18th Australasian Fluid Mechanics Conference
Launceston, Australia
3-7 December 2012

Differential diffusion of passive scalars with MMC mixing model in isotropic turbulent flow

L. Dialameh [1], B. Sundaram[1], M. J. Cleary[2] and A. Y. Klimenko [1]
[1]School of Mechanical and Mining Engineering
The University of Queensland, Queensland 4072, Australia
[2]School of Aerospace, Mechanical and Mechatronic Engineering
University of Sydney, Sydney, New South Wales 2006, Australia

Abstract

In theoretical models of turbulent mixing it is commonly assumed that the molecular diffusivities of all species are equal or that molecular diffusion is small compared with turbulent diffusion and that the effect of ignoring differences in the former is minor. The evidence suggests, however, that there are many situations where this simplification is not applicable. In this work the differential diffusion effects of passive scalars in statistically-stationary, isotropic turbulent flow are considered along with a modification to the multiple mapping conditioning (MMC) mixing model to account for these effects. Starting from identical initial conditions, each passive scalar field becomes self –similar with its variance decaying exponentially in time. The decay of the correlation coefficient between two mixing scalars with different diffusivities is then investigated and the results are compared with observations from previous DNS data available in the literature.

I. Introduction

In recent years the development of turbulent combustion models has mainly focused on the correct modelling of the chemistry-turbulence interactions. The molecular diffusivities of heat and mass of inert or reacting chemical species involved in turbulent flames are usually different, so that each scalar diffuses at a different rate. For example in hydrogen-air flames, which are the subject of many experiments, hydrogen diffuses about four times faster than air. Consequently, it is important to study differential diffusion; the phenomenon in which different scalars with different molecular diffusivities evolve differently from each other. Despite this, most theoretical models of turbulent flames effectively ignore differential diffusion, by assuming the same molecular diffusivities of all species and of heat (the unity Lewis number assumption) and neglecting the effects of molecular diffusion compared to turbulent diffusion. These assumptions are attractive because they lead to great modelling simplifications but as discussed below there is ample experimental and numerical evidence that unequal molecular diffusion often influences species compositions.

Drake et al. [5] reported experimentally observed differential diffusion in hydrogen-air flames and their measurements are consistent with the assumption that differential diffusion effects become smaller with increased turbulence at higher Reynolds numbers. Meier et al. [13] found quite significant differential diffusion effects on the temperature field for different H2/N2-air jet flames in the Reynolds number range between 6,200 and 8,800. Another experimental investigation of an H2/CO2 flame by Masri et al. [11] indicated that differential diffusion effects are still present for Re as high as 60,000 albeit with minor importance. A series of laser Rayleigh-scattering experiments has been performed by Dibble [4] to investigate the effects of differential molecular diffusion in turbulent non-reacting jet flows. A turbulent jet of a mixture of Freon and H2 exiting into coflowing air was studied at various Reynolds numbers. Smith et al. [17] observed large scale effects of differential diffusion in C02 diluted hydrogen flames at Re=30,000 and in another study a direct numerical simulation (DNS) was conducted [18] to examine the effect of varying Reynolds number on the differential diffusion of passive scalars in turbulent flow. The differential diffusion of passive scalars of different molecular diffusivities was also studied by Yeung and Pope [20] who performed DNS of statistically-stationary, isotropic turbulence. Nilsen and Kosaly [14] reported on a DNS investigation of differentially diffusing reacting scalars in isotropic, decaying turbulence. Their results demonstrate that effects due to differential diffusion decrease with increasing Reynolds numbers and increase with increasing Damköhler numbers.

