky (1982).

<table>
<thead>
<tr>
<th>$\Delta/\delta_{th}$</th>
<th>0.8</th>
<th>2.4</th>
<th>4.0</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta/\Lambda_{crit}$</td>
<td>0.065</td>
<td>0.200</td>
<td>0.327</td>
</tr>
</tbody>
</table>

Statistical behaviour of the unclosed sub-grid TSF

It is known that the sub-grid TSF exhibits CGT when the velocity jump across the flame due to heat release overcomes the effects of turbulent velocity fluctuation. In order to decide if CGT or gradient transport (GT) is favoured, Veynante et al., (1997) defined a non-dimensional parameter known as the Bray number, which is given by:

$$NB = \frac{\tau S_L}{2\alpha u'} ; \alpha \approx 0.5$$

(8)

It has been demonstrated by Veynante et al. (1997) that CGT is favoured in the case of $NB > 1$ ($NB < 1$). For the present database, the Bray number is $NB \approx 4.5 > 1$ for the inlet value of $u'/S_L$. Hence, predominance of CGT is expected for all flames discussed in this work. Lamioni et al. (2018) reported an enhancement of the extent of CGT for hydrodynamically unstable flames. Accordingly, the amount of CGT should increase from case A to B to C. This can indeed be seen from Fig. 5 where the mean values of the cosine of the angle $\alpha$ between $(\tilde{\rho}u_i\tilde{c} - \tilde{\rho}\tilde{u}_i\tilde{c})$ and $-\partial\tilde{c}/\partial x_i$ conditional upon $\tilde{c}$ are shown for $\Delta/\delta_{th} = 0.8, 4.0$ in cases A-C. A value
of \cos(\alpha) = -1 (+1) indicates that the TSF has the opposite (same) direction as the gradient of Favre filtered reaction progress variable.

\[ \cos(\alpha) = -1 (+1) \]

Figure 5. Profiles of the mean values of the cosine of the angle \( \alpha \) between \((\bar{\rho}\bar{u}_i \bar{c} - \bar{\rho} \bar{u}_i \bar{c})\) and \(-\partial \bar{c}/\partial x_i\) conditional upon \(\bar{c}\) for \(\Delta/\delta_{th} = 0.8, 4.0\) in cases A – C.

As the strength of the TSF cannot be judged from Fig. 5, the profiles of the mean values of normalised TSF projected in flame normal direction, i.e. \(\tau_j M_j/\rho_0 S_L\) conditional upon \(\bar{c}\) for filter widths \(\Delta/\delta_{th} = 0.8, 4.0\) (top) and \(\Delta/d_n = 0.03, 0.15\) (bottom) is shown in Fig. 6. Here, the flame normal vector is defined in terms of the Favre filtered reaction progress variable:

\[ \bar{M} = -\nabla \bar{c}/|\nabla \bar{c}|. \]

Figure 6 shows a pronounced increase of the CGT behaviour particularly for the variation of filter width to nozzle diameter ratio \(\Delta/d_n\). It is worth noting that \(M_i\) has the same orientation as \(-\partial \bar{c}/\partial x_i\) and consequently a negative (positive) value of \(\tau_j M_j\) represents CGT (GT).
Figure 6. Profiles of the mean normalised values of $(\bar{\rho} \bar{u}_c \bar{c} - \bar{\rho} \bar{u}_c \bar{c})$ projected in the resolved flame normal $\vec{M}$ direction (i.e. $\tau_i M_i / (\rho_0 S_L)$) conditional upon $\bar{c}$ for filter widths (top) $\Delta / \delta_{th} = 0.8, 4.0$ and (bottom) $\Delta / d_n = 0.03, 0.15$.

As discussed earlier, the curvature PDFs become increasingly skewed with increasing pressure and it will be interesting to see the interdependence of $\kappa_m \delta_{th}$ and $\tau_i M_i / (\rho_0 S_L)$. Figure 7 shows scatter plots of $\tau_i M_i / (\rho_0 S_L)$ against $\kappa_m \delta_{th}$ for two filter width $\Delta / \delta_{th} = 0.8, 4.0$ for cases A, B and C evaluated in the region of the flame brush $0.01 \leq \bar{c} \leq 0.99$. Curvature and flux values are calculated at the same location. It becomes clear that there is a strong negative correlation between $\kappa_m$ and $\tau_i M_i$ such that the TSF increases from nearly zero at large negative curvature values to a maximum obtained at small negative value $\kappa_m \approx -\varepsilon$ before it decreases again. It is worth mentioning that large positive curvature values do not occur for the present database (see Fig. 2).
Figure 7. Scatter plot of $\tau_i M_i / (\rho_0 S_L)$ against $\kappa_m \delta_{th}$ (left and right) for filter widths $\Delta / \delta_{th} = 0.8, 4.0$ and (top to bottom) 1bar, 5bar, 10bar together with the mean value of $\tau_i M_i / (\rho_0 S_L)$ conditional on curvature.

