Conditional moment closure for turbulent combustion

A.Y. Klimenko^a, R.W. Bilger^b

^aDepartment of Mechanical Engineering, University of Queensland, Brisbane, Qld 4072, Australia ^bDepartment of Mechanical and Mechatronic Engineering, University of Sydney, NSW, 2006, Australia

Abstract

This paper reviews the fundamentals of conditional moment closure (CMC) methods for the prediction of turbulent reacting flows, with particular emphasis on combustion. It also surveys several of the applications that have been made. CMC methods predict the conditional averages and higher moments of quantities such as species mass fractions and enthalpy, conditional on the mixture fraction or reaction progress variable having a particular value. A brief introduction is given to generalized functions and probability density function (pdf) methods. This is followed by an exposition on the various methods of derivation for the CMC equation and the general characteristics of this equation and its boundary conditions. Simplifications that can be made in slender layer flows such as jets and plumes are outlined and examples of application of the technique to such flows are given. The method allows the definition of a new class of simplified reactors related to the well known perfectly stirred reactor and plug flow reactor: these are outlined. CMC predictions are compared to experiment and direct numerical simulations for flows with homogeneous turbulence. Derivation and modeling of the equations for conditional variances and covariances are outlined and their use in second-order CMC illustrated. Brief review is made of progress on application of the method to problems involving differential diffusion, multiple conditioning, sprays and premixed combustion.

Keywords: Turbulent combustion; Conditional averaging; Modeling; Closure methods; Pollutant formation

Contents

Nomenclature
1. Introduction
1.1. Background
1.2. Outline of conditional moment closure methods
1.3. Outline of this review
2. Pdf methods in turbulent flows
2.1. Pdfs and conditional expectations
2.1.1. Pdfs and generalized functions
2.1.2. Joint pdfs
2.1.3. Conditional pdfs and conditional expectations
2.2 Differentiating generalized functions and pdfs
2.2.1 Generalized derivative
2.2.1. Scheralized activative
2.2.2. The gradient operator from the joint odf
2.2.3. Differentiating the joint put
2.2.4. Differentiating conditional expectations
2.3.1 The organization for the fine grained ref
2.3.1. The equation for the line-grained put
2.3.2. The joint par equation
2.3.3. The case of high Reynolds numbers
2.4.1 The conserved scalar par equation and its boundary conditions
2.4.1. Smooth pdfs
2.4.2. Smooth pdfs with fixed bounds
2.4.3. Intermittent pdfs
2.5. Summary of the major results
3. The conditional moment closure equation
3.1. The joint pdf method
3.1.1. The unclosed equation
3.1.2. Alternative derivation of the unclosed equation
3.1.3. Primary closure hypothesis
3.2. Decomposition method
3.2.1. The unclosed equation
3.2.2. Primary closure hypothesis
3.3. Comparing the primary closure hypotheses
3.4. Enthalpy equation in conditional moment closure
3.5. Note on averaging of the source terms
3.6. Note on conditional Favre averaging
3.7. The physical basis of conditional moment closure
3.7.1. Diffusion approximations and the analogy with a Markov process
3.7.2. Hypothesis of local similarity
3.8. Summary of the major results
4. Main features of the conditional moment closure equation
4.1. Possible closures of the remaining unclosed terms in the general conditional moment closure equations
4.2. Conservation integrals
4.2.1. Smooth pdfs with fixed bounds
4.2.2. Intermittent pdfs
4.2.3. Errors arising from the conditional scalar dissipation
4.3. Frozen and fast chemistry limits
4.4. Laminar flamelet model
4.5. Some further results for homogeneous turbulence
4.5.1. Low probability effects
4.5.2. Boundary conditions
4.6. Overview
5. Conditional moment closure in turbulent slender laver flows
5.1. Basics of the asymptotic analysis
5.2. Simple method

5.3. Integral method
5.4. Boundary conditions for the pdf integrals
5.4.1. Intermittent pdfs
5.4.2. Smooth pdfs
5.4.3. Summary of the boundary conditions
5.5. Conservation integrals
5.5.1. Errors arising from inconsistent scalar dissipation
5.6. Evaluation of the conditional scalar dissipation
5.6.1. Results for self-similar flows
5.6.2. Results for the planar wake
5.6.3. Method for local self similarity in jet flames
5.6.4. Other methods
5.7. Corrections of higher order
5.8. Results for some conditional moment closure predictions
5.8.1. Reacting scalar mixing layer
5.8.2. Turbulent jet diffusion flames
5.8.3. Round plumes in grid turbulence
6. Conditional moment closure for simplified turbulent reactors
6.1. The incompletely stirred reactor
6.1.1. Definition and governing equations
6.1.2. Application
6.1.3. Validity of ISR modeling
6.2. The dilution flow reactor
7. Conditional moment closure in homogeneous flows
7.1. Turbulent mixing reactors
7.2. DNS with one-step chemistry
7.2.1. Correctly modeling conditional scalar dissipation
7.2.2. Validity of the primary closure hypothesis
7.2.3. Closure of the chemical source term
7.3. Multi-step kinetics
8. Conditional variance equation and second order closure
8.1. The pdf method
8.2. Decomposition method
8.3. Discussion
8.4. Second order closure
9. Conditional moment methods: further topics
9.1. Differential diffusion
9.1.1. Derivation of the unclosed equation
9.1.2. Closure for non-reacting homogeneous flow
9.1.3. Closure for reacting flow
9.2. Conditional moment closure with multiple conditions
9.2.1. The pdf method
9.2.2. The decomposition method
9.2.3. Discussion
9.3. Conditional moment closure and spray combustion
9.3.1. Basics of droplet evaporation and combustion
9.3.2. Formulation of the conditional moment closure model
9.3.3. The mixture fraction pdf and dissipation in sprays
9.3.3.1. Non-inertial droplets
9.3.3.2. Inertial droplets
9.3.3.3. Remarks for practical applications
9.4. Conditional moment closure for turbulent premixed combustion
9.5. Conditional methods and Lagrangian modeling
10. Conclusions and perspectives
Acknowledgements
References

NOTATION

Roman Letters c reaction progress variable (or number density in Sec.9.3) a function (or interdroplet mixture fraction in Sec.9.3) f h enthalpy k the reaction rate constant $k_{\rm t}$ kinetic energy of turbulence l a length scale m mass flow rate p pressure r radius or distance from a droplet t time velocity vector V x Cartesian coordinates z Differential diffusion scalar D diffusion coefficient Da Damkohler number F arbitrary smooth function $G \equiv \langle K | \eta \rangle$ conditional expectation of variance K *H* Heaviside function $K = (Y'')^2$ conditional variance or a correlation function L integral length scale $N = D(\nabla \xi)^2$ scalar dissipation $O(\varepsilon)$ indicates a function of the same order as ε P probability density function, pdf Pe Peclet number $Q \equiv \langle Y | \xi = \eta \rangle$ conditional expectation of a reactive scalar Re Reynilds number R radius Sc Schmidt number $S_{\rm c}$ reaction term for c S_{η} isopleth surface ($\xi=\eta$) area T temperature U longitudinal average velocity component W chemical reaction term X normalized xmole fraction (Fig. 27) Y scalar, reactive scalar species mass fraction Z sample space variable for Y $\mathcal{P}(\mathcal{B})$ cumulative probability of event \mathcal{B} mesh size М Greek Letters γ intermittency δ Delta-function Delta-function ϵ a small number or parameter

 ε_t turbulent dissipation of energy

v kinematic viscosity

 $[\]rho$ density

- σ dispersion
- τ characteristic time scales or a time-like variable
- $\theta = \xi^2 \langle \xi \rangle^2$ unconditional variance
- normalized temperature difference (Fig. 38)
- ξ conserved scalar or mixture fraction
- η sample space variable for ξ
- $\psi = \delta(Z Y)$ the fine-grained pdf
- ζ sample space variable for c

 $\Gamma \quad \text{Gamma-function} \\ \Phi(\lambda) \equiv \langle \exp(i\lambda Y) \rangle \text{ characteristic function of } Y$

Subscript indices

- a an averaged value
- c related to a centerline or to variable c*i,j* related to different components of vectors t related to turbulence or turbulent region *min* related to the smallest value of conserved scalar
- max related to the largest value of conserved scalar
- *m* either *min* or *max*
- in related to the in-flow
- out related to the out-flow
- b either in or out
- e related to chemical equilibrium
- s related to the stoichiometric mixture ratio or related to the surface of a droplet
- K related to the Kolmogorov viscous scales
- R related to radius R or is a reference value
- η related to ξ and η or conditioned on $\xi = \eta$

Superscript indices

- derivative of a function
- or fluctuation with respect to unconditional mean
- indicates alternative or dummy values
- " fluctuation with respect to conditional mean
- * indicate integration or averaging over certain region

Overbar symbols

- ~ Favre averaging
- --- conventional averaging
- ^ normalized function or variable

Underscore symbols

- indicates a vector (for Greek letters)

Other special symbols

- < > ensemble averaging
- $\{ \}_R$ integration over cross-section limited by $|\mathbf{r}| < R$

1. INTRODUCTION

1.1. Background

turbulent flows present problems Reacting of important economic consequence in many fields of science. In engineering we are interested in efficiency, transfer predicting heat and pollutant formation in internal combustion engines and furnaces. product in chemical yield reactors. explosions, and the rate of spread overpressures in gas and toxic emissions from fires. In environmental science we are concerned with rates transformation of chemical in chimney plumes. oxidant production in photochemical and pollutant transformation biota smog, and growth in lakes. estuaries and coastal waters. Examples rivers. are coming into focus in oceanography and atmospheric science, for example in the ozone hole problem¹¹².

The problems are made difficult because of the fact that the rates of reaction of concern are highly nonlinear functions of temperature and The turbulence in the species concentrations. flow engenders mixing of nonuniformities in species and temperature, and the rates of this mixing usually fast compared with the rates of reaction. are not As a spatial temporal fluctuations in consequence, large and occur the scalar efforts quantities (composition, temperature, enthalpy, etc). and to terms of rates of reaction in average of express average values the inadequate ¹⁷. The be nonlinearity of the scalars prove to reaction rates give rise to terms involving correlations of the fluctuations, and these are usually as large as those involving only the average quantities. Often correlation terms are of the opposite sign, that the true mean rate the SO magnitude of reaction can be an order of or more smaller than that the obtained from using just average values in rate expression. This problem is an extremely difficult addition the already difficult to closure problem of prediction in nonreacting turbulent flows.

The problems of predicting flow and mixing of nonreacting scalars are difficult enough. Progress has been made using several approaches. One of considerable engineering approach interest in and applied science involves so-called "moment closure" methods. First moments are means or averages. Second moments are variances and covariances of the fluctuations about the averages. Third moments are triple correlations between the

1-1

fluctuations. In moment methods the Navier-Stokes and scalar conservation equations are used to derive equations for these moments. The exact equations for the first moments have terms involving the second moments. Exact equations for the second moments have terms involving the third moments. And so on. The equations for any level of moments have terms involving higher level moments. The system of equations is thus unclosed. In moment closure methods, closure is obtained by modelling the higher level moments in terms of the lower moments. First moment closure (often "order" closure) solves equations for the first moments called first (the averages) by expressing the second moments such as the Reynolds stresses and scalar fluxes in terms of the averages and their gradients. Mixing eddy viscosity methods are such first moment closure methods. length and still commonly used in environmental science and the They are in sciences. In engineering, much use is made of second moment geophysical closures which the triple correlation (order) in terms are modelled in terms of the first and second moments and their gradients. While these models are by no means universally applicable they have proved to be very useful in predictions for a wide range of flows^{73,143}.

the second moment level Such success in closure at been has not achieved for chemical reacting systems, however, high due to the nonlinearity of the reaction rate terms. This difficulty appears not only for the mean reaction rate in the first moment equations for the term the also in the equations for turbulent scalar fluxes where scalars. but correlations between the reaction and the scalar fluctuations rate are present. Moment closure methods have met with success for only a limited $fast^{7,21}$ of problems where the chemistry is or range where it is sufficiently simple⁴⁶.

General reviews of the problems of predicting turbulent reacting flows may be found in Chen & Kollmann²⁷, Fox^{39} , Libby & Williams⁸¹ and Pope¹¹⁴.

