A Comparison between Statistical Behaviours of Scalar Dissipation Rate between Homogeneous MILD Combustion and Premixed Turbulent Flames

Frederick W. Young, Hazem S. A. M. Awad, Khalil Abo-Amsha, Umair Ahmed and Nilanjan Chakraborty

School of Engineering, Newcastle University, Newcastle-upon-Tyne NE1 7RU, UK
* Correspondence: nilanjan.chakraborty@newcastle.ac.uk

Abstract: Three-dimensional Direct Numerical Simulations (DNS) data has been utilised to analyse statistical behaviours of the scalar dissipation rate (SDR) and its transport for homogeneous methane-air mixture turbulent Moderate or Intense Low oxygen Dilution (MILD) combustion for different O_2 dilution levels and turbulence intensities for different reaction progress variable definitions. Additional DNS has been conducted for turbulent premixed flames and passive scalar mixing for the purpose of comparison with the SDR statistics of the homogeneous mixture MILD combustion with that in conventional premixed combustion and passive scalar mixing. It has been found that the peak mean value of the scalar dissipation rate decreases with decreasing O_2 concentration for MILD combustion cases. Moreover, SDR magnitudes increase with increasing turbulence intensity for both MILD and conventional premixed combustion cases. The profiles and mean values of the scalar dissipation rate conditioned upon the reaction progress variable are found to be sensitive to the choice of the reaction progress variable definition. This behaviour arises due to the differences in the distributions of the species mass fractions within the flame. The strain rate contribution and the molecular dissipation term are found to be the leading order contributors in the scalar dissipation rate transport for MILD combustion; whereas, in conventional premixed flames, the terms rising from density variation and reaction rate gradient also play leading roles in addition to the strain rate and molecular dissipation contributions. By contrast, the terms due to density gradient and reaction rate gradient remain negligible in comparison to the leading order contributors in MILD combustion cases due to small density variation because of moderate temperature rise and small reaction rate gradient magnitudes. Furthermore, the qualitative behaviour of the strain rate contribution to the SDR transport in premixed flames is significantly different to that in the case of MILD combustion and passive scalar mixing. The findings of the current analysis indicate that the scalar dissipation rate statistics in MILD combustion show several qualitative similarities to the passive scalar mixing despite major differences with the SDR transport in conventional turbulent premixed flames. This further suggests that the scalar dissipation rate models, which were originally proposed in the context of passive scalar mixing, have the potential to be applicable for MILD combustion but the models for the premixed turbulent combustion may not be applicable for MILD combustion of homogeneous mixtures.

Keywords: MILD combustion; premixed combustion; passive scalar mixing; scalar dissipation rate; direct numerical simulations

1. Introduction

Scalar dissipation rate (SDR) is a quantity of fundamental importance in the analysis of turbulent reacting flows, as it characterises the rate of micromixing [1] and can be used for the reaction rate closure in turbulent premixed combustion [2]. Therefore, it is worthwhile to consider the evolution process of SDR within the flame for its closure in turbulent reacting flows. Although SDR statistics for turbulent premixed [2] and non-premixed [1,3]
combustion have been analysed extensively, limited effort has been directed to the analysis of the SDR statistics for Moderate or Intense Low oxygen Dilution (MILD) combustion despite the importance of SDR in its modelling [4–8].

MILD combustion is becoming increasingly popular because of the advantage it offers in terms of high energy efficiency and low pollutant (e.g., NOx) emission as a result of moderate temperature rise [9] and has applications in industrial furnaces within the ceramic, glass and chemical industries and combustion locally in the primary zone in gas turbines can take place in MILD mode. Therefore, high-fidelity modelling of MILD combustion is needed to utilise the potentials offered by MILD combustion mode and SDR closure plays a key role in this context because of its pivotal role in combustion and micro-mixing modelling [4–8].

It has recently been demonstrated that there are significant differences in the statistical behaviours of the reactive scalar gradient between homogeneous mixture MILD combustion and conventional turbulent premixed combustion [10], and these differences arise principally due to the weakened thermal expansion effects in MILD combustion as a result of small temperature rise. This further suggests that the statistical behaviour and the evolution of the SDR in MILD combustion of homogeneous mixtures are likely to be different from that in conventional premixed turbulent combustion [10]. Moreover, in the absence of heat release, the SDR transport of the passive scalar in MILD combustion may exhibit similarities with the SDR transport of the passive scalar in the case of pure mixing [10]. However, the similarities and differences in the SDR statistics between MILD combustion, conventional premixed turbulent flames and pure scalar mixing are yet to be analysed in the existing literature; however, this information is important for modelling of micromixing in MILD combustion [1–8]. This gap in the literature is addressed in the current study by considering Direct Numerical Simulation (DNS) data of homogeneous methane-air mixture combustion under MILD conditions for different turbulence intensities and oxygen dilution levels to analyse the statistical behaviour of the SDR of reactive scalars and the terms of their transport equations. Moreover, to compare the statistical behaviours of the SDR in MILD combustion with conventional premixed flames and passive scalar mixing, additional DNS simulations of representative statistically planar turbulent premixed flames and passive scalar mixing in a canonical configuration have been considered. In this respect, the objectives of the current analysis are:

(a) to demonstrate the effects of turbulence intensity and O2 dilution on the statistical behaviour of the SDR and the terms of its transport equation in MILD turbulent combustion

(b) to illustrate the differences in the SDR transport behaviour between homogeneous mixture MILD combustion and conventional premixed flames, and between homogeneous mixture MILD combustion and passive scalar mixing

(c) to provide physical explanations for (a) and (b) and indicate the modelling implications.