The above studies show that differential diffusion can be significant, especially for hydrogen containing mixtures. Therefore it is important to develop scalar mixing models which account for differential diffusion effects. Although the dominant paradigm is to model all diffusivities as being equal, some such differential diffusion models have been suggested in the context of probability density function (PDF) methods by Chen and Chang [1], conditional moment closure (CMC) methods by Kronenburg and Bilger [9,10] and laminar flamelet methods by Pitsch and Peters [15]. In large eddy simulation (LES) the turbulent (or subgrid) diffusion can be of the same order as molecular diffusion and it becomes harder to justify the equal diffusivity approximation. This is particularly so for the increasingly well resolved LES of later years. In this light, McDermott and Pope [12] presented a new approach for treating molecular diffusion in LES-FDF (filtered density function) methods. The effect of differential diffusion on spatial transport is simulated by evolving scalar values in a way that accounts for effective diffusional velocities. Transport of scalars with different diffusion coefficients is simulated by the same set of particles, which is important for computational efficiency. More recently Richardson and Chen [16] proposed a new approach for treating differential diffusion in turbulent premixed flames which ensures realizability for pairwise-exchange mixing models in general, and demonstrated its application in the Interaction by Exchange with the Mean (IEM) model and in the Euclidean Minimum Spanning Tree (EMST) model.

In principle, differential diffusion may cause differences in a) spatial transport and in b) joint distributions of scalars at a fixed location. In this work, we focus on the second problem and suggest a new approach for considering the differential diffusion effects in PDF methods by using effective conditioning of mixing (i.e. MMC). As differential diffusion occurs at the molecular scale, accurate modelling of such phenomena in statistical methods is a challenging task. In order to minimise the effect of

spatial transport and examine one-point joint characteristics with maximal accuracy, we consider differential diffusion of unreactive passive scalars in statistically-stationary, isotropic turbulent flow. We demonstrate the ability of the MMC mixing to account for refined properties of differential diffusion (such a loss of correlation between the scalars). Our modelling results are compared to the DNS performed by Yeung and Pope [20].
The remainder of this paper is organized as follows. A brief overview of the MMC mixing model and the suggested modification for considering the effects of differential diffusion are presented in Sec. II. Results including correlation coefficients and decay of variances and covariances are presented in Sec. III. In Sec. IV conclusions are drawn.

II. MMC mixing model and suggested modifications for differential diffusion

The MMC approach was originally suggested by Klimenko and Pope [6] as an effective combination of CMC, which is used for evaluation of reactive scalars, and generalised mapping closure (MC), which is used for consistent modelling of the conditional dissipation and PDFs of the conditioning variables. The MMC approach allows for a stochastic implementation that converts this model into a full PDF model with CMC-like properties. In stochastic form MMC fulfils the role of a mixing model which enforces localness within a reduced manifold consisting of reference variables that are related to the physical quantities in turbulent combustion. In the MMC mixing model mixing particles interact directly with each other, similarly to Curl's model [3], but the particle pairs are selected specifically to enforce localness in the reference space rather than randomly as is the case with inferior, non-local mixing models. The specific details of the particle pair selection and the associated model for mixing time scale are discussed by Cleary and Klimenko [2]. This implementation was originally developed for the equal diffusivity condition. The MMC mixing model can be modified to account for differential diffusivity as discussed in the next paragraph.

Consider two passive scalars Y_{I} and Y_{II} in an homogeneous, isotropic turbulent field each having a different molecular diffusivity denoted by D_{I} and D_{II} respectively. As mixing proceeds, the ensembles mean scalar variances $\left\langle Y_{\mathrm{I}}^2 \right\rangle$ and $\left\langle Y_{\mathrm{II}}^2 \right\rangle$ decay with time according to:

$$\frac{\partial \left\langle Y_{\mathrm{I}}^2 \right\rangle}{\partial t} = -2D_{\mathrm{I}} \left\langle \left(\frac{\partial Y_{\mathrm{I}}}{\partial x_i} \right)^2 \right\rangle = -\chi_{\mathrm{I}} \tag{1}$$

$$\frac{\partial \left\langle Y_{\mathrm{II}}^2 \right\rangle}{\partial t} = -2D_{\mathrm{II}} \left\langle \left(\frac{\partial Y_{\mathrm{II}}}{\partial x_i} \right)^2 \right\rangle = -\chi_{\mathrm{II}} \tag{2}$$