1.2. Outline of CMC Methods

In the Conditional Moment Closure (CMC) methods that are the subject this review, the underlying hypothesis is that most of the fluctuation of in the scalar quantities interest can often be associated with of

fluctuation of (usually) only one key quantity. Thus in nonpremixed the where there is mixing between two bodies or streams of fluid, problems. fuel and oxidant, say, the values of concentrations and temperature within the mixing field depend very strongly on the local instantaneous value of the mixture fraction, which variable. such as describes the some stoichiometry of the mixture. The mixture fraction is a conserved scalar and can be defined as the normalised mass fraction of an inert tracer with the fuel. If turbulent mixing occurs without introduced significant diffusion. differential molecular the mixture fraction gives the mass fraction of the material in the local mixture that originated in unmixed body of fluid denoted as fuel. The mass fraction of the local mixture that originated from the unmixed body of fluid denoted as oxidant, is then (For further minus the mixture fraction. clarification of this one important concept see Refs 9, 139). The balance equation for the mixture fraction has no chemical source term. As such, it is not subject to the difficulties associated with reactive scalars. and its mean and variance fields are often predicted with success by moment closure methods.

Figure 1 shows scatter plots of temperature and mass fraction of by advanced laser diagnostic hydroxyl radical, measured methods, as a function of the simultaneously made measurement of the mixture fraction. The measurements were made⁸⁵ in a piloted nonpremixed jet flame of methanol in air. It is seen that the temperature and OH are very strong nonlinear functions of mixture fraction. At a given position in the flame are very strong fluctuations in the mixture fraction about its there mean value. There are also very strong fluctuations of temperature and OH mass fraction about their mean values. But if we draw a curve through the middle of this scatter plot data, the departures from this curve are much less than the overall fluctuations. The curve is the function derived from conditional averaging.

Conditional moments are averages, covariances, etc, made subject to a certain condition being satisfied. In Fig. 1 the right hand side (RHS) of conditional the figure shows average values of temperature, Τ, and fraction, $Y_{\rm OH}$, conditional on hydroxyl radical mass the mixture fraction, ξ , having a prescribed value, η . We write these as $\langle T | \eta \rangle$ and $\langle Y_{OH} | \eta \rangle$. A formal definition of conditional averages will be found in more Section 2.1.3. In reducing the experimental data, the range of mixture fraction is divided into something like 30 'bins'. The data set is then sorted into 30 these bins. **Statistics** are then made for each bin: the average

1-3

temperature in the bin is taken as the conditional average for the mixture centre of the range of mixture fraction fraction value at the for that conditional variance of the fluctuations is taken as the variance bin. Α the fluctuations about the conditional average for for the data in that association that is mentioned bin. The close above between reactive scalars and the mixture fraction can be more precisely expressed in terms of the conditional variances being small compared with the square of the mean. This roundabout way of expressing conditional the close correlations Fig. 1 that are apparent in is necessary since the correlations are strongly nonlinear.

In premixed systems, it is thought that fluctuations in species mass fractions and in temperature are often closely associated with fluctuations in the reaction progress variable. This reaction progress variable is best defined in terms of the H₂O mass fraction in the mixture such that it has a value of zero in the unburnt fluid and a value of unity in the fully reacted mixture. Methods for premixed systems are not well advanced as yet, and in most of what follows we will confine ourselves to nonpremixed systems.

conditional In earlier work, averaging done subject the was to condition of whether the flow was turbulent or nonturbulent. This is Chen Kollmann²⁷ outline sometimes termed zone conditional averaging. & this approach in comparison with the conditional averaging used here.

In CMC methods. equations are derived and modelled for the conditional moments reactive scalars. With the modelling of the nonpremixed assumptions usually made for simpler problems in combustion, the CMC equation for the conditional average $\langle Y_i | \eta \rangle$ of the mass fraction of species *i* is

$$\frac{\partial \langle Y_i | \eta \rangle}{\partial t} + \langle \mathbf{v} | \eta \rangle \langle \nabla \langle Y_i | \eta \rangle - \langle N | \eta \rangle \frac{\partial^2 \langle Y_i | \eta \rangle}{\partial \eta^2} = \langle W_i | \eta \rangle$$
(1.1)

In this equation t is time, v is velocity, W_i is the rate of formation of species *i* per unit mass of the mixture and N is the scalar dissipation

$$N \equiv D \nabla \xi \cdot \nabla \xi$$

where D is the molecular diffusivity, assumed equal for all species. In deriving this equation, important modelling assumptions are made and these

discussed in some detail later. A similar equation is derived for the are These conditional average enthalpy. equations have unclosed source terms involving the conditional average of the species reaction rate and of the CMC methods usually make the assumption radiant transfer. that net closure for these terms can be made at the first conditional moment level. This is often possible for the conditional reaction rates since the fluctuations about the conditional averages are small and the conditional covariances can be neglected; but for some cases the closure is best done at the second moment level. It should be noted that the CMC equation, itself. is valid for large fluctuations about the conditional mean (conditional variance). Relatively small values of the conditional shown Figure 1 indicate conditional variances on that the expectations represent a good choice for the model property, however, variables. This is not assumed in CMC derivations. The physical basis of CMC lies on different grounds which are discussed in Section 3. The modelling of the conditional average velocity, $\langle \mathbf{v} | \eta \rangle$, and conditional average scalar dissipation, $\langle N|\eta \rangle$, is usually accomplished from information already existing on the velocity and mixing fields. In homogeneous flows and special simplifications turbulent shear layers, are possible. It is conditional scalar important that the average dissipation used is with that appearing in the transport equation for the pdf of consistent the mixture fraction. These modelling issues are discussed later.

Solution of the CMC equations usually proceeds in parallel with appropriate modelling for the velocity and mixture fraction fields. CMC can be used with any type of approach to the modelling of the flow and mixing, from simple entrainment models to large eddy simulation. Even experimental data can be used. If second order closure methods are used, terms of the mean mixing is expressed in and variance (usually the averaged^{6,37}) of Favre the density-weighted, i.e. mixture fraction. То obtain the mean density, which is needed for the flow/mixing calculation, the conditional mean density obtained from the CMC solution is weighted by A presumed form for the pdf is assumed and this the mixture fraction pdf. scaled to fit the required mean and variance. is In computing results for unconditionally averaged the species and temperature, the CMC conditional averaged values obtained from the calculation are also weighted by this pdf. For simplicity, the theoretical development works in terms of conventional pdfs rather than Favre pdfs presented here convenient^{6,7,9,47} usually more flows with which are in heat release.

1-5

Where appropriate, indications are given for transcription of formulae in terms of Favre pdfs.

Bilger⁹ outlines experimental data on the shapes of mixture fraction pdfs in various types of flows. In modeling, two different presumed forms are commonly used for the mixture fraction pdf: the clipped Gaussian form shown in Fig. 2 and the beta function form shown in Fig. 3 Of particular note is the behaviour near $\eta = 0$ and 1, the unmixed fluid states. In many two-stream mixing problems there are places in the flow where patches of essentially unmixed fuel and/or oxidiser are intermittently If present. these are treated as completely uncontaminated by the other stream fluid then the mixture fraction pdf will have delta functions at $\eta = 0$ and 1. Alternatively, it can be argued on the basis of the form of the convection diffusion equation, that these patches must be everywhere contaminated by other stream, even if the levels will be insignificantly the small. This viewpoint leads to pdfs which are continuous near $\eta = 0$ and 1. We shall refer to these as smooth pdfs and the others as intermittent pdfs.

It is noted here that in classical turbulent flows, such iets as wakes and boundary layers, these 'unmixed' patches of fluid are assumed to be non-vortical and hence non-turbulent. It has become usual to treat the interface between turbulent and non-turbulent fluid to be the same as that between unmixed or uncontaminated fluid and fluid that is partially mixed with $0 < \eta < l$, even though this question has not been satisfactorily investigated. In most practical systems the unmixed will streams be turbulent to some extent and the analogy between turbulence and mixing does not apply. We will not labour the issue here and will often refer to the partially mixed fluid as the turbulent fluid as has been the convention.

The formulae describing the clipped Gaussian form are as follows.

$$P(\eta) = \gamma_1 \delta(\eta) + \left(1 - \gamma_1 - \gamma_2 \right) P_t(\eta) + \gamma_2 \delta(1 - \eta)$$
(1.2)

$$P_{t}(\eta) = G(\eta)/I_{g} \qquad 0 \le \eta \le 1$$
(1.3)

where

$$G(\eta) \equiv \frac{1}{\sigma_{\rm g}(2\pi)^{1/2}} \exp\left(-\frac{(\eta - \xi_{\rm g})^2}{2\sigma_{\rm g}^2}\right)$$
(1.4)

and

$$I_{\rm g} = \int_{0}^{1} G(\eta) d\eta \tag{1.5}$$

Here $\delta(x)$ represents the Dirac delta function centered at x = 0, and γ_1 and γ_2 are the strengths of these delta functions in the unmixed fluids corresponding to $\xi = 0$ and $\xi = 1$, respectively. $P_t(\eta)$ is the pdf of the turbulent fluid which has a Gaussian form as shown with free parameters ξ_g and σ_g which are the mean and standard deviation of the unclipped Gaussian but are not simple moments of the clipped Gaussian $P_t(\eta)$. They can be related to the mean and variance of the mixture fraction by taking appropriate moments of $P(\eta)$, with γ_1 and γ_2 being regarded as free parameters, or, as is often assumed, having values corresponding to the clipped tails of the distribution

$$\gamma_1 = \int_{-\infty}^{0} G(\eta) d\eta; \qquad \gamma_2 = \int_{1}^{\infty} G(\eta) d\eta \qquad (1.6)$$

For jets far downstream from the potential core, $\gamma_2 \approx 0$ and an alternative proceedure⁵² is to use an empirical correlation for γ_1 .

The beta function distribution is given by

$$P(\eta) = \frac{\eta^{r-1} (1-\eta)^{s-1}}{I_{b}}; \qquad 0 \le \eta \le 1$$
(1.7)

where

$$I_{\rm b} \equiv \int_{0}^{1} \eta^{\rm r-1} (1-\eta)^{\rm s-1} d\eta = \frac{\Gamma(r)\Gamma(s)}{\Gamma(r+s)}$$
(1.8)

and $\Gamma(x)$ is the Gamma function. The parameters r and s are directly related to the mixture fraction mean, $\langle \xi \rangle$, and variance, $\langle \xi'^2 \rangle$, by

$$r = \langle \xi \rangle \left(\langle \xi \rangle \ \frac{1 - \langle \xi \rangle}{\langle \xi'^2 \rangle} - 1 \right); \qquad s = r \frac{1 - \langle \xi \rangle}{\langle \xi \rangle}$$
(1.9)

They are positive quantities, since

$$0 < (<\xi'^2>) < (<\xi>(1 - <\xi>))$$
 (1.10)

except for when $P(\eta)$ is composed entirely of delta functions at $\eta = 0$ and I. This positive nature of r and s ensures that $P(\eta)$ is integrable even though $P(\eta)$ may have "spikes", *i.e.* $P(\eta) \rightarrow \infty$ as $\eta \rightarrow 0$ if 0 < r < 1 and/or $\eta \rightarrow I$ if 0 < s < I. Such spikes are physically and mathematically different to the delta functions of strength γ_1 and γ_2 associated with intermittent pdfs such as the clipped Gaussian just described.

Of some importance to the theoretical development in the early Sections of this review is the fundamentally different character of these pdf forms at their upper and lower bounds. As detailed above, the clipped Gaussian form assumes that statistics in scalar space are intermittent in character at the upper and lower bounds and the pdf has delta function The beta function pdf, on the other hand, has components at these bounds. nature of the such description of the intermittent outer edges no of turbulent flows. In such regions the pdf asymptotes to infinity at the corresponding bound. These and other characteristics of the bounds of the pdf are examined with some care in the development that follows.

CMC methods were developed independently by each of us^{10,12,54}. As such, they are still quite new, and their potential and limitations have not yet been fully established. They have been applied to predictions of flows^{12,25,77,123,124,125} reacting with considerable experimental success. They have also been examined using data bases generated by direct simulation^{91,99,133} numerical also with considerable success. They appear over other advanced to have significant advantages methods of prediction of turbulent flows such as the stationary flamelet reacting laminar (SLFM)^{71,109}, methods Monte simulation of the joint probability Carlo (MC/jpdf)¹¹³ modelling⁵³. density function and linear eddy Direct comparisons with CMC are only now becoming available 91,125,130. The advantages are in the more rigorous basis for the modelling and in reduced methods computational cost (except for SLFM). CMC are, however, this time for flows without significant local extinction only validated at or reignition phenomena present. Second-order conditional moment closure methods and double conditional averaging methods are being developed to address these types of problems.