The rest of the paper is organised as follows. The mathematical background and numerical implementation pertaining to the analysis presented in this paper are discussed in the next two sections. Following that, the results are presented and subsequently discussed. The conclusions are drawn, and a summary of the main findings is provided in the final section of this paper.

2. Mathematical Background

In homogeneous mixture combustion, the progress of the chemical reaction is quantified by a reaction progress variable \(c\), which can be defined as [11]:

\[
c = (Y_a - Y_{a,u}) / (Y_{a,b} - Y_{a,u})
\]

where subscripts \(u\) and \(b\) refer to values in fully unburned and fully burned mixtures, respectively, and \(Y_a\) is the mass fraction of the species \(a\) based on which the reaction
progress variable is defined. The transport equation of reaction progress variable for homogeneous mixture combustion takes the following form \[11\]:

\[
\frac{\partial (\rho c)}{\partial t} + \frac{\partial (\rho u_j c)}{\partial x_j} = \frac{\partial}{\partial x_j} \left( \rho D_c \frac{\partial c}{\partial x_j} \right) + \dot{w}_c \tag{2}
\]

where \(\rho, u_j, D_c\) and \(\dot{w}_c\) are gas density, \(j^{\text{th}}\) component of the velocity vector, reaction progress variable diffusivity and reaction rate of reaction progress variable, respectively. The reaction rate of the reaction progress variable \(\dot{w}_c\) is defined as \[11\]:

\[
\dot{w}_c = \dot{w}_a / (Y_{a,b} - Y_{a,u}) \tag{3}
\]

It can be appreciated from Equations (1)–(3) that the values of \(D_c\) and \(\dot{w}_c\) are dependent on the choice of species \(\alpha\) based on which the reaction progress variable is defined. Here, \(c\) definitions based on CH\(_4\), O\(_2\), CO\(_2\) and H\(_2\)O mass fractions have been considered.

The scalar dissipation rate \(N_c\) is defined as \[1\]:

\[
N_c = D_c \nabla c \cdot \nabla c \tag{4}
\]

Taking the gradient of Equation (2) and multiplying the resulting equation by \(\nabla c\) yields a transport equation of SDR \(N_c\) in the following manner by ignoring the contribution arising from diffusivity gradients \[2,12\]:

\[
\frac{\partial (\rho N_c)}{\partial t} + \frac{\partial (\rho u_j N_c)}{\partial x_j} = \frac{\partial}{\partial x_j} \left( \rho D_c \frac{\partial c}{\partial x_j} \right) + T_1 + T_2 + T_3 - D_2 \tag{5}
\]

where,

\[
T_1 = -2D_c \left( \dot{w}_c + \nabla \cdot (\rho D_c \nabla c) \right) \frac{\partial c}{\partial x_i} \frac{\partial \rho}{\partial x_k} \tag{6a}
\]

\[
T_2 = -2\rho D_c \frac{\partial c}{\partial x_i} \frac{\partial u_i}{\partial x_j} \frac{\partial c}{\partial x_j} \tag{6b}
\]

\[
T_3 = 2D_c \frac{\partial \dot{w}_c}{\partial x_i} \frac{\partial c}{\partial x_j} \tag{6c}
\]

\[
D_2 = 2\rho D^2_c \frac{\partial^2 c}{\partial x_i \partial x_j} \frac{\partial^2 c}{\partial x_i \partial x_j} \tag{6d}
\]

The terms on the left-hand side represent the transient and advection terms. The term \(D_1\) represents the molecular diffusion of the SDR, whereas the terms \(T_1\) and \(T_2\) arise due to density gradient and flame normal strain rate, and are referred to as the density variation and scalar turbulence-interaction (or strain rate term) terms, respectively. The term \(T_3\) is the reaction rate contribution, which arises due to the correlation between the reaction rate and scalar gradients. The term \(D_2\) represents the molecular dissipation rate of the SDR. The statistical behaviours of \(N_c\), \(T_1\), \(T_2\), \(T_3\) and \((-D_2)\) will be analysed in Section 4 of this paper for MILD combustion for different turbulence intensities and O\(_2\) dilution levels, and also for conventional premixed flames for different definitions of reaction progress variable \(c\).

In the context of passive scalar mixing, the term \(T_3\) is identically zero and the contribution of \(\dot{w}_c\) in \(T_1\) is identically zero in the SDR transport equation where \(c\) should be interpreted as the concentration of a scalar that is being mixed.
3. Numerical Implementation