Where χ_{I} and χ_{II} are the scalar dissipation rates. The most important joint statistic is the covariance, $\left\langle Y_{\mathrm{I}} Y_{\mathrm{II}} \right\rangle$, which evolves as

$$\frac{\partial \left\langle Y_{\mathrm{I}} Y_{\mathrm{II}} \right\rangle}{\partial t} = -2\left(D_{\mathrm{I}} + D_{\mathrm{II}}\right) \left\langle \frac{\partial Y_{\mathrm{I}}}{\partial x_i} \frac{\partial Y_{\mathrm{II}}}{\partial x_i} \right\rangle = -\chi_{\mathrm{I,II}} \tag{3}$$

Where $\chi_{\mathrm{I,II}}$ denotes the joint dissipation. The cross-correlation Coefficient $\rho_{\mathrm{I,II}}$ between the two scalars is defined as:

$$\rho_{\mathrm{I,II}} = \frac{\left\langle Y_{\mathrm{I}} Y_{\mathrm{II}} \right\rangle}{\left[\left\langle Y_{\mathrm{I}}^2 \right\rangle \left\langle Y_{\mathrm{II}}^2 \right\rangle \right]^{1/2}}. \tag{4}$$

For two scalars with identical initial values and equal diffusivities the scalars would remain identical valued, and thus fully correlated, at all times. However, since the diffusivities are not equal the scalars become progressively displaced and statistically decorrelated from each other. The manner in which the decorrelation process occurs provides fundamental information on the basic mechanisms of differential diffusion.

In MMC (as in other PDF methods) the turbulent scalar fields, whose mean and covariance evolves according to "equation 1" through "equation 3", are modelled using an ensemble of Pope particles (notional particles which possess scalar quantities subject to a mixing operation). In the absence of spatial advection and diffusion, the scalars Y_{I} and Y_{II} evolve according to the mixing operation

$$\begin{aligned} Y_{\mathrm{I}}'^{*} &= Y_{\mathrm{I}}^{*}\left(1-\alpha\right) + \hat{Y}_{\mathrm{I,II}}^{*}\alpha \\ Y_{\mathrm{II}}'^{*} &= Y_{\mathrm{II}}^{*}\left(1-\alpha\right) + \hat{Y}_{\mathrm{I,II}}^{*}\alpha \end{aligned} \tag{5}$$

The mixing model in "equation 5" is a particle interaction model. Particles are mixed in pairs and the acute symbol indicates new values, $\hat{Y}_{\mathrm{I,II}}^{*}$ is the average of scalar values prior to mixing and α is the mixing extent which is a random number that is independent of the particle scalar values ,$\alpha = 1$ corresponds to complete mixing while $\alpha = 0$ corresponds to no mixing at all.
Defining $\beta=1-\alpha$, $\gamma=1-\beta^2$ and Δt the duration of the mixing step, "equation 6" indicates a link between the mean value of γ and the unconditional dissipation time of the flow, τ_D ,which is defined as the average value of scalar variances over the dissipation rate [8]:

$$\tau_D = \frac{4\Delta t}{\left\langle \gamma \right\rangle} \tag{6}$$

The mixing operation ideally satisfies a number of principles [19] the most important of these in the current context being decay of variance consistent with "equation 1" and "equation 2" to a Gaussian distribution (in homogeneous turbulence), linearity and independence of mixing, and localness in composition space. Traditional mixing models such as Curl's model [3] satisfy linearity and independence and can approximate decay to a Gaussian but violate the localness requirement leading to significant over prediction of conditional fluctuations of reactive scalars in jet flames [2]. High quality mixing models like MMC, on the other hand, satisfy all of these principles. The principle of localness is enforced in MMC through use of reference variables which are modelled to emulate the major statistics of scalar fields but are mathematically independent of them. The particle mixing pairs are selected so that the pair is in close proximity in the reference space. Here we introduce two reference variables, Z_{I} and Z_{II}, which are modelled by independent Ornstein-Uhlenbeck (OU) processes. These are stationary, Gaussian and Markovian diffusion process [6]. (Note that other types of reference variables including Lagrangian quantities from DNS or LES are possible [2].) The stochastic differential equation for $Z^*(t)$ (the value of Z_{I} and Z_{II} are MMC reference quantities also carried by the Pope particles) has the following form:

$$dZ_I^* = -\frac{Z_I^*(t)}{\tau_I}dt + \left(\frac{2}{\tau_I}\right)^{1/2} dW(t)$$

$$dZ_{II}^* = -\frac{Z_{II}^*(t)}{\tau_{II}}dt + \left(\frac{2}{\tau_{II}}\right)^{1/2} dW(t) \qquad (7)$$