1.3 Outline of This Review

In this review we seek to set out in a tutorial way the fundamentals

of CMC methods. present some of the practical matters involved in computing solutions and show some of the results that have been obtained. We start from the premise that the reader has a basic familiarity with is available in by turbulence theory, such as the books Tennekes & Lumlev¹³⁵ or Pope 115 , and with the theory of combustion and turbulent combustion such as is available in the book by Turns¹³⁹. Readers concerned primarily with applications of CMC may wish to skim Sections 2 and 3 and begin close study at Section 4 where the main features of the CMC equation and its application are discussed.

The development of CMC theory requires mathematical tools not often available to graduate students. Accordingly, in Section 2 we outline the fundamentals of probability density function (pdf) methods and their basis We show used the theory of generalized functions. how these in are to derive the transport equation for the joint pdf of two scalars which is derivation of the CMC equation. We consider one basis for the also the transport equation for the pdf of the mixture fraction, which is an adjoint equation to the CMC equation, and which may be used to determine the conditional average scalar dissipation, an important quantity in the conditions to be CMC equation. The boundary used in integrating this mixture fraction pdf equation are considered in some detail for various shapes. For those readers not wishing to follow the derivations pdf in detail, a summary of the main results is given at the end of the Section.

The various methods of deriving the first-order CMC equation (for the conditional averages) are presented in Section 3 including outline an of the primary closure hypotheses used in closing the unclosed convection and diffusion terms in the equation. The CMC equation for enthalpy is presented separately. Closure for the conditional average reaction rate source terms that appear in the enthalpy equation is and the discussed. The role of Favre averaging in CMC is outlined. A more detailed discussion of the closure hypotheses from the point of view of Markov process theory and local similarity follows. The Section concludes with a summary of the main results obtained.

main features of the CMC equation are The elucidated in Section 4, velocity including modeling of conditional and conditional dissipation, and of its relation the fast chemistry limit SLFM. to and to Simplifications that in homogeneous problems outlined. The appear are specification of boundary conditions is also elucidated.

Special simplifications are available in turbulent shear layers and

1-9

turbulent slender layer flows. Asymptotic analysis indicates that other the structure of the CMC equations in shear layers is quite different from traditional boundary layer equations for conventional means. Section 5, In shown variation of conditional it is that the cross-stream averages of scalars be neglected in reactive is small and can the first order. Integration of the CMC and pdf transport equations across the layer eliminates terms involving the cross-stream component of the conditional The conditional dissipation, velocity. averaged across the flow, can be from the pdf transport equation of the mixture fraction; and the obtained boundary conditions needed for this are considered in some detail for both intermittent and smooth pdfs. Solutions obtainable in self-similar flows quasi-self-similar, presented. For flows which are only are algorithms are which of the conditional presented the computation scalar ease for CMC flames, Results computations in jet dissipation. reacting scalar mixing layers, and plumes are shown in comparison with experimental data.

The use of CMC in defining novel simple reactor concepts is reviewed in Section 6. The Incompletely Stirred Reactor (ISR) is a generalisation of the well-known Perfectly Stirred Reactor (PSR) and the Dilution Flow Reactor (DFR) is a generalisation of the well-known Plug Flow Reactor (PFR). For definitions of the PSR and PFR see Ref. 139 In the PSR all are uniform within the reactor and equal to the exit values. In scalars generalisation of this, the ISR, the inlet is not fully our premixed and mixture fraction may be non-uniform within the reactor; but the condition uniformity is assumed for the conditional averages of of the reactive The ISR may prove to be a useful model for nonpremixed reactors scalars. such as gas turbine primary zones in which there is strong recirculation and for which the chemical time scales are of the same order as the mixing scales. In the Plug Flow Reactor (PFR) composition is time assumed uniform across the flow: in its DFR counterpart this uniformity is not mixture fraction but conditional required for the is assumed for the averages of reactive scalars. The DFR may prove to be a useful model for nonpremixed reactors such as gas turbine dilution zones and NOx reburn These concepts are introduced in Section 6 even though zones in furnaces. their regimes of validity are as yet not well defined.

A number of studies have been made of the validity of CMC methods using Direct Numerical Simulations (DNS) as the data base. These have made flows with decaying homogeneous turbulent fields, been in flow usually, but not always with homogeneously distributed scalar fields.

1-10

Results are shown in Section 7 from a number of studies with simple and complex chemistry. The Section opens with presentation of CMC predictions of experimental results in chemical reactors with quasi-homogeneous turbulence.

Derivation equations for the conditional of the variances and covariances in nonpremixed systems is presented in Section 8. The consequences of large fluctuation or intermittency in the scalar Results dissipation are discussed. of predictions for the reacting scalar mixing layer are shown against the experimental data. The use of these predictions in second order conditional moment closure is outlined.

In Section 9, progress on the use of conditional moment methods in a number of problems, which require more intricate choice of conditioning are discussed. These include the use of averaging with multiple variables. conditions for multi-stream mixing problems, CMC for premixed combustion and for sprays, the incorporation of differential diffusion effects into the CMC method and a brief discussion on the applications of conditional methods to Lagrangian modelling.

Conclusions and recommendations are summarised in Section 10.

2. PDF METHODS IN TURBULENT FLOWS

A feature of CMC modeling is the rigorous basis for the formulation of the CMC equations and the transparency of the modeling that is done. For this to be demonstrated it is necessary to use mathematical tools that available to workers in this field. Accordingly, are often not in this Section, we review some basic facts related to pdfs and conditional We consider expectations. also the formalism of generalized functions the delta function) which provides very effective (such as a and relatively simple technique for deriving the governing equations pdfs for and conditional moments in turbulent flows. We introduce formulae and will used later in the derivation methods which be of the conditional The transport equation for the pdf of the conserved moment equations. or mixture fraction is important as it is essentially scalar an adjoint equation to the CMC equation and can be used to obtain the conditional scalar dissipation. This is done by integrating it in mixture fraction space. Questions arise as to the nature of the boundary conditions to be applied at $\eta = 0$ and *I*, and how these depend on the structure assumed for the pdf at these bounds - whether smooth or intermittent.

For those readers not wishing to read this Section detail. in а summary of the main results needed in ensuing sections is given at the Pope¹¹³ end. For those readers wishing to explore this material further, gives a more comprehensive review of pdf methods in turbulent combustion, while Lighthill⁸³ and Bracewell²⁰ give more information generalized on functions.

2.1. Pdfs and Conditional Expectations

2.1.1. Pdfs and generalized functions

In turbulent flows. values of the velocity components and scalars such concentrations and temperature stochastic, fluctuating as are variables. Important properties of stochastic variables be can by their probabilities. We prefer "stochastic characterized the term variable" the commonly used "random to more variable", since in the variables of interest are continuous in turbulence space and time and do have the characteristics of white noise. The cumulative not

2-1

probability $\mathcal{P}(Y < Z)$ of a stochastic variable Y is the probability of an event occurring which has Y < Z, where Z is the sample space variable for Y, that is, a particular value in the range of Y. The cumulative probability $\mathcal{P}(Y < Z)$ is a monotonic function of Z. The cumulative probability $\mathcal{P}(Y < Z)$ tends to 0 as $Z \rightarrow \infty$ and $\mathcal{P}(Y < Z)$ tends to 1 as $Z \rightarrow +\infty$. The probability density function (pdf) P is the derivative of the corresponding cumulative probability

$$P(Z) = \frac{d\mathcal{P}(Y < Z)}{dZ}$$
(2.1)

The probability of $Z_1 \leq Y < Z_2$ is given by $\mathcal{P}(Y < Z_2) - \mathcal{P}(Y < Z_1)$ and, if the Eq.(2.1) exists as a usual derivative in derivative, the product $P\Delta Z$ specifies the probability of the event $Z \leq Y < Z + \Delta Z$ for any small ΔZ . The relationship of the functions $\mathcal{P}(Y < Z)$ and P(Z) is shown in Fig. 4 and Thi s f i gures illustrate the case of finite Z_2 - Z_1 (Fig. 4) Fig. 5 and the case of $\Delta Z \equiv Z_2 - Z_1 \rightarrow 0$ (Fig. 5).

its Another basic concept for a stochastic variable is expectation. Expectations are also called mean values and are limit the of average values for large number realizations of the flow. An expectation is the which usually result of averaging is denoted by angular brackets. Probability density functions and expectations are closely related to each other. The mean or expected value of Y, $\langle Y \rangle$ is given by

$$\langle Y \rangle = \int_{-\infty}^{+\infty} Z P(Z) dZ = \int_{-\infty}^{+\infty} Z d\mathcal{P}(Y \langle Z)$$
(2.2)

The expectation of any deterministic function, F(Y), which depends on the stochastic variable Y is given by

$$\langle F(Y) \rangle = \int_{-\infty}^{+\infty} F(Z) P(Z) dZ = \int_{-\infty}^{+\infty} F(Z) d\mathcal{P}(Y \langle Z)$$
(2.3)

averaging understood so-called ensemble averaging. Here is as Let us imagine that an experiment is duplicated in n_a many different places under and values $Y_{\rm k} = Y({\bf x},t)$ $(k=1,2,...,n_a)$ identical conditions are measured in experiments for given **x** and t. Y_k represents these the value of the realization. The ensemble mean value is stochastic value Y in the kth

determined by $\langle Y \rangle = n_a^{-1} \sum_k Y_k$. The value of n_a must be very large. In practice other ways of determining average values may be more convenient. Under certain conditions time averaging can be used. In this paper, however, we imply that averaging means ensemble averaging.

Equation (2.1) can be transformed so that the cumulative probability is expressed in terms of expectations

$$\mathcal{P}(Y < Z) = \int_{-\infty}^{Z} P(Z^{\circ}) dZ^{\circ} = \int_{-\infty}^{+\infty} H(Z - Z^{\circ}) P(Z^{\circ}) dZ^{\circ} = \langle H(Z - Y) \rangle$$
(2.4)

where *H* is the Heaviside function: H(Z)=0 for $Z \le 0$ and H(Z)=1 for Z>0. Differentiating Eq.(2.4) with respect to *Z* and taking into account that $P(Z^{\circ})$ does not depend on *Z* we obtain the identity

$$P(Z) = \int_{-\infty}^{+\infty} \delta(Z - Z^{\circ}) P(Z^{\circ}) dZ^{\circ} = \langle \delta(Z - Y) \rangle$$
(2.5)

which is very convenient representation of the pdf. Here δ is Dirac's been introduced derivative delta function which has as the of the Heaviside function $\delta = dH/dZ$ ($\delta(Z)$ is zero everywhere except at Z=0 where the delta function is infinite). This derivative does not exist as a usual derivative since H is a discontinuous function. The delta function is a so-called generalized function, and it has a more precise formal definition than that just introduced.

The formal definition of the delta function specifies how it affects Equation (2.5) is not suitable as a strict definition ordinary functions. of P because: 1) P is not an arbitrary function and we have used Eq.(2.5)to determine the properties of *P* rather than to introduce the delta function; and 2) $\mathcal{P}(Y \le Z)$ can be a discontinuous function of Z and this means that the pdf function P introduced by Eq.(2.1) can itself involve The delta function can be strictly defined by describing delta functions. its properties: the function which affects any function F(Z)(which belongs to the certain class of "good" functions 83) such that

$$\int_{-\infty}^{+\infty} F(Z) \,\delta(Z) \,dZ = F(0) \tag{2.6}$$

the delta function. In general, this definition depends on is what we understand as the class of "good" functions. Normally these functions Fmust be smooth enough and rapidly tend to zero as $Z \rightarrow \pm \infty$. We assume here that the function F is continuous and has as many derivatives as required. the definition in Eq.(2.6), delta function is an even According to the function $\delta(-Z) =$ $\delta(Z)$. Generalized functions, f_{g} , defined are by describing their properties under integration. This means the specification of: 1) the variable which the over product $f_{\circ}F$ is to be result arbitrary integrated: 2) the of the integration for any and function F which belongs to the class of good functions.