The statistical behaviours of the SDR and the terms of its transport equation have been analysed using 3D DNS data generated using a compressible code known as SENGA2 [13]. In SENGA2, the standard transport equations of mass, momentum, energy, and species for turbulent reacting flows are solved in the context of the finite-difference approach. The first and second-order spatial differentiation operations are approximated using a tenth-order central difference scheme for the internal grid points and the order of accuracy gradually reduces to a one-sided fourth-order scheme at the non-periodic boundaries. A fourth-order low-storage Runge–Kutta scheme is used for explicit time advancement. The non-periodic boundaries are specified according to Navier-Stokes Characteristic Boundary Condition (NSCBC) approach [14]. A skeletal chemical mechanism comprising 16 species and 25 reactions (including 10 reversible reactions) has been taken to represent the chemical kinetics of \( \text{CH}_4/\text{Air} \) combustion [15]. A domain size of 10 mm \( \times \) 10 mm \( \times \) 10 mm is discretised by a Cartesian grid of \( 252 \times 252 \times 252 \) where uniform grid spacing has been used for the simulations of MILD \( \text{CH}_4 \)-air mixtures with \( \text{O}_2 \) dilution percentages of 3.5% and 4.8% by volume for an equivalence ratio of \( \phi = 0.8 \) and an initial temperature of about 1500 K (i.e., \( T_0 \approx 1500 \) K). The unburned gas temperature of 1500 K is higher than the autoignition temperature (i.e., \( T_{\text{ign}} = 1100 \) K based on well-stirred reactor calculations [16–20]) for \( \text{CH}_4 \)-air mixtures with \( \text{O}_2 \) dilution percentages of 3.5% and 4.8% by volume for an equivalence ratio of \( \phi = 0.8 \). The computational domain has a reflecting inflow with specified density on the left-hand side boundary in the \( x \)-direction and a partially non-reflective outflow boundary condition is specified for the right-hand side boundary in the \( x \)-direction. All other directions are taken to be periodic.

DNS calculations of statistically planar conventional turbulent premixed flames of \( \phi = 0.8 \) with an unburned gas temperature of 300 K have been conducted for a domain size of 20 mm \( \times \) 10 mm \( \times \) 10 mm, which is discretised by a uniform Cartesian grid of 504 \( \times \) 252 \( \times \) 252. A longer domain is taken for turbulent premixed flame simulations to allow for flame propagation in the mean direction of flame propagation which is taken to align with \( x \)-direction. For premixed flame simulations, the domain boundaries in the direction of mean flame propagation are taken to be partially non-reflecting and transverse boundaries are taken to be periodic.

The grid spacing ensures that the flame thickness \( \delta_{\text{th}} = (T_P - T_0) / \max |\nabla T|_L \) (with \( T_r \), \( T_P \) and \( T_0 \) being the instantaneous, product and reactant temperatures, respectively, and subscript \( L \) refers to the values in the corresponding 1D unstretched premixed flame) is resolved by using at least 12 grid points and the Kolmogorov length scale \( \eta \) is resolved by at least 1.5 grid points for all turbulent cases.

The thermochemical parameters in the unburned gas, including the mole fractions of the reactants, unstretched laminar burning velocity, unburned gas temperature and thermal flame thickness, for the MILD and premixed flame cases, are shown in Table 1. The simulations have been carried out for different initial values of turbulence intensities \( u'/S_L \) for a given normalised value of integral length scale of turbulence \( l/\delta_{\text{th}} \) where \( S_L \) is the unstretched laminar burning velocity for the corresponding mixture composition and unburned gas temperature and \( l \) is the integral length scale of turbulence. The initial values of \( u'/S_L \) and \( l/\delta_{\text{th}} \) along with the values of Damköhler number \( Da = lS_L / u'\delta_{\text{th}} \) and Karlovitz number \( Ka = (u'/S_L)^{3/2}(l/\delta_{\text{th}})^{-1/2} \) are listed in Table 2. The values of \( Da \) and \( Ka \) indicate that the cases considered here occupy nominally the same location in the Borghi–Peters diagram [21] and represent combustion within the thin reaction zones regime [21].
Table 1. Thermochemical conditions.

<table>
<thead>
<tr>
<th>Case</th>
<th>$X_{O_2}$</th>
<th>$X_{CO_2}$</th>
<th>$X_{H_2O}$</th>
<th>$X_{CH_4}$</th>
<th>$X_{N_2}$</th>
<th>$S_L$ (m/s)</th>
<th>$T_0$ (K)</th>
<th>$\delta_{th}$ (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MILD–3.5% $O_2$</td>
<td>0.035</td>
<td>0.066</td>
<td>0.132</td>
<td>0.014</td>
<td>0.753</td>
<td>2.3</td>
<td>1500</td>
<td>0.8</td>
</tr>
<tr>
<td>MILD–4.8% $O_2$</td>
<td>0.048</td>
<td>0.061</td>
<td>0.121</td>
<td>0.019</td>
<td>0.751</td>
<td>3.2</td>
<td>1500</td>
<td>0.6</td>
</tr>
<tr>
<td>Premixed</td>
<td>0.194</td>
<td>0.0</td>
<td>0.0</td>
<td>0.077</td>
<td>0.729</td>
<td>0.3</td>
<td>300</td>
<td>0.5</td>
</tr>
</tbody>
</table>

Table 2. Initial Turbulence parameters.

<table>
<thead>
<tr>
<th>Case</th>
<th>$u'/S_L$</th>
<th>$l/\delta_{th}$</th>
<th>$Da$</th>
<th>$Ka$</th>
</tr>
</thead>
<tbody>
<tr>
<td>MILD–3.5% $O_2$</td>
<td>4.0</td>
<td>2.5</td>
<td>0.62</td>
<td>5.06</td>
</tr>
<tr>
<td></td>
<td>8.0</td>
<td>2.5</td>
<td>0.31</td>
<td>14.31</td>
</tr>
<tr>
<td>MILD–4.8% $O_2$</td>
<td>4.0</td>
<td>2.5</td>
<td>0.62</td>
<td>5.06</td>
</tr>
<tr>
<td></td>
<td>8.0</td>
<td>2.5</td>
<td>0.31</td>
<td>14.31</td>
</tr>
<tr>
<td>Premixed</td>
<td>4.0</td>
<td>2.5</td>
<td>0.62</td>
<td>5.06</td>
</tr>
<tr>
<td></td>
<td>8.0</td>
<td>2.5</td>
<td>0.31</td>
<td>14.31</td>
</tr>
</tbody>
</table>