Where W(t) is a Weiner process (random walk) and τ_I and τ_{II} are selected so that the statistics of Z*(t) match (as much as possible) the statistics of real turbulence. Thus we have two time scale ratios:

$$r_1 = \frac{\tau_I}{\tau_D} \qquad (8)$$

$$r_2 = \frac{\tau_{II}}{\tau_I} \qquad (9)$$

For the scalar Y_I mixing is localised in an extended space given by (Z_I, μZ_{II}), while for scalar Y_{II} mixing is localised in an extended space given by (μZ_I, Z_{II}). μ is a weighting factor that is required to control the rate of decorrelation of the differentially diffusing scalars. Its value is typically small. In previous MMC modelling, where the equal diffusivity approximation has been made, the reference space has been the same for all scalars. This new idea of using different reference spaces, with different characteristic diffusion times, allows the differential diffusivity of Y_I and Y_{II} to be modelled implicitly by the mixing model. The benefit of this approach (particularly for practical inhomogeneous flows) is that a single set of Pope particles can be used to model differentially diffusing scalars.

In non-local mixing models, like Curl's model, the decay of scalar variances is strongly determined by the mixing time scale τ_D in "equation 6". In MMC, on the other hand, the localisation of mixing in the reference space directly links the dissipation of the variances of scalars Y_I and Y_{II} to the dissipation of the variances of reference variables Z_I and Z_{II}. The latter are controlled by the parameters r_1 and r_2 while τ_D has a much reduced level of influence and controls only the so called *minor fluctuations* of Y with respect to Z. In the present work we set an equal α for both Y_I and Y_{II}.

The model has three independent parameters: r_1, r_2 and μ. The first two are selected to give the correct rate of decay of $\langle Y_I^2 \rangle$ and $\langle Y_{II}^2 \rangle$. μ is selected to ensure the correct rate of decorrelation of the differentially diffusing scalars. For $\mu = 0$ the cross-correlation coefficient, $\rho_{\mathrm{I,II}}$, approaches zero very quickly, whereas for $\mu = 1$, $\rho_{\mathrm{I,II}} = 1$ and Y_I and Y_{II} remain perfectly correlated and do not differentially diffuse. The selection of the three parameters is demonstrated in the next section.

III. Results

The modified MMC model is applied to the case of differential diffusion of two passive scalars in statistically-stationary isotropic turbulent flow. The modelling is performed using 10,000 Pope particles which are initialised by $Y_I^* = Y_{II}^*$ and $Z_I^* = Z_{II}^*$. Results are compared to the DNS performed by Yeung and Pope [20].

"Figure 1" shows $\langle Y_{\mathrm{I}}^2 \rangle$ and $\langle Y_{\mathrm{II}}^2 \rangle$ versus time. As can be seen the dissipation of scalar fluctuations by mixing occurs at different rates for two scalars due to their different diffusivities. The results show exponential decay of the variances in time, represented by approximately straight lines of constant slopes on the linear-log plot. A reasonable match between the MMC predicted decay rate and the DNS data is obtained by setting the time scale ratios $r_1 = 8$ and $r_2 = 1.3$. The figure also includes the MMC predicted covariance $\langle Y_{\mathrm{I}} Y_{\mathrm{II}} \rangle$.

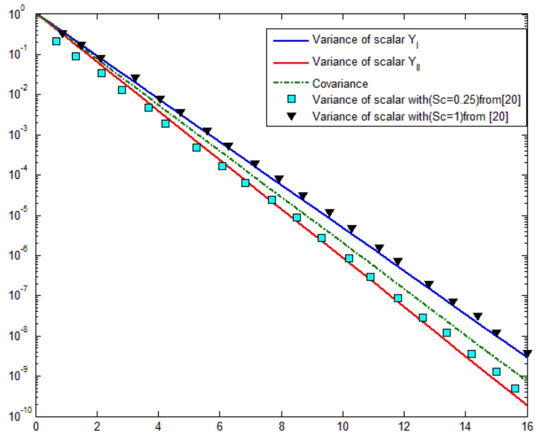

Figure 1.Scalar variances and covariance versus time.