With this more formal definition we can now prove that Eq.(2.5) is the property of the delta function introduced by Eq.(2.6). Equation (2.6) defines the delta function located at Z=0. This definition can be extended to involve the delta function located at $Z=Z_0$

$$F(Z_0) = \int_{-\infty}^{+\infty} F(Z+Z_0) \,\delta(Z) \,dZ = \int_{-\infty}^{+\infty} F(Z^\circ) \,\delta(Z^\circ-Z_0) \,dZ^\circ$$
(2.7)

This equation can be formally derived from Eq.(2.6) by substituting $F_0(Z) \equiv F(Z+Z_0)$ for F(Z) and introducing the new variable $Z^\circ = Z+Z_0$. It is easy to see that the new function F_0 also belongs to the class of good functions.

After formal substitution of Y for Z_0 and Z for Z° , Eq.(2.7) is averaged over Y

$$\langle F(Y) \rangle = \langle \int_{-\infty}^{+\infty} F(Z) \,\delta(Z-Y) \,dZ \rangle = \int_{-\infty}^{+\infty} F(Z) \,\langle \delta(Z-Y) \rangle \,dZ$$
(2.8)

Since Eq.(2.8) is valid for any F(Z), upon comparing Eq.(2.8) and Eq.(2.3) we obtain $P(Z) = \langle \delta(Z-Y) \rangle$ which is Eq.(2.5). Equation (2.5) is the basic equation which is used to derive the governing equation for the pdf, P(Z). The governing equation is derived first for $\psi = \delta(Z-Y)$ and then averaged. This technique is not the only one possible. Sometimes the characteristic function $\Phi(\lambda) \equiv \langle \exp(i\lambda Y) \rangle$, which is the Fourier transform of the pdf, is used for derivation of the pdf equations 72 . While $\Phi(\lambda)$ has the advantage analytical function, derivations using $\Phi(\lambda)$ usually of being an require more steps.

2.1.2. Joint pdfs

In this section generalize the single-variable probability we introduced in the previous section to the case of n stochastic functions variables Y_i and their joint pdf. The joint cumulative probability $\mathcal{P}(\mathbf{Y} < \mathbf{Z})$ stochastic variables Y_i (*i*=1,2,...*n*) is the probability of a set of that $Y_n < Z_n$ are jointly valid. The joint pdf $Y_1 < Z_1$, $Y_2 < Z_2$, ..., and the average value of a deterministic function $F(\mathbf{Y})$ $P(\mathbf{Z}) \equiv P(Z_1, \dots, Z_n)$ are given by

$$P(\mathbf{Z}) = \frac{\partial^{n} \mathcal{P}(\mathbf{Y} < \mathbf{Z})}{\partial Z_{1} \partial Z_{1} \dots \partial Z_{n}}$$
(2.9)

$$\langle F(\mathbf{Y}) \rangle = \int_{\infty} F(\mathbf{Z}) P(\mathbf{Z}) dZ_1 dZ_2 ... dZ_n$$
 (2.10)

Here, and further on, the integral is taken over all possible values of Z. If Р is an ordinary function and $\Delta \mathbf{Z}$ is small. product the $P(\mathbf{Z}) \Delta Z_1 \Delta Z_2 \dots \Delta Z_n$ of the joint pdf P and the volume of the box with center **Z** and sides $\Delta \mathbf{Z}$ specifies the probability of **Y** being in this box. If the is the product of the joint pdf one-variable pdfs $P(\mathbf{Z}) = P(Z_1) P(Z_2) \dots P(Z_n),$ the stochastic variables Y_{i} are statistically independent. We do not introduce any special notation for the joint pdf. The number of the stochastic variables is specified by the number of arguments Z_i in P. Surface and contour plots of a Gaussian two-variable joint pdf are shown in Figs 6 and 7

Joint cumulative probabilities and joint pdfs can also be introduced using the functions H and δ

$$\mathcal{P}(\mathbf{Y} < \mathbf{Z}) = \langle H(Z_1 - Y_1) H(Z_2 - Y_2) \dots H(Z_n - Y_n) \rangle$$
(2.11)

$$P(\mathbf{Z}) = \langle \psi \rangle; \quad \psi \equiv \delta(Z_1 - Y_1) \,\delta(Z_2 - Y_2) \dots \,\delta(Z_n - Y_n) \tag{2.12}$$

where the new function ψ is introduced for convenience. This function ψ is called the fine-grained pdf. Equations (2.9) and (2.12) are similar to Eqs.(2.1) and (2.5) for a single stochastic variable.

2.1.3. Conditional pdfs and conditional expectations

Now we turn to consideration of conditional pdfs and conditional Conditional pdfs and conditional expectations are the pdfs expectations. selected realizations and expectations determined for certain among the ensemble of all realizations of the flow. The criterion for selection of these realizations is the fulfillment of a particular condition which is specified. The conditional cumulative probability $\mathcal{P}(\mathfrak{A}|\mathfrak{B})$ is the cumulative probability of the event A determined for these realizations which meet the condition appearing on the right-hand side of the vertical bar (that is for these realizations for which statement B is valid). For example, if the event \mathfrak{A} can be specified as $Y_1 < Z_1$ and the condition \mathfrak{B} can be specified as $Y_2 < Z_2$ the corresponding conditional probability can be written as $\mathcal{P}(Y_1 < Z_1 \mid Y_2 < Z_2)$. According to Bayes theorem ¹⁰⁸, the joint probability of two events \mathfrak{A} and \mathfrak{B} both occurring, $\mathcal{P}(\mathfrak{A},\mathfrak{B})$, can be decomposed into

$$\mathcal{P}(\mathfrak{A},\mathfrak{B}) = \mathcal{P}(\mathfrak{A} \mid \mathfrak{B}) \mathcal{P}(\mathfrak{B}) \tag{2.13}$$

The conditional pdf $P(Z_1|\mathfrak{B})$ is the probability density for Y_1 being at the value Z_1 for those realizations which meet the condition appearing on the right-hand side of the vertical bar. In general, the condition \mathfrak{B} should have a non-zero probability. If \mathfrak{B} represents a condition which never occurs, $P(Z_1|\mathfrak{B})$ cannot be determined. If the condition \mathfrak{B} is specified as $Y_2=Z_2$, this means that $Z_2-\Delta Z \leq Y_2 < Z_2+\Delta Z$ for sufficiently small ΔZ (note that the probability of $Y_2=Z_2$ may be zero). We consider here two stochastic variables Y_i , i=1,2. Their joint probability function $P(Z_1,Z_2)$ can be also decomposed into the product of the pdf of Z_2 multiplied by the pdf of Y_1 conditional on $Y_2=Z_2$

$$P(\mathbf{Z}) \equiv P(Z_1, Z_2) = P(Z_1 | Y_2 = Z_2) P(Z_2)$$
(2.14)

Equation (2.14) determines the conditional pdf $P(Z_1 | Y_2 = Z_2)$. If the variables Z_1 and Z_2 are statistically independent then $P(Z_1, Z_2) = P(Z_1)P(Z_2)$ and $P(Z_1 | Y_2 = Z_2) = P(Z_1)$.

The conditional expectation $\langle Y_2 | \mathfrak{B} \rangle$ is the average of Y_2 over the sub-ensemble of these realizations which meet the condition appearing on

the right-hand side of the vertical bar. If the condition \mathfrak{B} is specified as $Y_2=Z_2$, this has the same meaning as in Eq.(2.14). The conditional expectation is determined by

$$= \int_{-\infty}^{+\infty} Z_{1}P(Z_{1}|Y_{2}=Z_{2}) dZ_{1} = \frac{\int_{-\infty}^{+\infty} Z_{1}P(Z_{1},Z_{2}) dZ_{1}}{P(Z_{2})}$$
(2.15)

The conditional expectation is a function of the variable Z_2 which averaging. We may condition of specifies the use the brief notation $< Y_1 | Z_2 >$ which means exactly the same as $\langle Y_1 | Y_2 = Z_2 \rangle$. The conditional averages and conditional pdfs are shown in Fig. 8 the case of for a "banana" pdf. This figure illustrates the differences between $P(Z_2 | Y_1 = a)$ and $P(Z_2 | Y_1 = b)$ for conditional pdfs and between $\langle Y_1 | Z_2 \rangle$ and $\langle Y_2 | Z_1 \rangle$ for conditional expectations. The following properties of the conditional Eq.(2.15). The expectations are derived from conventional (unconditional) mean value is given by the integral

$$\langle Y_1 \rangle = \int_{-\infty}^{+\infty} \langle Y_1 | Y_2 = Z_2 \rangle P(Z_2) dZ_2$$
 (2.16)

If variables Y_1 and Y_2 are statistically independent the conditional expectation $\langle Y_1 | Y_2 = Z_2 \rangle$ does not depend on Z_2 and coincides with the unconditional expectation $\langle Y_1 \rangle$. For any deterministic function $F(Y_2)$ which does not depend on Y_1 , the conditional expectation of the product Y_1F is given by

$$\langle Y_1 F(Y_2) | Y_2 = Z_2 \rangle = \langle Y_1 | Y_2 = Z_2 \rangle F(Z_2)$$
 (2.17)

If Y_1 is a deterministic function of Y_2 (that is $Y_1=F(Y_2)$) then, according to Eq.(2.17), the conditional expectation is the function, itself: $\langle F(Z_2) | Y_2=Z_2 \rangle = F(Z_2).$

Using the formalism of the delta function the conditional expectations can be also expressed as

$$P(Z_2) =$$
 (2.18)

In order to prove this equation we multiply Eq.(2.18) by any good function

 $F(Z_2)$ and integrate over Z_2 . The left-hand and right-hand sides of Eq.(2.18) then take the forms

$$\int_{-\infty}^{+\infty} F(Z_2) < Y_1 | Y_2 = Z_2 > P(Z_2) dZ_2 = \int_{-\infty}^{+\infty} P(Z_2) dZ_2 =$$

$$\int_{-\infty}^{+\infty} F(Z_2) < Y_1 \delta(Z_2 - Y_2) > dZ_2 = < Y_1 \int_{-\infty}^{+\infty} F(Z_2) \delta(Z_2 - Y_2) dZ_2 > =$$

Here, in the second equation, Y_1 does not depend on Z_2 and Y_1 is put outside the integration sign. Since both sides of Eq.(2.18) are thus found to be identical for any arbitrary function F, this proves Eq.(2.18). Equation (2.18) is often used in pdf derivations. This equation is more general than Eq.(2.5) since Eq.(2.5) can be obtained by substituting $Y_1=1$ into Eq.(2.18). Equation (2.18) can be generalized for multiple conditions

$$\langle Y_0 | \mathbf{Y} = \mathbf{Z} \rangle P(\mathbf{Z}) = \langle Y_0 \psi \rangle$$
(2.19)

where ψ is specified in Eq.(2.12) $\mathbf{Y}=(Y_1,...,Y_n)$, $\mathbf{Z}=(Z_1,...,Z_n)$ and Y_0 is not included in the set $(Y_1,...,Y_n)$.

2.2. Differentiating Generalized Functions and Pdfs

show how to differentiate In this section we generalized functions technique for the derivation of then we apply this identities and some which involve pdfs and conditional expectations. These identities will be used later when we consider the governing equation for the joint pdf.

2.2.1. Generalized derivative

We consider here functions or generalized functions for which normal derivatives do not exist. If the derivative of the function f exists, we can write for any good function F

$$\int_{-\infty}^{+\infty} F(Z)f'(Z)dZ = \left(Ff\right)_{-\infty}^{+\infty} - \int_{-\infty}^{+\infty} F'(Z)f(Z)dZ = - \int_{-\infty}^{+\infty} F'(Z)f(Z)dZ$$
(2.20)

Equation (2.20) results from integration by parts and taking into account that the good function F rapidly tends to zero as $Z \rightarrow \pm \infty$. If, on the other the derivative $f' \equiv df/dZ$ does exist hand. not as a normal equation derivative, this "non-generalized" becomes the definition of the generalized derivative. This derivative is а generalized which function has properties specified by Eq.(2.20). For example, let f be the Heaviside function: f(Z)=H(Z). Equation (2.20) takes the form

$$\int_{-\infty}^{+\infty} F(Z)H'(Z)dZ = -\int_{-\infty}^{+\infty} F'(Z)H(Z)dZ = -\int_{0}^{+\infty} F'(Z)dZ = -\left(F(Z)\right)_{0}^{+\infty} = F(0)$$

This equation coincides with the definition of the delta function in Eq.(2.6) which proves that $\delta(Z)=H'(Z)\equiv dH/dZ$. The derivative of the delta function $\delta'\equiv d\delta/dZ$ is also a generalized function and it is defined by the equation

$$\int_{-\infty}^{+\infty} F(Z) \,\delta'(Z) \,dZ = - \int_{-\infty}^{+\infty} F'(Z) \,\delta(Z) \,dZ = -F'(0)$$
(2.21)

The higher derivatives of the delta function $\delta^{(n)} \equiv d^n \delta / dZ^n$ are introduced in a similar way

$$\int_{-\infty}^{+\infty} F(Z) \,\delta^{(n)}(Z) \,dZ = (-1)^n \int_{-\infty}^{+\infty} F^{(n)}(Z) \,\delta(Z) \,dZ = (-1)^n \left[\frac{d^n F}{dZ^n} \right]_{Z=0}$$
(2.22)

We should emphasize here, that if f' is a generalized function, Eq.(2.20) looks like a mere integration by parts, but it actually represents the definition of f'. If f' happens to be a ordinary function, Eq.(2.20) is valid and still represents simple integration by parts. This gives а simple rule for checking any equation which may involve generalized differentiation: multiply this equation by a good function F and formally integrate it by parts so that all generalized derivatives are excluded.