The $x - y$ plane of the diagrams in Fig. 7 with the $x$ axis represented by $\kappa_m \delta_{th}$ and the $y$ axis represented by $F = \tau_i M_i / (\rho_0 S_L)$ can be divided into four quadrants...
\( V_{i=1,...,4} \) characterised by \( F > 0, \kappa_m > 0; \ F < 0, \kappa_m > 0; \ F < 0, \kappa_m < 0; \ F > 0, \kappa_m < 0 \).

The percentage of samples within the flame front \((0.01 \leq c \leq 0.99)\) in different quadrants, for (left) \( \Delta/\delta_{th} = 0.8 \) and (right) \( \Delta/\delta_{th} = 4.0 \) is shown in Fig. 8, which is consistent with the observation made from Fig. 7 that positive values of \( F \) occur very rarely. Figure 8 clearly shows that for low pressures (case A) negative curvatures dominate (which is consistent with the mean flame geometry) whereas for the high pressure case C the positive bulges lead to a dominance of mildly positive curved flame elements, despite the negative curvature of the mean flame geometry.

**Figure 8. Percentage of samples within the flame front \((0.01 \leq c \leq 0.99)\) in different quadrants, for (left) \( \Delta/\delta_{th} = 0.8 \) and (right) \( \Delta/\delta_{th} = 4.0 \).**

As observed before, high pressure flames are prone to hydrodynamic instabilities due to the decreasing flame thickness. This increases the flame area, the turbulent flame speed and hence the volumetric heat release, which, in general, favours CGT (Chakraborty and Cant, 2009b; Gao et al., 2015). Furthermore, due to streamline convergence upstream of negatively curved flame elements, the velocity increases which possibly further promotes this effect and might explain the maximum values of \( \tau_i M_i \) for small negative curvatures. However, interestingly the TSF attains very small negative values in very large negative curvature regions \((\kappa_m \delta_{th} \ll 1)\) and the flame-flame interaction in this region might be a possible reason behind this behaviour.
Figure 7 also demonstrates a curvature dependence of TSF which possibly is not captured in many existing CGT models.

**LES modelling of the unclosed sub-grid TSF**

This section discusses the LES modelling of the sub-grid TSF based on two different metrics. The Pearson correlation coefficient is used to measure the degree of linear dependence between the TSF evaluated from DNS and the different model expressions given in Eqs. 3-7. It has been evaluated and averaged in the range $0.1 \leq \bar{c} \leq 0.9$. 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. As the correlation coefficient is invariant with respect to constant multipliers, the correct magnitude of the TSF cannot be judged on this basis. Therefore, the second criterion is the magnitude of the model expression (again projected in flame normal direction and denoted $\tau_n$) conditional on the Favre filtered reaction progress variable $\bar{c}$.

For easier presentation the negative of the GHM model is shown in Fig. 9, which demonstrates that the negative correlation between $\tau_{n^{GHM}}$ and $\tau_n$ increases with increasing pressure. This is the third indication that the amount of CGT increases with pressure. Further, as a general trend, the correlation strength decreases with increasing filter width, consistent with earlier analysis (see Gao et al. 2015; Klein et al. 2016, 2018d). The RFM model performs satisfactorily for small filter sizes and the performance improves consistently with increasing pressure. The CGM, CPR and HFM models exhibit reasonable correlations, while the highest values are obtained for the CPR model. Figure 9 also shows that the correlation coefficient between TSFs obtained from DNS and model predictions for a given value of $\Delta/\delta_{th}$ exhibits higher values.
than that in the case of a given value of $\Delta/d_n$, which is consistent with the statement that the model performance is likely to deteriorate for high pressures for a given fixed mesh size.