The evolution of the particle scalar values Y_I^* and Y_{II}^* over time is illustrated in "figure 2 "showing a scatter plot of Y_I^* versus Y_{II}^* at approximately 16 eddy turn-over times. The two scalars are initially equal (red line with slope of unity) but due to differential diffusivity the two scalars slowly decorrelate (green line with slope less than unity).

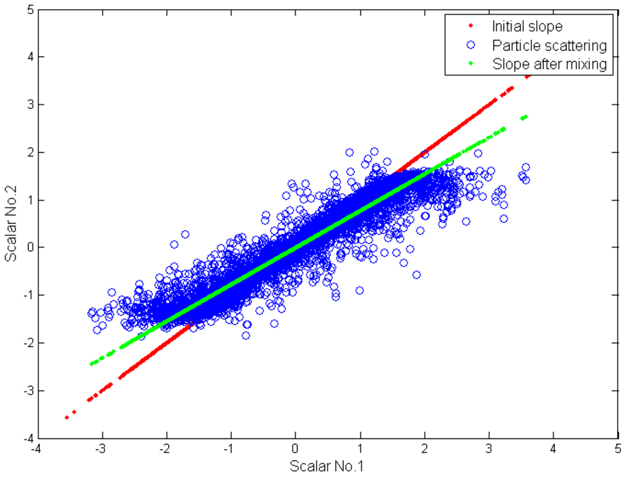

Figure 2.Particle scatter plot of Y_I^* versus Y_{II}^* at 5 eddy turn-over times.

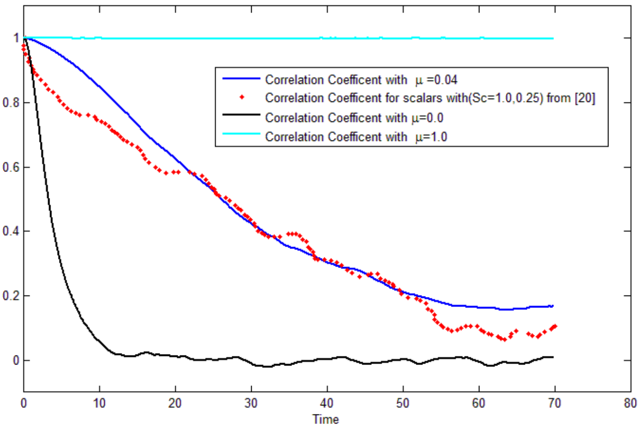

Figure 3.Correlation coefficient versus time

The decorrelation of the differentially diffusing scalars is now evaluated quantitatively. “Figure 3” shows the correlation coefficient versus time. MMC predictions are shown for three different values of μ. As expected $\mu = 1$ results in full correlation at all times (this is equivalent equal species diffusivity). It is evident that $\mu = 0$ leads to too rapid decorrelatoin within about 10 eddy turn-over times whereas the true time to decorrelation is an order of magnitude greater. The relatively good match between the MMC and DNS is obtained by setting $\mu = 0.04$.

IV. Conclusions

A modified form of the MMC mixing model is developed to account for the effects of differential diffusion. The new model is examined for the case of differential diffusion of two passive scalars in statistically stationary, isotropic turbulent flow in comparison with the DNS results of Yeung and Pope [20]. Specifically, we demonstrate the ability of the model to predict the different rates of decay of the scalar variances and at the same time also being able to control and emulate their rate of decorrelation. While the purpose of this paper is to demonstrate principal feasibility of consistent emulation of variance decay and decorreletion due to differential diffusion within the flamework of PDF methods, the future work will investigate dependence of modelling parameters on Le and Re and focus on giving practical recommendations for modelling of differential diffusion.

Acknowledgments

This research is financially supported by the Australian Research Council under grant number DP120102294.