Let us consider a few examples of identities which will be utilized

later. We will use formal differentiation by parts to prove that

$$\frac{d}{dt}\delta\left(Z - Y(t)\right) = -\frac{\partial}{\partial Z}\left(\frac{dY}{dt}\delta\left(Z - Y(t)\right)\right)$$
(2.23)

where Y is a function of the variable t. Equation (2.23) is multiplied by a good function F(Z) and integrated over Z, with the left-hand side becoming

$$\int_{-\infty}^{+\infty} F(Z) \frac{d}{dt} \delta\left(Z - Y(t)\right) dZ = \frac{d}{dt} \int_{-\infty}^{+\infty} F(Z) \delta\left(Z - Y(t)\right) dZ = \frac{dF(Y(t))}{dt} = F'(Y) \frac{dY}{dt}$$

and this is identical to the right-hand side since the integrated by parts

$$\int_{-\infty}^{+\infty} F(Z) \frac{d}{dZ} \left(\frac{dY}{dt} \,\delta\left(Z - Y(t)\right) \right) dZ = \int_{-\infty}^{+\infty} \frac{dY}{dt} \,\delta\left(Z - Y(t)\right) \frac{dF(Z)}{dZ} dZ = \frac{dY}{dt} F'(Y)$$

This proves Eq.(2.23). We note that Y does not depend on Z in Eq.(2.23) so that dY/dt can be taken outside of the differentiation sign $\partial/\partial Z$. The form of the equation, as it is written in Eq.(2.23), is usually more convenient for our purposes.

We note that it is quite convenient with delta to operate the function as if it was an ordinary (non-generalized) function. In many this yields correct results. А sounder viewpoint cases is that any equation involving the function implies that this equation delta is valid when applied to a good function F.

The rules for operating with multiple delta functions are similar to the rules discussed above. We consider the variables Y_{i} (*i*=1,...*n*) which The derivative depend on time, t. time of the fine-grained pdf,ψ, specified in Eq.(2.12) is given by

$$\frac{d\Psi}{dt} = -\sum_{i=1}^{n} \frac{\partial}{\partial Z_{i}} \left(\frac{dY_{i}}{dt} \; \Psi \right) \equiv -\frac{\partial}{\partial Z_{i}} \left(\Psi \frac{dY_{i}}{dt} \right)$$
(2.24)

Equation (2.23) is applied in Eq.(2.24) to each of the delta functions in the product ψ . The summation convention over repeated dummy indexes is

applied here and further on (that is, the summation sign " Σ " is omitted). Equation (2.24) can be checked by multiplying Eq.(2.24) by the good function $F(\mathbf{Z})$ and integrating over \mathbf{Z}

$$\int_{\infty} F(\mathbf{Z}) \frac{d\Psi}{dt} dZ_{1}...dZ_{n} = \frac{d}{dt} \int_{\infty} F(\mathbf{Z}) \Psi dZ_{1}...dZ_{n} = \frac{dF(\mathbf{Y}(t))}{dt} = \frac{\partial F}{\partial Y_{i}} \frac{dY_{i}}{dt}$$
$$-\int_{\infty} F(\mathbf{Z}) \frac{\partial}{\partial Z_{i}} \left(\Psi \frac{dY_{i}}{dt} \right) dZ_{1}...dZ_{n} = \int_{\infty} \Psi \frac{dY_{i}}{dt} \frac{\partial}{\partial Z_{i}} \left(F(\mathbf{Z}) \right) dZ_{1}...dZ_{n} = \frac{\partial F}{\partial Y_{i}} \frac{dY_{i}}{dt}$$

The first step uses the multidimensional form of Eq.(2.21) and the last integral in this equation is formally integrated by These parts. mathematical transformations are similar to the proof of Eq.(2.23). If the Y_{i} depend not only on time t but also on coordinates $\mathbf{x} = (x_1, x_2, x_3)$, partial derivatives should be substituted for the ordinary derivatives in Eq.(2.24)

$$\frac{\partial \Psi}{\partial t} = -\frac{\partial}{\partial Z_{i}} \left(\Psi \frac{\partial Y_{i}}{\partial t} \right)$$
(2.25)

Let consider the differentiation of the product $f_{s}\Psi$, where us assumed to smooth function and specified $f_s = f_s(t)$ is be a ψ is as in differentiation Eq.(2.12). The rules are similar the differentiation of to ordinary functions

$$\frac{d\psi f_s}{dt} = f_s \frac{d\psi}{dt} + \psi \frac{df_s}{dt}$$
(2.26)

This equation is tested as usual: the left-hand side of Eq.(2.26) is multiplied by $F(\mathbf{Z})$ and integrated over \mathbf{Z}

$$\int_{\infty} F(\mathbf{Z}) \frac{d\Psi f_s}{dt} dZ_1 \dots dZ_n = -\frac{d}{dt} \int_{\infty} F(\mathbf{Z}) \Psi f_s dZ_1 \dots dZ_n = -\frac{d}{dt} \left(F(\mathbf{Y}(t)) f_s(t) \right)$$

while after similar operations the right-hand side takes the form

$$\int_{\infty} F(\mathbf{Z}) \left(f_{s} \frac{d\psi}{dt} + \psi \frac{df_{s}}{dt} \right) dZ_{1} \dots dZ_{n} = f_{s} \frac{d}{dt} \int_{\infty} F(\mathbf{Z}) \psi dZ_{1} \dots dZ_{n} + \frac{df_{s}}{dt} \int_{\infty} F(\mathbf{Z}) \psi dZ_{1} \dots dZ_{n} = f_{s} \frac{dF(\mathbf{Y}(t))}{dt} + F(\mathbf{Y}(t)) \frac{df_{s}(t)}{dt}$$

It is easy to see that the both sides are equal since both F and f_s are smooth functions which can be differentiated in the usual way.

2.2.2. The gradient operator

We consider equations which involve gradient some the operator. Variables $Y_{\rm i}$ assumed here be dependent the coordinates are to on $\mathbf{x} = (x_1, x_2, x_3)$. The gradient operator applied to ψ yields

$$\nabla \Psi = - \frac{\partial}{\partial Z_i} \left(\Psi \nabla Y_i \right)$$
(2.27)

This equation is a vector equation with components that are specified by the derivatives $\partial/\partial x_1$, $\partial/\partial x_2$, $\partial/\partial x_3$. Each of these components is similar to Eq.(2.25). Let us consider the derivatives of higher order which result from the divergence operator being applied to Eq.(2.27). We use the vector analog of Eq.(2.26), $\nabla \cdot (\mathbf{f}_s \psi) = \psi \operatorname{div}(\mathbf{f}_s) + \mathbf{f}_s \cdot \nabla \psi$, and also assume that the Y_i are smooth functions of the coordinates.

$$\nabla^{2} \Psi = -\nabla \cdot \left(\frac{\partial \Psi \nabla Y_{i}}{\partial Z_{i}} \right) = -\frac{\partial \Psi \nabla^{2} Y_{i}}{\partial Z_{i}} - \frac{\partial (\nabla \Psi \cdot \nabla Y_{i})}{\partial Z_{i}} = -\frac{\partial \Psi \nabla^{2} Y_{i}}{\partial Z_{i}} + \frac{\partial^{2} \Psi (\nabla Y_{i} \cdot \nabla Y_{i})}{\partial Z_{i} \partial Z_{j}}$$

Note that Eq.(2.27) is used twice and that ∇Y_i does not depend on Z.

For pdf derivations we will need a slightly more complicated form of this equation which involves the molecular diffusion term. First Eq.(2.27) is multiplied by ρD (*D* is the molecular diffusion coefficient which is assumed to be similar for all scalars $D_i=D$; ρ is the density) and then we take the divergence of the product. The values of ρ and *D* do not depend on Z_i . After some manipulation we obtain

$$\operatorname{div}\left(\rho D\nabla\Psi\right) = -\frac{\partial}{\partial Z_{i}}\left(\operatorname{div}\left(\psi\rho D\nabla Y_{i}\right)\right) = -\frac{\partial}{\partial Z_{i}}\left(\psi\operatorname{div}(\rho D\nabla Y_{i})\right) + \frac{\partial^{2}}{\partial Z_{i}\partial Z_{j}}\left(\psi\rho D(\nabla Y_{i}:\nabla Y_{j})\right)$$
(2.28)

This equation is an identity and does not involve any modeling of the properties of turbulent scalar transport. If ρ and D are not constants, the term on the left-hand side of Eq.(2.28) is more conveniently written

as

$$\operatorname{div}\left(\rho D\nabla\psi\right) = \operatorname{div}\left(\nabla(\psi\rho D)\right) - \operatorname{div}\left(\psi\nabla(\rho D)\right)$$
(2.29)

Another equation which has some similarities with Eq.(2.28) is obtained by applying the divergence operator to $\psi \rho D \nabla Y_i$

$$\operatorname{div}\left(\psi\rho D\nabla Y_{i}\right) = \psi \operatorname{div}\left(\rho D\nabla Y_{i}\right) - \frac{\partial}{\partial Z_{j}}\left(\psi\rho D(\nabla Y_{i} \cdot \nabla Y_{j})\right)$$
(2.30)

Here *i* is any of 1,...,n if Y_i is one of the variables $Y_1,...,Y_n$ in Eq.(2.12) or *i=0* otherwise.

2.2.3. Differentiating the joint pdf

The equations for ψ derived above can be considered as equations for the fine-grained pdf. The variables Y_i are stochastic variables which depend on time and coordinates. Now we can derive some equations involving the joint pdf $P(\mathbf{Z})$. The simplest equation represents the time derivative of the pdf. After averaging, Eq.(2.25) takes the form

$$\frac{\partial P(\mathbf{Z})}{\partial t} = -\frac{\partial}{\partial Z_{i}} \left\{ < \frac{\partial Y_{i}}{\partial t} \mid \mathbf{Y} = \mathbf{Z} > P(\mathbf{Z}) \right\}$$
(2.31)

Equations (2.12) and (2.19) have been taken into account in arriving at this result.

We will need some equations which involve the gradient operator. The technique for their derivation is similar: the equations involving the fine-grained pdf are averaged and then Eqs.(2.12) and (2.19) are applied. Combining Eqs.(2.28) and (2.29) we obtain after averaging

$$\operatorname{div}\left(\langle \rho D \nabla \psi \rangle\right) = \nabla^{2}\left(\langle \rho D | \mathbf{Y} = \mathbf{Z} \rangle P\right) - \operatorname{div}\left(\langle \nabla(\rho D) | \mathbf{Y} = \mathbf{Z} \rangle P\right) = (2.32)$$
$$= -\frac{\partial}{\partial Z_{i}}\left(\langle \operatorname{div}(\rho D \nabla Y_{i}) | \mathbf{Y} = \mathbf{Z} \rangle P\right) + \frac{\partial^{2}}{\partial Z_{i} \partial Z_{j}}\left(\langle \rho D(\nabla Y_{i} \cdot \nabla Y_{j}) | \mathbf{Y} = \mathbf{Z} \rangle P\right)$$

where $P = P(\mathbf{Z})$. Averaging of Eq.(2.30) yields

$$\operatorname{div}\left(<\!\rho D\nabla Y_i | \mathbf{Y}=\mathbf{Z}>\!P\right) =$$

$$= \left(< \operatorname{div}(\rho D \nabla Y_{i}) | \mathbf{Y} = \mathbf{Z} > P \right) - \frac{\partial}{\partial Z_{j}} \left(< \rho D (\nabla Y_{i} \cdot \nabla Y_{j}) | \mathbf{Y} = \mathbf{Z} > P \right)$$
(2.33)

Note that these equations are identities and their derivation does not use the scalar transport equation. Equations (2.32) and (2.33) have been derived for the case of $D_i=D$, but a more general result with $D_i \neq D$ can also be obtained in a similar manner (see Section 9.1.1).