![Figure 9. Correlation between $(\bar{\rho} \bar{u}_i \bar{c} - \bar{\rho} \bar{u}_i \bar{c})$ and different model expressions averaged over all components (from left to right GHM, CGM, RFM, CPR and HFM) for cases A-C and filter width (top) $\Delta/\delta_{th} = 0.8, 4.0$ and (bottom) $\Delta/d_n = 0.03, 0.16$.](image)

Figure 10 shows the mean values of normalised $(\bar{\rho} \bar{u}_i \bar{c} - \bar{\rho} \bar{u}_i \bar{c})$ and different model predictions projected in the resolved flame normal direction (i.e. $\tau_i M_i/(\rho_0 S_L)$) conditional upon $\bar{c}$ for $\Delta/\delta_{th} = 0.8, 4.0$ in cases A-C. The general observations from Fig. 9 hold also true for Fig. 10. The CGM and CPR models perform reasonably well for small filter widths (e.g. $\Delta/\delta_{th} = 0.8$) whereas the GHM models fail to predict the DNS data. The magnitude of the RMF model is relatively close to the DNS results with a small underprediction, whereas the CPR model overpredicts the sub-grid flux obtained from DNS data. For large filter widths (e.g. $\Delta/\delta_{th} = 4.0$), the CPR and RFM model overpredict the flux magnitude but both models capture the shape of the flux conditional on $\bar{c}$. The quantitative predictions of the CGM and HFM models are relatively poor and the GHM model continues to fail. For large filter widths, the scale
gradient models (CGM and HFM) considerably underpredict the TSF magnitude obtained from DNS data, and this effect is even more pronounced in Fig. 11 where the variation of the mean values of \( \tau_i M_i/(\rho_0 S_L) \) conditional upon \( \tilde{c} \) for a given value \( \Delta/d_n \) is shown.

Figure 10. Profiles of the mean normalised value of \( (\bar{\rho} \bar{u}_i \bar{c} - \bar{\rho} \bar{u}_i \bar{c}) \) and different model predictions projected in the resolved flame normal direction \( \vec{M} \) (i.e. \( \tau_i M_i/(\rho_0 S_L) \)) for (left) filter widths \( \Delta/\delta_{th} = 0.8 \) and (right) \( \Delta/\delta_{th} = 4.0 \). Cases A-C are shown from top to bottom.
Figure 11. Profiles of the normalised mean value of \((\bar{\rho} \bar{u}_i \bar{c} - \bar{\rho} \bar{u}_i \bar{c})\) and different model predictions projected in the resolved flame normal direction \(M\) (i.e. \(\tau_i M_i/(\rho_0 S_L)\)) for \(\Delta/d_n = 0.15\) for (left) case A and (right) case C.

4. DISCUSSION

This section discusses the implications of the findings reported in section 3. Firstly, it is worth noting that there are different ways to analyse the CGT behaviour and quantifying its strength. In fact, one can find three different representations in the paper that allow for making statements regarding the CGT behaviour: Fig. 5 (the cosine between flux and negative gradient of reaction progress), 6 (the flux projected in flame normal direction) and 9 (the correlation coefficient between flux and GHM model). In Fig. 5, the magnitude of the flux does not play a role in contrast to Fig. 6. Similarly, as the correlation coefficient is invariant with respect to constant positive multipliers (i.e. \(corr(a \cdot X, b \cdot Y) = corr(X, Y)\)), it is likely that the strength does not play a role in the results shown in Fig. 9. Figures 5 and 9 indicate differences in CGT behaviour with respect to the orientation of the two vectors - even for the smallest filter width, whereas this is not the case for \(\Delta/\delta_{th} = 0.8\) in Figure 6. It is worth mentioning that the increasing amount of CGT should not be attributed to increasing Karlovitz numbers for cases A to B to C, as CGT effects are typically quantified in terms of the Bray number which is constant for all flames considered in this work. To further illustrate this point one additional 1
bar flame, case E, has been analysed from the extended database presented in Klein et al. (2018a,b). The ratio of $l/\delta_{th}$ has been modified in case E such that cases C and E have same values of $u'/S_L$ and $l/\delta_{th}$ and thus they occupy the same location on the regime diagram. Consequently, case E has the same Karlovitz number as case C but corresponds to a 1 bar case rather than 10 bar. The results (see Appendix) indicate a reduced amount of CGT relative to Case C (10 bar) although both Bunsen flames occupy the same location on the regime diagram and have the same Karlovitz number. Instead, cases A and E have been found to show similar amount of CGT.