The MILD combustion simulations involve two stages. The first stage mimics the mixing of the products of combustion with the reactant mixture. The second stage considers the subsequent combustion process [16–20]. The species field resulting from the first stage serves as the initial as well as the inflow fields for the second stage. The first stage is representative of what happens inside the recirculation zone of a real MILD combustor. The mixture at stage 1 is prepared according to the following steps [16–20]:

- Turbulent velocity fluctuations are initialised by a homogenous isotropic field generated using a well-known pseudo-spectral method [22] following the Batchelor–Townsend spectrum [23].
- 1D laminar premixed flames are simulated for the thermochemical conditions shown in Table 1. An initial bimodal distribution of $c$ based on CH$_4$ mass fraction with prescribed integral length scale $l_c = 0.0015$ m is then developed using the methodology by Eswaran and Pope [24]. The 1D laminar profiles of species mass fraction for $\phi = 0.8$ are specified as a function of the progress variable $c$ based on the CH$_4$ mass fraction. These functions alongside the bimodal distribution of $c$ are used to initialise the density and species mass fractions corresponding to atmospheric pressure and an unburned gas temperature of 1500 K (i.e., $T_0 = 1500$ K).
- The generated bimodal scalar fields in the previous step are then allowed to evolve under the generated turbulence for about 1 eddy turnover time ($l/u'$) in a periodic domain mimicking the EGR in MILD combustion. At the end of this step, the mean and variance of the preprocessed $c$ field are $\langle c \rangle \approx 0.50$ and $\langle c^2 \rangle \approx 0.09$, respectively, where $\langle \cdot \rangle$ is the mean value evaluated over the whole domain. The temperature in the preprocessed mixture has a variation of about $\pm 0.3\%$ of its mean value.

The prepared mixture at the end of the previous stage will then be fed into the domain through the inlet with a mean velocity $U_{in} = 20$ m/s by scanning a plane through it following the previous work by Minamoto, Swaminathan and co-workers [16–20]. In the case of the passive scalar mixing simulations, the generated bimodal field of the scalar is allowed to evolve under turbulence without any chemical reactions. The species mass fractions, temperature and density for turbulent premixed flame simulations are initialised by the steady unstretched 1D laminar premixed flame with $\phi = 0.8$. A pseudo-spectral method [22] is used to initialise turbulent velocity fluctuations by a homogeneous isotropic field with prescribed values of $u'$ and $l$ according to following the Batchelor–Townsend spectrum [23].
The MILD combustion simulations have been continued for 2.5 through-pass times (i.e., \( t_{sim} = 2.5L_x/U_{mean} \)), which amounts to more than 10 initial eddy turnover times for the cases considered here. This ensures that the statistics extracted at the end of the simulation remains independent of the homogeneous isotropic velocity field used for initial and inflow conditions. The premixed turbulent flame simulations under decaying turbulence need to be conducted for \( t_{sim} = \max(1/u', \delta_{th}/S_L) \) in order to ensure that the statistics extracted at the end of the simulation remain independent of initial homogeneous isotropic turbulence. In the present analysis, the premixed flame simulations have been continued for one chemical timescale (i.e., \( t_{sim} = \delta_{th}/S_L \)), which amounts to 1.3 and 2.7 initial eddy turnover times (i.e., \( 1.3l/u' \) and \( 2.7l/u' \)) for initial \( u'/S_L = 4.0 \) and \( 8.0 \) cases, respectively. By that time, the flame surface area and volume-integrated reaction rate of the reaction progress variable reached quasi-steady state. These simulation times remain comparable to several previous studies [16–20,25–32], which contributed significantly to the fundamental understanding and modelling in the past.

4. Results and Discussion

The instantaneous views of \( c = 0.5 \) isosurface for MILD combustion, passive scalar mixing, and conventional premixed flame cases are shown in Figure 1. It can be seen from Figure 1 that the \( c = 0.5 \) isosurface extends over a significant part of the simulation domain in the MILD combustion cases, whereas this isosurface separates reactants from products in turbulent premixed flames. Moreover, there is a significant amount of interaction of reaction progress variable isosurfaces in MILD combustion cases, whereas this is not prevalent in the premixed flame cases. The distributions of \( c = 0.5 \) isosurfaces for passive scalar mixing are found to be qualitatively similar to the \( c = 0.5 \) isosurfaces shown for the MILD combustion cases. This can be further confirmed from the spatial distributions of \( c \) based on CH\(_4\) mass fraction and the natural logarithm of its normalised SDR \( \ln(N_c \times l_0/u'_0) \) in the central midplane, which are shown in Figures 2–4 for MILD combustion, premixed combustion and passive scalar mixing cases, respectively, where \( u'_0 \) and \( l_0 \) correspond to the initial rms velocity and integral length scale for the MILD combustion case with 4.8% O\(_2\) dilution with \( u'/S_L = 4.0 \) and \( l/\delta_{th} = 2.5 \) and the logarithm of the SDR is used to clearly demonstrate the variations of scalar gradients within the computational domain. It can be seen from Figures 2–4 those high values of \( \ln(N_c \times l_0/u'_0) \) are distributed over a large part of the domain in the MILD cases and this tendency strengthens with decreasing O\(_2\) concentration. By contrast, high SDR values are obtained within the reaction zone separating reactants and products in the premixed flame cases.