2.2.4. Differentiating conditional expectations

It is well-known that the conventional (unconditional) averaging and differentiation can be commuted

$$\langle \frac{\partial Y}{\partial t} \rangle = \frac{\partial \langle Y \rangle}{\partial t}$$
 (2.34)

This equation can normally be applied in engineering applications and we derivations have used it repeatedly in made so far. [Mathematically, Eq.(2.34) needs further qualification. For example, it is possible that $\partial < Y > /\partial t$ $\partial Y/\partial t$ exists but does not.] Equation (2.34)is valid for conventional expectations but it is not necessarily valid for conditional expectations since the condition of averaging may also be dependent on the variable t. The equation for the differential of a conditional mean can be derived by differentiating the product

$$\frac{\partial}{\partial t} \left(Y_1 \ \delta(Z_2 - Y_2) \right) = \frac{\partial Y_1}{\partial t} \ \delta(Z_2 - Y_2) - \frac{\partial}{\partial Z_2} \left(Y_1 \frac{\partial Y_2}{\partial t} \ \delta(Z_2 - Y_2) \right)$$
(2.35)

The conditional mean is determined by Eq.(2.18) so that Eq.(2.35) after averaging takes the form

$$\frac{\partial \langle Y_1 | Y_2 = Z_2 \rangle}{\partial t} P(Z_2) + \langle Y_1 | Y_2 = Z_2 \rangle \frac{\partial P(Z_2)}{\partial t}$$
$$= \langle \frac{\partial Y_1}{\partial t} | Y_2 = Z_2 \rangle P(Z_2) - \frac{\partial}{\partial Z_2} \left(\langle Y_1 \frac{\partial Y_2}{\partial t} | Y_2 = Z_2 \rangle P(Z_2) \right)$$
(2.36)

It is seen that there are two additional terms in Eq.(2.36)and the first term on the left-hand side is not necessarily equal to the first term on (2.36)includes Eq.(2.31) the right-hand side. Equation which can be derived from Eq.(2.36) by substituting $Y_1=1$. Equation (2.36) can be easily expectations with generalized for conditional multiple conditions. If Y_2 does not depend on t, Eq.(2.36) takes the form

$$\frac{\partial \langle Y_1 | Y_2 = Z_2 \rangle}{\partial t} = \langle \frac{\partial Y_1}{\partial t} | Y_2 = Z_2 \rangle$$
(2.37)

When the condition of averaging does not depend on t, differentiating with respect to t and conditional averaging can be commuted.

2.3. The Pdf Equations

In this section the joint pdf equation for a set of the scalar fields Y_i (i=1,...,n) in a turbulent flow is derived. Each of the scalars is a function of time t and physical coordinates **x** and, assuming Fickian diffusion, is governed by the scalar transport equation

$$\rho \frac{\partial Y_i}{\partial t} + \rho \mathbf{v} \cdot \nabla Y_i - \operatorname{div} \left(\rho D_i \nabla Y_i \right) = \rho W_i$$
(2.38)

2.3.1. The equation for the fine grained pdf

In this section we derive the equation for the fine grained pdf ψ based the (2.38). The pdf which is on transport equation transport is then obtained averaging of the equation equation by for ψ. Substituting $\partial Y_i/\partial t$ determined by Eq.(2.38) into Eq.(2.25) we obtain

$$\frac{\partial \Psi}{\partial t} = \frac{\partial}{\partial Z_i} \left(\Psi \left(\mathbf{v} \cdot \nabla Y_i - \frac{1}{\rho} \operatorname{div}(\rho D_i \nabla Y) - W_i \right) \right)$$
(2.39)

Equation (2.27) multiplied by \mathbf{v} yields

$$\mathbf{v} \cdot \nabla \boldsymbol{\Psi} = - \frac{\partial}{\partial Z_i} \left(\boldsymbol{\psi} \left(\mathbf{v} \cdot \nabla Y_i \right) \right)$$
(2.40)

Note that **v** does not depend on the independent sample space variable Z_i . Combining Eq.(2.39) and Eq.(2.40) we obtain

$$\rho \frac{\partial \Psi}{\partial t} + \rho \left(\mathbf{v} \cdot \nabla \Psi \right) + \frac{\partial}{\partial Z_{i}} \left(\Psi \operatorname{div}(\rho D_{i} \nabla Y) \right) = - \frac{\partial}{\partial Z_{i}} \left(\Psi \rho W_{i} \right)$$
(2.41)

Density, ρ , is independent of the independent sample space variable Z_i but ρ is a stochastic function of t and \mathbf{x} which can be stochastically

dependent on Y_i . The continuity equation

$$\frac{\partial \rho}{\partial t} + \operatorname{div}(\rho \mathbf{v}) = 0 \tag{2.42}$$

multiplied by ψ and added to Eq.(2.41) transforms Eq.(2.41) into the divergent form

$$\frac{\partial \rho \Psi}{\partial t} + \operatorname{div} \left(\rho \mathbf{v} \Psi \right) + \frac{\partial}{\partial Z_{i}} \left(\Psi \operatorname{div} (\rho D_{i} \nabla Y) \right) = - \frac{\partial}{\partial Z_{i}} \left(\rho \Psi W_{i} \right)$$
(2.43)

The divergent form of the equation for ψ is more convenient for averaging. Equation (2.43) is a transport equation for the fine grained joint pdf ψ . mathematical identity. It essentially incorporates This equation is not а which transport in turbulent the equations govern scalar flows. If all diffusion coefficients $D_i = D$, Eq.(2.28) transforms Eq.(2.43) are the same into the form

$$\frac{\partial \rho \psi}{\partial t} + \operatorname{div} \left(\rho \mathbf{v} \psi \right) - \operatorname{div} \left(\rho D \nabla(\psi) \right) + \\ + \frac{\partial^2}{\partial Z_i \partial Z_j} \left(\psi \rho D(\nabla Y_i \cdot \nabla Y_j) \right) = - \frac{\partial}{\partial Z_i} \left(\psi \rho W_i \right)$$
(2.44)

2.3.2. The joint pdf equation

According to Eq.(2.19) averaging of Eq.(2.43) yields the equation for the joint pdf $P(\mathbf{Z})$

$$\frac{\partial \langle \rho | \mathbf{Y} = \mathbf{Z} \rangle P}{\partial t} + \operatorname{div} \left(\langle \rho \mathbf{v} | \mathbf{Y} = \mathbf{Z} \rangle P \right) + \frac{\partial}{\partial Z_{i}} \left(\langle \operatorname{div}(\rho D_{i} \nabla Y) | \mathbf{Y} = \mathbf{Z} \rangle P \right) =$$

$$= - \frac{\partial}{\partial Z_{i}} \left(\langle \rho W_{i} | \mathbf{Y} = \mathbf{Z} \rangle P \right)$$
(2.45)

Equation (2.45)is not the only possible form of the joint pdf If all of the scalars have similar diffusion coefficients equation. $D_i = D$, last term on the left-hand side of Eq.(2.45) can be transformed into the which explicitly involves the form the scalar dissipation terms $D(\nabla Y_i \cdot \nabla Y_i)$. This form can be obtained by substituting Eq.(2.32) into Eq.(2.45) or by substituting Eq.(2.29) into Eq.(2.44) and averaging

$$\frac{\partial <\rho | \mathbf{Y} = \mathbf{Z} > P}{\partial t} + \operatorname{div} \left(<\rho \mathbf{v} | \mathbf{Y} = \mathbf{Z} > P \right) + \frac{\partial^2}{\partial Z_i \partial Z_j} \left(<\rho D(\nabla Y_i \cdot \nabla Y_j) | \mathbf{Y} = \mathbf{Z} > P \right) + \operatorname{div} \left(<\nabla(\rho D) | \mathbf{Y} = \mathbf{Z} > P \right) - \nabla^2 \left(<\rho D | \mathbf{Y} = \mathbf{Z} > P \right) = -\frac{\partial}{\partial Z_i} \left(<\rho W_i | \mathbf{Y} = \mathbf{Z} > P \right)$$
(2.46)

equations These pdf transport are derived using the scalar transport equation (2.38). Equations (2.45) and (2.46) are exact equations but they are unclosed. This means that some of the coefficients of Eqs.(2.45) and (2.46) (these are $\langle \rho \mathbf{v} | \mathbf{Y} = \mathbf{Z} \rangle$, $\langle \rho D(\nabla Y_i \cdot \nabla Y_i) | \mathbf{Y} = \mathbf{Z} \rangle$ and $\langle \operatorname{div}(\rho D_i \nabla Y) | \mathbf{Y} = \mathbf{Z} \rangle$) are not known. Such unclosed equations need further modeling to transform the equations into a closed form.

2.3.3. The case of high Reynolds numbers

If the Reynolds number, Re, is high, the last two terms on the left-hand side of Eq.(2.46) can be neglected. These terms correspond to macro-transport by molecular diffusion. When Re is large the kinematic the viscosity coefficient, v, and diffusion coefficient, D, relatively are small (we assume here that Eq.(2.46) to have been normalized by a suitable velocity, length and density so that $v \sim D \sim Re^{-1}$).

We consider the limit $D \rightarrow 0$ (that is $\text{Re} \rightarrow \infty$) in order to estimate the dependence of some gradients and functions on D. If Re is large enough (D is small enough), some of the functions do not depend on D while others indicate a significant dependence on D (or v). The instantaneous gradient can be estimated as $\nabla Y \sim Y D^{-1/2}$ (since Y can have different scaling, ∇Y is proportional to Y). This estimate follows from the fact that the mean $\langle D(\nabla Y)^2 \rangle$ scalar dissipation specifies the rate of the dissipation of the scalar *Y* fluctuations. The dissipation is linked to other macro-parameters of turbulence and does not depend on D. That is $\langle D(\nabla Y)^2 \rangle \sim 1$ as $D \to 0$ and $Re \rightarrow \infty$. Here we refer to the important property of developed turbulence independence of its macro-characteristics from the Reynolds number ^{98,115,135}. The diffusivity coefficient, D, and the density, ρ, are $\nabla D \sim D D^{-1/2} = D^{1/2}$ the concentrations of species so functions of that and $\nabla \rho \sim \rho D^{-1/2}.$ The characteristic values of instantaneous gradients are large in turbulent flows due The relatively to small-scale fluctuations. smaller. gradients of mean values are For large Re, turbulent scalar transport is not dependent on D. Both conditional and unconditional means

smooth functions of coordinates. This is reflected in estimates $\nabla < Y > Y > Y$ are $\nabla < Y_1 | Y_2 = Z_2 > Y_1$. (It should be emphasized that these estimates are and quite approximate and reflect only the dependence on or independence from It will be demonstrated in Sec.5 that the study of more refined Re. properties of some turbulent flows indicate that $\nabla < Y_2$ and $\nabla < Y_1 | Y_2 = Z_2$ can be of different orders.) The unconditional mean value of the gradient ∇Y $\langle \nabla Y \rangle = \nabla \langle Y \rangle \sim Y.$ can be estimated by Estimation of the conditional expectation of the gradients estimation is simple of not as as unconditional expectations since, according to Eq.(2.36), $<\nabla Y_1 | Y_2 = Z_2 >$ is conditional necessarily equal $\nabla < Y_1 | Y_2 = Z_2 >$. The order of the not to expectation $\langle \nabla Y_1 | Y_2 = Z_2 \rangle$ can not exceed the order of instantaneous gradient ∇Y_1 so that $\langle \nabla Y_1 | Y_2 = Z_2 \rangle \langle \nabla Y_1 \sim Y_1 D^{-1/2}$.

The estimates of gradients of scalars and their functions in turbulent flows at high Re can be summarized in the following simple rules :

1) The gradients of conditional and unconditional expectations have dependence on D similar to the expectations themselves.

2) An unconditional expectation of a gradient is identical to the gradient of the unconditional expectation $\langle \nabla Y \rangle = \nabla \langle Y \rangle$.