References

[1] Chen, J.Y. & Chang, W.C., Modelling differential diffusion effects in turbulent nonreacting-reacting Jets with stochastic mixing models, Combust. Sci. and Tech., 133,343-375, 1998.

[2] Cleary, M.J. & Klimenko, A.Y., A detailed quantitative analysis of sparse-Lagrangian filtered density function simulations in constant and variable density reacting jet flows, *Phys. Fluids ,*23, 115102, 2011.

[3] Curl, R.L., Dispersed phase mixing: 1. theory and effects in simple reactors, *Aiche Journal,* 9, 2, 175–181,1963.

[4] Dibble, R. W.& Long ,M.B., Investigation of differential diffusion in turbulent jet flows using planar laser Rayleigh scattering, *Combustion and Flame* ,143 ,2005, 644–649

[5] Drake, M. C. , Lapp, M. , Penney, C. M. , Warshaw, S. & Gerhold, B. W., Measurements of temperature and concentration fluctuations in turbulent diffusion flames using pulped Raman spectroscopy, *18th Symposium (International)on Combustion ,The Combustion Institute, Pittsburgh,* 1521–1531, 1981.

[6] Karlin, S., & Taylor, H.M., *A Second Course in Stochastic Processes*, Academic Press, New York, 1981.

[7] Klimenko, A.Y. & Pope, S.B., The modelling of turbulent reactive flows based on multiple mapping conditioning, *Phys. Fluids*, 15, 2003.

[8] Klimenko, A.Y., Matching conditional moments in PDF modelling of nonpremixed combustion, *Combustion and Flame,* 143, 369–385, 2005.

[9] Kronenburg, A., & Bilger, R. W., Modelling of differential diffusion effects in nonpremixed nonreacting turbulent flow, *Phys. Fluids*, 9, 1435-1447, 1997.

[10] Kronenburg, A., & Bilger, R. W., Modelling Differential Diffusion in Nonpremixed Reacting Turbulent Flow: Model Development, *Cmbust. Sci. and Tech.,* 166, 195-227, 2001.

[11] Masri, A.R., Dibble, R.W. & Barlow, R.S. ,Chemical kinetic effects in nonpremixed flames of H_2/Co_2 fuel, *Combustion and Flame*, 91, 285, 1992.

[12] McDermott,R.& Pope, S.B., A particle formulation for treating differential diffusion in filtered density function methods, *Journal of Computational Physics,* 226 ,947–993, 2007.

[13] Meier. W., Vyodorov, A.O., Bergmann, V. & Stricker, W. ,Simultaneous Raman/LIF measurements of major species and NO in turbulent H2-air diffusion flames, *Appl. Phys*., B 63,79, 1996.

[14] Nilsen, V. & Kosaly, G, Differential diffusion in turbulent reacting flows, *Combustion and Flame*, 117, 493, 1999.

[15] Pitsch, H. & Peters. N., A consistent flamelet formulation for nonpremixed combustion considering differential diffusion effects, *Combustion and Flame*, 114, 26,1998.

[16] Richardson ,E.S.,& Chen, J.H., Application of PDF mixing models to premixed flames with differential diffusion, *Combustion and Flame* ,159 , 2398–2414, 2012.

[17] Smith, L.L., Dibble. R.W., Talbot, L., Barlow, R.S. & Caner, C.D. , Laser Raman scattering measurements of differential diffusion in nonreacting and reacting laminar and turbulent jet flow, *31st Aerospace Science Meeting & Exhibit, AIAA*-93-0804,1993.

[18] Smith.N.S.A, Differential diffusion of passive scalars in homogenous turbulence: implications for combustion modelling, *Australian Symposium on Combustion*, 61-65, 1999.

[19] Subramaniam, S., Pope, S.B., A Mixing Model for Turbulent Reactive Flows based on Euclidean Minimum Spanning Trees, *Combustion and Flame*, 115, 4, 487-514, 1998.

[20] Yeung, P.K. & Pope,S.B., Differential diffusion of passive scalar in isotropic turbulence, *Phys. Fluids,* 5, 2467-2478 , 1993.