A comparison between Figures 2 and 4 reveals that the SDR distribution in the case of passive scalar mixing is qualitatively similar to that in the MILD combustion cases. Figures 2–4 further suggest that the SDR magnitude increases with increasing turbulence intensity for MILD combustion, premixed combustion and passive scalar mixing cases considered here. This can further be substantiated from Figure 5a,b, where the profiles of the mean values of the normalised SDR \( N_c \times l_0/u'_0 \) conditioned upon \( c \) for different definitions of reaction progress variable \( c \) for MILD and premixed combustion cases are shown, respectively.
Figure 1. Instantaneous views of $c = 0.5$ isosurface for homogeneous mixture MILD combustion with 3.5% and 4.8% O$_2$ concentration (1st and 2nd row), premixed combustion (3rd row) and passive scalar mixing (4th row) for initial $u'/S_L = 4.0$ (1st column) and 8.0 (2nd column).
Figure 2. Spatial distributions of $c$ based on CH$_4$ mass fraction (1st column) and the natural logarithm of its normalised SDR $\ln(N_c \times l_0/\mu'_0)$ (2nd column) in the central midplane for homogeneous mixture MILD combustion with 3.5% (1st and 3rd row) and 4.8% O$_2$ concentration (2nd and 4th row) for initial $u'/S_L = 4.0$ (1st and 2nd row) and 8.0 (3rd and 4th row).
A comparison between Figures 2 and 4 reveals that the SDR distribution in the case of passive scalar mixing is qualitatively similar to that in the MILD combustion cases. Figures 2–4 further suggest that the SDR magnitude increases with increasing turbulence intensity for MILD combustion, premixed combustion and passive scalar mixing cases.

Figure 3. Spatial distributions of $c$ based on CH$_4$ mass fraction (1st column) and the natural logarithm of its normalised SDR $\ln(N_c \times l_0/\bar{u}'_0)$ (2nd column) in the central midplane for the premixed combustion for initial $u'/S_L = 4.0$ (1st row) and 8.0 (2nd row).

Figure 4. Spatial distributions of $c$ based on CH$_4$ mass fraction (1st column) and the natural logarithm of its normalised SDR $\ln(N_c \times l_0/\bar{u}'_0)$ (2nd column) in the central midplane for passive scalar mixing for initial $u'/S_L = 4.0$ (1st row) and 8.0 (2nd row).
considered here. This can further be substantiated from Figure 5a, b, where the profiles of the mean values of the normalised SDR $N_c \times l_0/u'_0$ conditioned upon $c$ for different definitions of reaction progress variable $c$ for MILD and premixed combustion cases are shown, respectively. (a) MILD combustion (3.5\% O$_2$-red, 4.8\% O$_2$-blue), (b) premixed combustion (magenta), and (c) passive scalar mixing (green) cases considered here with $u'/S_L = 4$ (solid line) and $u'/S_L = 8$ (dashed line).

**Figure 5.** Profiles of the mean values of the normalised SDR $N_c \times l_0/u'_0$ conditioned upon $c$ for different definitions of reaction progress variable $c$ for MILD combustion (3.5\% O$_2$-red, 4.8\% O$_2$-blue), premixed combustion (magenta), and passive scalar mixing (green) cases considered here with $u'/S_L = 4$ (solid line) and $u'/S_L = 8$ (dashed line).
It can be seen from Figure 5a,b that the peak mean value of $N_c \times I_0/u'_0$ increases with increasing turbulence intensity for the MILD combustion cases. The mean values of $N_c \times I_0/u'_0$ conditional upon $c$ for passive scalar mixing are shown in Figure 5c, which also shows an increasing trend of the SDR with increasing turbulence intensity. However, the peak value of the normalised SDR for turbulent premixed flames does not change appreciably with increasing turbulence intensity. It can further be seen that the peak mean value of $N_c \times I_0/u'_0$ decreases with increasing dilution (i.e., from 4.8% to 3.5% O$_2$ by volume), which is a consequence of the thicker flame and smaller flame speed. Moreover, the mean values of $N_c \times I_0/u'_0$ in turbulent MILD combustion cases have been found to be considerably smaller than the $N_c \times I_0/u'_0$ values of premixed flame cases listed in Table 1. This is consistent with the significant drop in $|\nabla c|$ in turbulent MILD combustion cases in comparison to the corresponding laminar premixed flame cases, which has been explained elsewhere by the present authors [10]. Furthermore, a comparison between Figure 5a,b reveals that there are significant qualitative differences in the mean variation of $N_c \times I_0/u'_0$ for different choices of reaction progress variable for MILD combustion and premixed flame cases. For both cases, it has been found that the highest mean SDR values are obtained for the CH$_4$ mass fraction-based reaction progress variables and the values for O$_2$ and H$_2$O mass fraction-based reaction progress variable remain comparable, and relatively smaller mean SDR values are obtained for CO$_2$ mass fraction-based reaction progress variable. The relative thicknesses of the reaction layers of different species are responsible for differences in mean SDR values for different choices of $c$. Furthermore, a comparison between Figure 5a,c reveals that the SDR profiles between the MILD combustion and passive scalar mixing cases are both qualitatively and quantitatively similar, which suggests a predominantly mixing-driven behaviour in MILD combustion cases. In order to explain the behaviours of $N_c$ in response to the variations of turbulence intensity, O$_2$ dilution and the choices of reaction progress variable, it is worthwhile to analyse the statistical behaviours of the terms of the SDR $N_c$ transport equation (Equation (5)).