3) A conditional expectation of the gradient of a scalar variable or its function can increase the order of the variable or function by $\leq D^{-1/2}$.

We consider first the case of D=const, ρ =const. Since $\nabla(\rho D)=0$, we need to estimate only the last term on the left-hand side of Eq.(2.46). This term can be written as $\rho D \nabla^2 P$ and estimated by $\rho D \nabla^2 P \sim \rho D P$. If D is small (Re is large), this term is small and can be neglected. Let us consider the more complicated case when ρ and D are not constants and two the left-hand estimate the last terms on side of Eq.(2.46). According to Rule 1, the Laplace operator applied to an averaged value does not increase the order so that the term $\nabla^2 (\langle \rho D | \mathbf{Y} = \mathbf{Z} \rangle P) \sim \langle \rho D | \mathbf{Y} = \mathbf{Z} \rangle P$ ~ ρD can be neglected. Let us estimate the next term div($\langle \nabla(\rho D) | \mathbf{Y} = \mathbf{Z} \rangle P$). The divergence operator is applied to the average value and so does not increase the order of the term. Gradients inside the conditional brackets should be estimated as explained above so that $div(\langle \nabla(\rho D) | \mathbf{Y} = \mathbf{Z} \rangle) \leq \nabla(\rho D) \sim$ $0D^{1/2}$ and this is negligible at high Re. Now we retain only the most significant terms in Eq.(2.32) so that

$$\frac{\partial}{\partial Z_{i}} \left(\langle \operatorname{div}(\rho D \nabla Y_{i}) | \mathbf{Y} = \mathbf{Z} \rangle P \right) = \frac{\partial^{2}}{\partial Z_{i} \partial Z_{j}} \left(\langle \rho D (\nabla Y_{i} \cdot \nabla Y_{j}) | \mathbf{Y} = \mathbf{Z} \rangle P \right)$$
(2.47)

This equation transforms Eq.(2.45) into the form

$$\frac{\partial \langle \rho | \mathbf{Y} = \mathbf{Z} \rangle P}{\partial t} + \operatorname{div} \left(\langle \rho \mathbf{v} | \mathbf{Y} = \mathbf{Z} \rangle P \right) + \frac{\partial^2}{\partial Z_i \partial Z_j} \left(\langle \rho D(\nabla Y_i \cdot \nabla Y_j) | \mathbf{Y} = \mathbf{Z} \rangle P \right) =$$
$$= - \frac{\partial}{\partial Z_i} \left(\langle W_i \rho | \mathbf{Y} = \mathbf{Z} \rangle P \right)$$
(2.48)

This form of the joint pdf equation is used extensively in the derivation of the CMC equation. This equation has n+4 independent variables and involves the conditional expectations of velocity and dissipation tensor $D(\nabla Y_i \cdot \nabla Y_i)$. These terms are unclosed. Equation (2.48) is quite universal. of governs the probability density function any scalar It values satisfying equations which can be written in the form of Eq.(2.38) with D_i = D.

Equation (2.47) is often used in pdf modeling. Let us also prove that

$$<\operatorname{div}(\rho D\nabla Y_{i})|\mathbf{Y}=\mathbf{Z}>P = \frac{\partial}{\partial Z_{j}}\left(<\rho D(\nabla Y_{i}\cdot\nabla Y_{j})|\mathbf{Y}=\mathbf{Z}>P\right)$$

$$(2.49)$$

This equation is not a mere integral of Eq.(2.47). Equation (2.49) being integrated over Z_i involves the arbitrary rotational vector function α_i which obeys $\partial \alpha_i / \partial Z_i = 0$ (note that vector $\langle \operatorname{div}(\rho D \nabla Y_i) | \mathbf{Y} = \mathbf{Z} \rangle P$ is not uniquely determined by its by divergence $\partial (\langle \operatorname{div}(\rho D \nabla Y_i) | \mathbf{Y} = \mathbf{Z} \rangle P) / \partial Z_i)$. Let us estimate side of Eq.(2.33). the left-hand Similar previous the term on to estimations we have $\operatorname{div}(\langle \rho D \nabla Y_i | \mathbf{Y} = \mathbf{Z} \rangle P) \leq \rho D \nabla Y_i \sim \rho D^{1/2}$. This term can be neglected and this proves Eq.(2.49).

2.4. The Conserved Scalar Pdf Equation and its Boundary Conditions

In this section we consider the pdf of a conserved scalar ξ which is not involved in chemical reactions and satisfies Eq.(2.38) with zero source term

$$\rho \frac{\partial \xi}{\partial t} + \rho \mathbf{v} \cdot \nabla \xi - \operatorname{div} \left(\rho D \nabla \xi \right) = 0$$
(2.50)

In practice, ξ can be the mixture fraction defined in section 1.2. The

sample space variable which corresponds to ξ is denoted by η . The pdf $P(\eta)$ is governed by the equation

$$\frac{\partial <\rho |\xi=\eta>P(\eta)}{\partial t} + \operatorname{div}\left(<\rho \mathbf{v} |\xi=\eta>P(\eta)\right) = -\frac{\partial^2 <\rho N |\xi=\eta>P(\eta)}{\partial \eta^2}$$
(2.51)

where $N \equiv D(\nabla \xi)^2$. This equation is a direct consequence of Eq.(2.48).

In this section we consider some conditions for the pdf $P(\eta)$ (specifically for the product $\langle \rho N | \xi = \eta \rangle P(\eta)$) at the bounds of the pdf, η_{\min} and η_{max} . It is assumed the conserved scalar pdf takes positive values only inside the closed interval $\eta_{min} \leq \eta \leq \eta_{max}$. We consider here the boundary one-variable scalar conditions for the conserved pdf only. These conditions will be utilized later. The technique developed in this section is, however, quite general and can be used in more complicated cases. It is assumed in Eq.(2.51) that the Reynolds number is large. In this section we will consistently keep this assumption and neglect macro-transport by molecular diffusion. Investigation of the boundary conditions for pdfs in of moderate Re is considered by Klimenko and Bilger⁵⁹. the case As outlined in Section 1.2 different types of pdfs are in current use in turbulence modeling. Examples include clipped Gaussian pdfs (Fig. 2) which are inherently intermittent and beta function pdfs (Fig. 3) which do involve delta functions at These types not η_{min} and η_{max} . can have different boundary conditions and need separate consideration. Furthermore, we include the consideration of η_{min} and η_{max} varying in space and time.

In order to obtain the boundary conditions we derive the transport equation for the function $F(\xi)$ which is assumed to be an arbitrary good function. By using Eq.(2.50) and chain differentiation of $F(\xi) = F\left[\xi(\mathbf{x},t),\mathbf{x},t\right]$, we obtain

$$\rho \frac{\partial F}{\partial t} + \rho \left(\mathbf{v} \cdot \nabla F \right) - \operatorname{div} \left(\rho D \nabla F \right) = \rho N F''$$
(2.52)

where

$$F'' \equiv \frac{\partial^2 F(\xi)}{\partial \xi^2}$$
; $N \equiv D(\nabla \xi)^2$

Equation (2.52) is averaged with the use of the continuity equation and the transport by molecular diffusion div $\langle \rho D \nabla F \rangle$ is neglected
$$\frac{\partial \langle \rho F \rangle}{\partial t} + \operatorname{div}\left(\langle \rho v F \rangle\right) = - \langle \rho N F'' \rangle$$
(2.53)

2.4.1. Smooth pdfs

pdfs any generalized components, These do not have e.g. delta functions. The transport equation for $\langle F \rangle$ can be also derived from the equation. Equation (2.51) is multiplied by $F(\eta)$ and integrated over pdf the interval $\eta_1 \le \eta \le \eta_2$ where $\eta_{\min} < \eta_1 < \eta_2 < \eta_{\max}$. (Note that η and $F(\eta)$) are independent of t and \mathbf{x} and hence $F(\eta)$ can be taken inside the derivatives.) The term on the right-hand side of Eq.(2.51) is integrated by parts

$$\int_{\eta_{1}}^{\eta_{2}} \frac{\partial \langle \rho | \xi = \eta \rangle FP(\eta)}{\partial t} d\eta + \int_{\eta_{1}}^{\eta_{2}} \operatorname{div} \left\{ \langle \rho \mathbf{v} | \xi = \eta \rangle FP(\eta) \right\} d\eta = - \int_{\eta_{1}}^{\eta_{2}} F'' \langle \rho N | \xi = \eta \rangle P(\eta) d\eta - \left[\frac{\partial \langle \rho N | \xi = \eta \rangle P(\eta)}{\partial \eta} F \right]_{\eta_{1}}^{\eta_{2}} + \left[\langle \rho N | \xi = \eta \rangle P(\eta) F' \right]_{\eta_{1}}^{\eta_{2}}$$

If η_1 and η_2 depend on t and x, the first two integrals on the left-hand side are transformed according to

$$\frac{\partial}{\partial t} \int_{\eta_{1}}^{\eta_{2}} \langle \rho | \xi = \eta > FP(\eta) d\eta = \int_{\eta_{1}}^{\eta_{2}} \frac{\partial \langle \rho | \xi = \eta > FP(\eta)}{\partial t} d\eta + \left[\frac{\partial \eta_{i}}{\partial t} \langle \rho | \xi = \eta > FP(\eta) \right]_{\eta_{1}}^{\eta_{2}}$$
$$\operatorname{div} \left(\int_{\eta_{1}}^{\eta_{2}} \langle \rho \mathbf{v} | \xi = \eta > FP(\eta) d\eta \right) = \int_{\eta_{1}}^{\eta_{2}} \operatorname{div} \left(\langle \rho \mathbf{v} | \xi = \eta > FP(\eta) \right) d\eta + \left[\nabla \eta_{i} \langle \rho \mathbf{v} | \xi = \eta > FP(\eta) \right]_{\eta_{1}}^{\eta_{2}}$$

where η_i is for either η_1 or η_2 . These equations yield

$$\frac{\partial}{\partial t} \int_{\eta_{1}}^{\eta_{2}} \langle \rho | \xi = \eta > FP(\eta) d\eta + \operatorname{div} \left(\int_{\eta_{1}}^{\eta_{2}} \langle \rho \mathbf{v} | \xi = \eta > FP(\eta) d\eta \right) = - \int_{\eta_{1}}^{\eta_{2}} F'' \langle \rho N | \xi = \eta > P(\eta) d\eta + \left[\left(\frac{\partial \eta_{i}}{\partial t} \langle \rho | \xi = \eta > + \nabla \eta_{i} \langle \rho \mathbf{v} | \xi = \eta > \right) P(\eta) F - \frac{\partial \langle \rho N | \xi = \eta > P(\eta)}{\partial \eta} F \right]_{\eta_{1}}^{\eta_{2}} +$$

+
$$\left[<\rho N | \xi = \eta > P(\eta) F' \right]_{\eta_1}^{\eta_2}$$
 (2.54)

Assuming $\eta_1 = \eta_{min} + \varepsilon$ and $\eta_2 = \eta_{max} - \varepsilon$ and taking the limit $\varepsilon \rightarrow 0$ (or $\eta_1 \rightarrow \eta_{min}$ $\eta_2 \rightarrow \eta_{max}$ for infinite η_{min} and η_{max}) we obtain

$$\frac{\partial \langle \rho F \rangle}{\partial t} + \operatorname{div}\left(\langle \rho v F \rangle\right) = - \langle \rho N F'' \rangle + \left[J_{P} F \right]_{\min}^{\max} + \left[\langle \rho N | \xi = \eta \rangle P(\eta) F' \right]_{\min}^{\max}$$
(2.55)

where

$$J_{P} \equiv \langle \rho | \xi = \eta \rangle P(\eta) \frac{d\eta_{m}}{dt} - \frac{\partial \langle \rho N | \xi = \eta \rangle P(\eta)}{\partial \eta}$$

$$\frac{d\eta_{m}}{dt} \equiv \frac{\partial \eta_{m}}{\partial t} + \frac{\langle \rho v | \xi = \eta \rangle}{\langle \rho | \xi = \eta \rangle} \cdot \nabla \eta_{m}$$

$$F' \equiv \frac{\partial F(\eta)}{\partial \eta} ;$$

$$\left[\cdot \right]_{\min}^{\max} \equiv \left[\cdot \right]_{\max} - \left[\cdot \right]_{\min} ; \qquad \left[\cdot \right]_{m} \equiv \lim_{\eta \to \eta_{m}} \left(\cdot \right)$$

$$(2.56)$$

and index "m" is for either "min" or "max".