This paper analyses five models and based on their observed performances the possible ways to improve the models is discussed here. There is no way to fix the GHM model as this model is not able to capture CGT transport which is prominent in the flames discussed here. The RFM model consists of both CGT and GT transport contributions. The turbulence intensity is very low for the present database ($u'/S_L = 1.0$) which suggests that one encounters $u'_\Delta \ll S_L$ for the SGS contribution. As a result of this, the RFM model is dominated by the second contribution $-\rho_0 S_L M_i (\bar{c} - \bar{c})$. As can be seen from Figs. 10 and 11, the RFM model in some cases underpredicts but in most cases overpredicts the magnitude of the TSF. This potentially could be corrected by altering the model parameter $C_L = 0.12$ in Eq. 4. A least square fit for determining this parameter reveals that the optimal value varies considerably in the range from $0.55 C_L$ to $2.32 C_L$ for the results presented in Figs. 10 and 11. Furthermore, by increasing the gradient contribution (by increasing $C_L$), the profile becomes erroneously skewed towards the burned gas side. Therefore adjusting $C_L$ is not enough to obtain a satisfactory prediction from the RFM model. A similar CGT / GT balance holds for the HFM model which is dominated by the term $\bar{\rho} D_{an} \Delta^2 \bar{s}_{ik} (\partial \bar{c} / \partial x_k)$ which bears similarity with the Clark model $\bar{\rho} (\Delta^2/12) (\partial \bar{u}_i / \partial x_k) (\partial \bar{c} / \partial x_k)$. In fact, both models show similar results as indicated in Figs. 10 and 11.
10 and 11 and the CGM model will be discussed in more detail in the following. The CGM model can essentially be derived by approximating the convolutional filter using a Taylor series. This approximation loses accuracy with increasing filter width. This is the main reason for the deterioration of the CGM model for large filter width. The second reason is that the gradients in the CGM model have to be approximated using finite difference formulas, typically central differences. In a real LES, the accuracy of approximation decreases with increasing filter (i.e. grid) size and it can be shown by using the modified wavenumber diagram that this results in underprediction of the magnitude of the gradients (Klein et al. 2017). A simple multiplicative correction is not an option here because the modelled flux has a different profile in reaction progress variable space than the true flux obtained from DNS (see Figs. 10 and 11). One could, in principle, filter a laminar flame profile, calculate the flux and use the ratio with the CGM model, evaluated on the filtered data, as a correction (which will depend on reaction progress variable). However, this approach is unlikely to be general and will fail for higher turbulence intensities. Hence, this approach is not recommended. The CPR model is the sum of the CGM model and the RFM model which both have been discussed before. The CPR model was suggested by Klein et al. (2018d) with the idea that the CGM model part will result in good correlations for all flux components and that the RFM contribution will properly predict the mean value of TSF conditional on $\tilde{c}$. Equation 7 does not contain model constants because the CPR model should be considered a possible modelling strategy rather than a final model. In fact, Fig. 9 shows that the CPR model has the highest correlation coefficients among all models considered in this work. However, it overpredicts the TSF magnitude for the current database. By using a least square fit for determining a correction factor multiplying the whole model, the optimal parameter has been found in the range between 0.63 and 0.75 with a mean value of 0.68, for the results presented in Figs. 10 and 11.

By setting $C_L = 0$ in the RFM model a pure CGT type model is obtained:
\[ \tau_{i}^{GT} = -\rho_0 S_L M_i (\bar{c} - \bar{c}) \]  \hspace{1cm} (9)

The correlation coefficients of this model are nearly identical to the RFM model with only very small differences. However, the model overpredicts the TSF for all cases and all filter widths due to the missing GT contribution. By using a least square fit for determining a correction factor multiplying the whole model, the optimal parameter has been found in the range between 0.71 and 0.90 with a mean value of 0.82, for the results presented in Figs. 10 and 11. For the present database, this model seems to be the overall best available modelling option. It is worth noting though that the variable \( \bar{c} \) in this model is unknown and requires modelling itself in real LES and the same holds true for the RFM model. The modelling of \( \bar{c} \) is discussed in Chakraborty and Cant (2007). It is also worth mentioning that this model will not account for the curvature dependence of the TSF which has been illustrated in Fig. 7, and explicit modelling of curvature dependence could be a future possibility. The performance of this model is very satisfactory for the present database, but it will fail for higher turbulence intensities when the magnitude of GT flux increases.