The mean contributions of the terms on the right-hand side of Equation (5) (i.e., $\{D_1, T_1, T_2, T_3, -D_2\} \times I_0^2/\rho_0 u'_0^2$) conditioned upon $c$ for MILD, premixed flame and passive scalar mixing cases are shown in Figures 6–8, respectively, for different choices of $c$, where $\rho_0 u'_0$ is the unburned gas density for MILD case with 4.8% O$_2$ (low dilution) and $u'/S_L = 4.0$. For all cases, the mean value of the molecular dissipation term $(-D_2)$ acts as a leading order sink in the SDR transport. It can be seen from Figures 6–8 that the mean molecular diffusion term $D_1$ assumes mostly positive values towards the unburned gas side before assuming negative values following positive values towards the burned gas region for all cases. Although the magnitude of the mean value of $D_1$ is either comparable to or greater than the leading order sink contribution of the molecular dissipation term $(-D_2)$, the molecular diffusion term $D_3$ redistributes the SDR but does not generate/dissipate it, and its volume integrated value vanishes in all cases.

The mean contribution of the term arising from density variation $T_1$ remains negligible in comparison to the leading order contribution of the molecular dissipation term $(-D_2)$ for all MILD combustion cases and this term is identically zero in the passive scalar mixing case with a small Mach number as in the case considered here. By contrast, the mean value of $T_1$ remains positive throughout the flame front and its magnitude is not negligible in comparison to the other leading order contributors in the premixed flame cases.
Figure 6. Profiles of the mean contributions of the terms on the right-hand side of the SDR transport equation (i.e., \( \{D_1, T_1, T_2, T_3, -D_2\} \times \frac{l_0^2}{\rho_0 u_0'^2} \)) conditioned upon \( c \) for different definitions of reaction progress variable \( c \) for homogeneous mixture MILD combustion cases with 3.5% (1st and 3rd row) and 4.8% \( O_2 \) concentration (2nd and 4th row) for initial \( u'/S_L = 4.0 \) (1st and 2nd row) and 8.0 (3rd and 4th row).

The mean value of the scalar-turbulence interaction term \( T_2 \) assumes positive values for the MILD combustion and passive scalar mixing cases. In contrast, the mean value of \( T_2 \) remains negative throughout the flame and its magnitude remains comparable to that of \( T_1 \) in the premixed combustion cases considered here.
Figure 7. Profiles of the mean contributions of the terms on the right-hand side of the SDR transport equation (i.e., $\{D_1, T_1, T_2, T_3, -D_2\} \times \rho \partial u / \rho_0 u_{\star}^2$) conditioned upon $c$ for different definitions of reaction progress variable $c$ for premixed combustion cases with initial $u'/S_L = 4.0$ (1st row) and 8.0 (2nd row).

Figure 8. Profiles of the mean contributions of the terms on the right-hand side of the SDR transport equation (i.e., $\{D_1, T_1, T_2, T_3, -D_2\} \times \rho \partial u / \rho_0 u_{\star}^2$) conditioned upon $c$ for different definitions of reaction progress variable $c$ for passive scalar mixing cases with initial $u'/S_L = 4.0$ (1st row) and 8.0 (2nd row).

The mean contribution of the reaction rate contribution $T_3$ assumes positive values for a major part of the flame before assuming negative values towards the burned gas side. This term is the leading order contributor in the conventional premixed combustion
cases (see Figure 8), whereas this term is identically zero for passive scalar mixing. In the MILD combustion cases, the magnitude of the mean value of $T_3$ remains negligible in comparison to that of $T_2$ and $(-D_2)$. It can be seen from Figures 6–8 that the magnitudes of the mean values of the terms of $\{D_1, T_1, T_2, T_3, -D_2\} \times \frac{l_0^2}{\rho_{old} u_0^2}$ increase with increasing turbulence intensity.

The aforementioned trends of the different terms of the SDR transport equation remain unchanged for different definitions of reaction progress variable $c$; however, the magnitudes of the mean values of the terms for the CH$_4$ mass fraction-based reaction progress variable remain greater than other options of reaction progress variables, whereas the values for O$_2$ and H$_2$O mass fraction-based reaction progress variables remain comparable and relatively smaller values are obtained for CO$_2$ mass fraction-based reaction progress variable. This is also consistent with the relative magnitudes of the SDR based on different definitions of the reaction progress variable observed in Figure 5a. Moreover, the transitions from positive to negative values for the distributions of the mean values of $D_1$ and $T_3$ across the flame front are dependent on the choice of $c$ for both MILD and premixed flame cases and these differences originate due to different distributions of species and their reaction rates within the flame front. For the MILD combustion cases, the magnitudes of the mean values of the terms of the SDR transport equation decrease with decreasing O$_2$ concentration. The reductions of the magnitudes of the source and sink terms of the SDR transport equation are consistent with the decreasing trends of the SDR with decreasing O$_2$ concentration, and turbulence intensity (see Figure 5).

The major differences in behaviours of $T_1$ and $T_2$ between MILD combustion and premixed flame cases can be explained in the following manner. The term $T_1$ originates due to density variation caused by temperature changes. In premixed flames, the gas density decreases from the unburned gas side to the burned gas side due to temperature rise, whereas density does not change appreciably across the flame in the case of MILD combustion. This can be substantiated from Figure 9 where the mean gas density $\rho$ conditioned upon $c$ for both homogeneous MILD combustion with 3.5% O$_2$ and conditional premixed combustion corresponding to initial $u'/S_L = 4.0$ are exemplarily shown. Similar qualitative behaviour has been observed for other turbulence intensities and O$_2$ dilution levels and thus is not shown for the sake of brevity.

The differences in behaviour in terms of density variation between MILD and premixed combustion cases also affect the statistical behaviour of $T_2$. The term $T_2$ can be expressed as [33–35]:

$$T_2 = -2\rho N_c \left( e_a \cos^2 \theta_A + e_{\beta} \cos^2 \theta_\beta + e_{\gamma} \cos^2 \theta_\gamma \right)$$  \hspace{1cm} (7)
where $e_\alpha, e_\beta$ and $e_\gamma$ are the most extensive, intermediate and the most compressive principal strain rates, respectively, and $\theta_\alpha, \theta_\beta$ and $\theta_\gamma$ are the angles between $\nabla c$ and the eigenvectors associated with $e_\alpha, e_\beta$ and $e_\gamma$, respectively. Equation (7) suggests that a preferential collinear alignment between $\nabla c$ and the most extensive eigendirection (i.e., $\cos^2 \theta_\alpha = 1.0$) leads to a negative value of $T_2$. By contrast, a preferential collinear alignment between $\nabla c$ and the most compressive eigendirection (i.e., $\cos^2 \theta_\gamma = 1.0$) leads to a positive value of $T_2$. It was demonstrated elsewhere [33–35] that $\nabla c$ preferentially aligns with the most compressive eigendirection (i.e., $\cos^2 \theta_\gamma = 1.0$) when turbulent straining dominates over the strain rate induced by thermal expansion as a result of density change. Thus, in the case of passive scalar mixing, $\nabla c$ aligns with the most compressive eigendirection [33–35]. In contrast, a preferential alignment between $\nabla c$ and the most extensive eigendirection (i.e., $\cos^2 \theta_\alpha = 1.0$) is obtained when the strain rate induced by thermal expansion overwhelms underlying turbulent straining.

It was shown based on scaling arguments by Chakraborty and Swaminathan [33,34] that a preferential alignment of $\nabla c$ with the eigenvector associated with $e_\alpha$ ($e_\gamma$) is obtained when $\tau Da \gg 1$ ($\tau Da \ll 1$) where $\tau = (T_{ad} - T_0)/T_0$ is the heat release parameter. The values of $\tau$ in the MILD combustion cases are 0.22 and 0.16 for the 4.8% and 3.5% $O_2$, respectively, whereas it assumes a value of 5.69 for the premixed case. The value of $\tau$ is negligibly small and $Da < 1$ in MILD combustion cases, and, thus, $\tau Da \ll 1$ is obtained for these cases, whereas $\tau Da$ remains greater than unity for the premixed combustion case considered here. Accordingly, $\nabla c$ preferentially aligns with the eigenvector associated with $e_\gamma$ for MILD combustion cases, whereas in premixed flame cases a preferential alignment of $\nabla c$ with the eigenvector associated with $e_\alpha$ obtained. This can be substantiated from Figure 10 where the probability density functions (PDFs) of $|\cos \theta_\alpha|$, $|\cos \theta_\beta|$ and $|\cos \theta_\gamma|$ are shown for different values of $c$ for CH$_4$ mass fraction-based reaction progress variable and a qualitatively similar trend is observed for other definitions of reaction progress variable. It can be seen from Figure 10 that the probability of finding $|\cos \theta_\gamma| = 1.0$ is predominant for the MILD combustion and passive scalar mixing cases, whereas the probability of $|\cos \theta_\alpha| = 1.0$ is predominantly obtained for premixed combustion cases.

The foregoing discussion suggests that the statistical behaviours of the SDR and its transport for homogeneous mixture MILD combustion are qualitatively different from that in conventional premixed combustion. However, the statistical behaviours of the SDR and its transport in turbulent MILD combustion are qualitatively similar to that of passive scalar mixing where the leading order balance is maintained between $T_2$ and $(−D_2)$. By contrast, $T_1$ and $T_3$ play significant roles in addition to $T_2$ and $(−D_2)$ in the SDR transport in premixed flames and the qualitative behaviour of $T_2$ in premixed flames can be markedly different from the MILD combustion cases.

This suggests that the chemical time scale associated with density change and strain rate induced by thermal expansion plays a key role in the SDR behaviour in premixed turbulent flames, but the turbulent mixing timescale is the key timescale in determining the SDR statistics in homogeneous mixture MILD combustion. Thus, the linear relaxation based SDR closure [1,2] (i.e., $\ddot{N}_c \propto \ddot{\epsilon} c^{n} / k$ with $\ddot{N}_c = \bar{p} N_c / \bar{p} k$ and $\ddot{\epsilon}$ being the Favre-averaged SDR, turbulent kinetic energy and its dissipation rate, respectively, and $c^{n} = c - \bar{c} / \bar{p}$ is the Favre-fluctuation of reaction progress variable with the overbar suggesting a Reynolds averaging operation), which was originally proposed for passive scalar mixing and considers only turbulent timescale, has the potential to be applicable for turbulent MILD combustion. The closures of Favre-averaged/filtered SDR for premixed combustion were discussed elsewhere [2], and the SDR closure for MILD combustion is beyond the scope of this analysis but will be considered in the future.
Figure 10. PDFs of $|\cos \theta_\alpha|$, $|\cos \theta_\beta|$, and $|\cos \theta_\gamma|$ (1st to 3rd column) for (1st and 2nd rows) MILD combustion (4.8% solid line, 3.5% dashed line), (3rd and 4th rows) premixed combustion and (5th and 6th rows) passive scalar mixing cases with initial $u'/S_L = 4.0$ (1st, 3rd, 5th row) and 8.0 (2nd, 4th and 6th row).
5. Conclusions