Although this affects only a small region of the flame surface, explicit modelling of curvature dependence could be a future possibility. Gradient type models (or equivalently scale similarity type models) exhibit deterioration of their performances for large filter widths, and therefore are unlikely to provide reasonable closure for high pressure flames. The CPR model represents a new modelling strategy which merits further investigation. A note of caution is that the values of all the model parameters reported in this section are only valid for the present database and need to be adjusted for other combustion regimes, potentially using a dynamic approach where averaging is done conditional on reaction progress variable (Klein et al., 2015).
5. CONCLUSIONS

The effect of pressure on the statistical behaviour and modelling of sub-grid turbulent scalar fluxes (TSFs) in turbulent premixed Bunsen flames have been analysed using three-dimensional DNS data. While many physical effects do not change with pressure, provided the respective quantities are properly normalised, the decreasing flame thickness with increasing pressure promotes fluid-dynamic instabilities and this acts in favour of counter gradient transport. To analyse these effects, DNS data for three different turbulent premixed Bunsen flames at pressure levels of 1, 5, 10 bar, have been explicitly filtered for a range of filter widths in order to extract the terms of the transport equation of Favre-filtered reaction progress variable, and the sub-grid TSF. The latter quantity has been further used to assess the performance of a variety of algebraic LES closures for the sub-grid TSF. The simulation results indicate that the behaviours of the terms in the Favre-filtered reaction progress variable transport remain similar for different pressure levels, provided they are appropriately normalised by the combination of unburned density, laminar burning velocity and thermal flame thickness. However, as the filter size to flame thickness ratio is likely to be large for high pressure flames, the qualitative and quantitative behaviours of the terms is likely to change with increasing pressure. Using different indicators, it has been demonstrated that the extent of counter-gradient transport (CGT) increases with increasing pressure. It has been suggested that the reason for the increased CGT is the higher volumetric heat release due to enhanced flame wrinkling combined with streamline convergence upstream of negatively curved flame elements. While a subset of the models shows high correlation for the sub-grid TSFs and predicts the magnitude of the sub-grid TSFs reasonably well for small filter sizes, several models considerably underpredict the sub-grid turbulent scalar flux for large filter widths. It is worth noting that this paper focuses on the flames located on the boundary of the wrinkled and the corrugated flamelets regimes to demonstrate the effects of hydrodynamic instabilities on
statistical behaviour and closure of sub-grid TSFs. It is worth assessing if the effects of Darrieus-Landau instability sustain once the turbulence intensity increases, which will form the basis of future investigations.

APPENDIX

Table 3. Inlet flow parameters for case E.

<table>
<thead>
<tr>
<th>Case</th>
<th>( P/P_0 )</th>
<th>( Re_d )</th>
<th>( Re_t )</th>
<th>( U_B/S_L )</th>
<th>( u'/S_L )</th>
<th>( l/d_n )</th>
<th>( l/\delta_{th} )</th>
<th>( Ka )</th>
<th>( Da )</th>
</tr>
</thead>
<tbody>
<tr>
<td>E</td>
<td>1.0</td>
<td>399</td>
<td>41.22</td>
<td>6.0</td>
<td>1.0</td>
<td>3/5</td>
<td>16.13</td>
<td>0.25</td>
<td>16.13</td>
</tr>
</tbody>
</table>

Figure 12. Profiles of the mean values of the cosine of the angle \( \alpha \) between \((\vec{\rho u}_t \bar{c} - \vec{\rho u}_t \bar{c})\) and \(-\partial \bar{c}/\partial x_i\) conditional upon \( \bar{c} \) for \( \Delta/\delta_{th} = 4.0 \) in case E.

ETHICS STATEMENT

This work did not involve any active collection of human data.

COMPETING INTERESTS STATEMENT

We have no competing interests.

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REFERENCES


Klein, M., Kasten, C., Chakraborty, N., Mukhadiyev, N. and Im, H.G., 2018d. Turbulent scalar fluxes in H2-air premixed flames at low and high Karlovitz numbers, Combust. Theory Modelling, 22, 1-16.