The statistical behaviors of the scalar dissipation rate (SDR) and its transport for homogeneous mixture turbulent MILD combustion have been analysed for different O\textsubscript{2} dilution levels and turbulence intensities for different choices of reaction progress variable using DNS data. Additional DNS data for turbulent premixed flames and passive scalar mixing are also considered for comparing the SDR statistics of the homogeneous mixture MILD combustion with conventional premixed combustion and passive scalar mixing. The high values of SDR are obtained throughout the combustion volume in the MILD combustion cases, whereas the high SDR values are obtained in a small part of the combustion volume within the reaction zone in premixed flames. The SDR magnitudes are smaller in MILD combustion cases than in conventional premixed flame cases. However, the SDR magnitude drops with decreasing O\textsubscript{2} concentration in MILD combustion cases. For both MILD and conventional premixed combustion, the SDR increases with increasing turbulence intensity. The distribution of the mean value of the SDR conditioned upon the reaction progress variable is sensitive to the choice of the reaction progress variable definition. Higher magnitudes of SDR are obtained for CH\textsubscript{4} and O\textsubscript{2} mass fraction-based reaction progress variable than in the case of reaction progress variable definitions based on CO\textsubscript{2} and H\textsubscript{2}O mass fractions. This difference in the SDR behaviour arises due to the differences in the distributions of the aforementioned species within the flame. It has been found that the strain rate contribution and the molecular dissipation term remain the leading order contributors in the SDR transport for MILD combustion, whereas in conventional premixed flames the terms rising from density variation and reaction rate gradient also play leading roles in addition to the strain rate and molecular dissipation contributions. By contrast, the SDR equation terms due to density gradient and reaction rate gradient remain negligible in comparison to the leading order contributors in MILD combustion cases due to small density variation as a result of moderate temperature rise and small reaction rate gradient magnitudes. Furthermore, the qualitative behaviour of the strain rate contribution to the SDR transport in premixed flames is significantly different to that in the case of MILD combustion, and this term acts as a leading order source and sink, respectively, for the MILD and premixed combustion cases considered here. This difference originates due to the differences in the alignment of the scalar gradient to local strain rate eigenvectors, which in turn arises as a result of the differences in thermal expansion effects between MILD and conventional and premixed combustion cases. The profiles of the mean value of the reaction rate contribution to the SDR transport conditioned upon reaction progress variable have been found to be sensitive to the choice of the reaction progress variable definition due to the differences in reaction rate distribution for different species within the flame front. The findings of the current analysis indicate that the SDR transport in MILD combustion shows several qualitative similarities to the passive scalar mixing despite major differences with the SDR transport in conventional turbulent premixed flames. This further suggests that the linear relaxation based SDR closure, which considers only the turbulent timescale and was originally proposed in the context of passive scalar mixing [1,2], has the potential to be applicable for MILD combustion but the SDR models for the premixed turbulent combustion may not be applicable for MILD combustion of homogeneous mixtures. Therefore, the findings of the current analysis provide the platform for future analyses for the development of high-fidelity models of Favre-averaged/filtered SDR for MILD combustion processes.

It is also worth noting that the effects of mean shear are absent in the canonical configuration considered here. However, the physical mechanisms, which determine the evolution of the scalar dissipation rate, occur at small scales that do not depend on the large-scale forcing and the small-scale behaviour shows the attributes of homogeneous isotropic turbulence. It was indeed found in the past that the reactive scalar statistics for turbulent premixed flames obtained from DNS of laboratory-scale configuration [36] have been found to be qualitatively consistent with the corresponding findings in the canonical
configuration without mean shear [37,38]. However, further analyses will be necessary in configurations with mean shear to confirm the findings of the present analysis.

Finally, it is worth noting that it is not possible to obtain direct experimental validations of the findings presented in this paper because the measurements of SDR and the terms of its transport equation in three dimensions remain difficult. However, the SDR models for premixed flames, which were proposed based on DNS data [39–42], have been found to work well also for RANS [43–46] and LES [47–50] of experimental configurations. This can be considered as indirect evidence of the appropriateness of scalar dissipation rate statistics analysis using DNS data. Despite this, further experimental and computational analyses will be necessary for validation of the findings of this paper.


Funding: EPSRC grants (EP/S025154/1 and EP/R029369/1).

Data Availability Statement: The data that support the findings of this study are available from the corresponding author upon reasonable request.

Acknowledgments: The financial support of EPSRC (EP/S025154/1) and the computational support of Cirrus, Rocket and ARCHER (EP/R029369/1) are gratefully acknowledged.

Conflicts of Interest: The authors declare no conflict of interest.

References

10. Awad, H.S.; Abo-Amsha, K.; Ahmed, U.; Chakraborty, N. Comparison of the reactive scalar gradient evolution between homogeneous MILD combustion and premixed turbulent flames. Energies 2021, 14, 7677. [CrossRef]
Energies 2022, 15, 9188


42. Gao, Y.; Chakraborty, N.; Swaminathan, N. Dynamic scalar dissipation rate closure for Large Eddy Simulations of turbulent premixed combustion: A Direct Numerical Simulations analysis. Flow Turb. Combust. 2015, 95, 775–802. [CrossRef]

43. Kolla, H.; Swaminathan, N. Strained flamelets for turbulent premixed flames II: Laboratory flame results. Combust. Flame 2010, 157, 1274–1289. [CrossRef]