Equations (2.53) and (2.55) must be identical for any function F. This specifies the boundary conditions required

 $J_{\rm P} \rightarrow 0 \quad \text{as} \quad \eta \rightarrow \eta_{\rm m}$ (2.57)

$$<\rho N | \xi = \eta > P(\eta) \rightarrow 0 \text{ as } \eta \rightarrow \eta_m$$
 (2.58)

In deriving Eqs.(2.57) and (2.58) it has been consistently assumed that molecular $\operatorname{Re} \rightarrow \infty$. In general, the question of neglecting transport by the vicinity of η_{min} and η_{max} is quite problematical since diffusion in the pdf structure near these bounds may depend on D. If η_{min} and η_{max} are molecular if not constants and the transport by diffusion is not terms⁵⁹. neglected, additional Eq.(2.56) involves some

As discussed in detail in Section 4, it can be quite important that the behavior of the pdf and conditional dissipation of the conserved scalar at the bounds of η are properly modeled. In order to assess this behavior we use Eq.(2.54) and assume $\eta_1 \rightarrow \eta_{min}$ and $\eta_{min} < \eta_2 < \eta_{max}$. Equation (2.54) takes the form

$$\frac{\partial}{\partial t} \int_{\min n}^{\eta_2} |\xi = \eta > FP(\eta) d\eta + \operatorname{div} \left(\int_{\min n}^{\eta_2} |\varphi = \eta > FP(\eta) d\eta \right) + \int_{\min n}^{\eta_2} F'' < \rho N |\xi = \eta > P(\eta) d\eta =$$
$$= \left[J_P F \right]_{\min n}^{\eta_2} + \left[< \rho N |\xi = \eta > P(\eta) F' \right]_{\min n}^{\eta_2} = \left[J_P F \right]_{\eta_2} + \left[< \rho N |\xi = \eta > P(\eta) F' \right]_{\eta_2}^{\eta_2}$$

Here the boundary conditions in Eqs.(2.57) and (2.58) are taken into account. The integrals on the left-hand side of this equation tend to zero as $\eta_2 \rightarrow \eta_{min}$. If $\eta_2 = \eta$ (η is close to η_{min} but $\eta \neq \eta_{min}$) we suggest that these integrals can be roughly estimated by the simple integral

$$\int_{\min}^{\eta} FP(\eta^{\circ})d\eta^{\circ}$$

which yields

$$\int_{\min} FP(\eta^{\circ}) d\eta^{\circ} \sim J_{P}F + \langle \rho N | \xi = \eta \rangle P(\eta)F' \text{ as } \eta \neq \eta_{\min}$$

A similar equation is valid for $\eta \rightarrow \eta_{max}$. Assuming power-law asymptotes

$$P(\eta) \sim |\eta - \eta_m|^{\alpha}, \langle \rho N | \xi = \eta \rangle \sim |\eta - \eta_m|^{\beta} \text{ as } \eta \rightarrow \eta_m$$

where α is always greater than -1 (otherwise $P(\eta)$ can not be integrated), we obtain using Eq. (2.56)

$$\left|\eta - \eta_{m}\right|^{\alpha+1} \sim \frac{d\eta_{m}}{dt} \left|\eta - \eta_{m}\right|^{\alpha} + \left|\eta - \eta_{m}\right|^{\alpha+\beta-1} + \left|\eta - \eta_{m}\right|^{\alpha+\beta} \text{ as } \eta \rightarrow \eta_{m}$$
(2.59)

Equating the power-law dependencies of the first and the second terms on the right-hand side, since these are the largest terms, we obtain $\beta=1$,

$$\langle \rho N | \xi = \eta \rangle \sim | \eta - \eta_m |$$
 as $\eta \rightarrow \eta_m$ (2.60)

2.4.2. Smooth pdfs with fixed bounds

In many cases the conserved scalar pdfs are assumed to have fixed bounds (that is η_{min} and η_{max} are constants). In this case the boundary conditions (2.57) and (2.58) take the forms

$$\frac{\partial \langle \rho N | \xi = \eta \rangle P(\eta)}{\partial \eta} \to 0 \text{ as } \eta \to \eta_{m}$$
(2.61)

$$\langle \rho N | \xi = \eta > P(\eta) \rightarrow 0 \quad \text{as} \quad \eta \rightarrow \eta_m$$
 (2.62)

If the pdf bounds are infinite these conditions are quite obvious. Let us estimate β . Assuming $d\eta_m/dt=0$ in Eq.(2.59) we obtain $\alpha+\beta-1=\alpha+1$ and

$$\langle \rho N | \xi = \eta \rangle \sim |\eta - \eta_m|^2 \text{ as } \eta \to \eta_m$$
 (2.63)

Note that the estimates in Eqs.(2.60) and (2.63) are rough and the exact some weaker functions (for example asymptote can involve of $|\eta - \eta_m|$ $\ln |\eta - \eta_m|$). For the special case of a Gaussian pdf it is known⁷ that the conditional scalar dissipation is independent of η in uniform density its unconditional value. Figure 9 demonstrates the nce equal to qualitative shapes of pdfs and corresponding conditional dissipation for three different cases considered above.

2.4.3. Intermittent pdfs

Intermittency is conventionally associated with the presence or not of vorticity fluctuations in the fluid. Here we are going to associate it with the presence or absence of the scalar. We shall for simplicity of the flow is like that of an initially language assume that nonturbulent nonturbulent surrounding fluid. The diffusion gas jet into a of vorticity the nonturbulent fluid is assumed to be accompanied by diffusion of into

In such intermittent flows the contaminated turbulent the scalar. spots alternate stochastically with uncontaminated nonturbulent spots. For two-stream mixing with initial values of $\xi = \eta_{min}$ in first stream and $\xi = \eta_{max}$ in the second stream, the intermittent pdfs involve delta functions:

$$P(\eta) = \gamma_{\min}\delta(\eta - \eta_{\min}) + \gamma_t P_t(\eta) + \gamma_{\max}\delta(\eta - \eta_{\max})$$
(2.64)

where $\gamma_{\min} + \gamma_t + \gamma_{\max} = l$ and γ_t is called the intermittency factor. The components delta function of the pdf correspond to the vortex-free uncontaminated regions. these regions the values of In the conserved scalar are constants (either $\xi = \eta_{min}$ or $\xi = \eta_{max}$). The values of η_{min} and The component P_t is smooth and corresponds to are constants. the η_{max} fully turbulent region The dissipation where $\eta_{min} < \eta < \eta_{max}$. in the vortex-free regions is zero N = 0 at $\eta = \eta_m$ so that

$$\langle \rho N | \xi = \eta \rangle P(\eta) = \gamma_t \langle \rho N | \xi = \eta \rangle P_t(\eta)$$
(2.65)

where η_m is for either η_{min} or η_{max} . Note that P=0 and $P_t=0$ for $\eta < \eta_{min}$ and for $\eta > \eta_{max}$.

has been derived as an equation The pdf equation (2.51) for the expectation of the generalized function ψ and it is still valid for an pdf. We treat Eq.(2.51) equation for intermittent as an generalized functions P which is valid for all η (- $\infty < \eta < +\infty$). We multiply Eq.(2.51) by an arbitrary good function $F(\eta)$ and integrate it over all η . This yields

$$\frac{\partial \langle \rho F \rangle}{\partial t} + \operatorname{div}\left(\langle \rho v F \rangle\right) = \int_{-\infty}^{+\infty} \frac{\partial^2 \langle \rho N | \xi = \eta \rangle P(\eta)}{\partial \eta^2} F(\eta) d\eta = \int_{-\infty}^{+\infty} \langle \rho N | \xi = \eta \rangle P(\eta) F'' d\eta = \langle \rho N F'' \rangle$$
(2.66)

significant difference derivations Eqs.(2.55) There is in the of and (2.66).the derivation of Eq.(2.55) the pdf equation (2.51) is treated In as an equation for ordinary function and the integral is taken over the (where which excludes any possible interval $\eta_1 \leq \eta \leq \eta_2$ $\eta_{\min} < \eta_1 < \eta_2 < \eta_{\max}$) singularity at the bounds. The integral in Eq.(2.66) is taken over all η . The product $\langle \rho N | \xi = \eta \rangle P(\eta)$, as it is indicated in Eq.(2.65), does not have generalized component, but its second derivative could have any such components. The modification of the integral on the right-hand side of Eq.(2.66) seems to be an integration by parts but this modification rules of operating with generalized functions represents the (see Eq.(2.20)). Equation (2.66) is consistent with Eq.(2.53).

The product $\langle \rho N | \xi = \eta \rangle P(\eta)$ represents an ordinary, smooth function and we are interested in determining its limits $\eta \rightarrow \eta_m$. The integral in Eq.(2.66) can be modified

$$\int_{-\infty}^{+\infty} \langle \rho N | \xi = \eta \rangle P(\eta) F'' d\eta = \int_{\eta_{\min}}^{\eta_{\max}} \langle \rho N | \xi = \eta \rangle P(\eta) F'' d\eta = \int_{\eta_{\min}}^{\eta_{\max}} \frac{\partial^2 \langle \rho N | \xi = \eta \rangle P(\eta)}{\partial \eta^2} F(\eta) d\eta + \left[\langle \rho N | \xi = \eta \rangle P(\eta) F' \right]_{\min}^{\max} - \left[\frac{\partial \langle \rho N | \xi = \eta \rangle P(\eta)}{\partial \eta} F \right]_{\min}^{\max}$$
(2.67)

The notations are the same as in Eq.(2.55). We substitute Eqs.(2.64) and (2.67) into Eq.(2.66). The function F is an arbitrary function which can have the arbitrary values of $F(\eta_{\min})$, $F(\eta_{\max})$, $F'(\eta_{\min})$ and $F'(\eta_{\max})$. The function F is a continuous function so that $F(\eta_m)=[F]_m$. Equating the terms which are multiplied by $F'(\eta_{\min})$, $F'(\eta_{\max})$, $F(\eta_{\min})$ and $F(\eta_{\max})$ we obtain

$$\langle \rho N | \xi = \eta > P(\eta) \rightarrow 0 \text{ as } \eta \rightarrow \eta_{\min}$$
 (2.68)

$$<\rho N|\xi=\eta>P(\eta) \rightarrow 0 \text{ as } \eta \rightarrow \eta_{\max}$$
 (2.69)

$$\frac{\partial \gamma_{\min} < \rho |\xi = \eta_{\min} >}{\partial t} + \operatorname{div} \left(\gamma_{\min} < \rho \mathbf{v} |\xi = \eta_{\min} > \right) = - \left[\frac{\partial < \rho N |\xi = \eta > P(\eta)}{\partial \eta} \right]_{\min}$$
(2.70)

$$\frac{\partial \gamma_{\max} < \rho |\xi = \eta_{\max} >}{\partial t} + \operatorname{div} \left(\gamma_{\max} < \rho \mathbf{v} |\xi = \eta_{\max} > \right) = \left[\frac{\partial < \rho N |\xi = \eta > P(\eta)}{\partial \eta} \right]_{\max}$$
(2.71)

It can be seen that the product $\langle \rho N | \xi = \eta \rangle P(\eta)$ has zero limit at $\eta \rightarrow \eta_m$ but the limit of its derivative is not necessarily zero. Kuznetsov and Sabelnikov⁷² assuming carried out a similar analysis that $< N | \xi = \eta >$ has This is an additional assumption which is non-zero limit at $\eta \rightarrow \eta_m$. not properties Eq.(2.51). additional determined by the of This assumption results in $P(\eta) \rightarrow 0$ as $\eta \rightarrow \eta_m$.

2.5 Summary of the Major Results

The techniques, which have been systematically introduced in this section, can be used for derivation of the equations and transport various pdfs (probability density identities for functions) in turbulent are extensively used in conditional flows. These equations methods. The techniques based manipulations with the delta function and are on its The delta derivatives. function is a generalised function and often needs This present initial difficulties special treatment. may some for have much experience field. We think, someone who does not in this rewarding that overcoming these initial difficulties should be however. since the technique is powerfull and, at the same time, is relatively simple to use in partical derivations. Here, we provide a summary of the main results obtained in this section.

The transport equation for the multivariate scalar pdf $P(Z_1, Z_2, ..., Z_n)$ forms Eq.(2.45)has two equivalent and Eq.(2.46). The first of these forms is most commonly used in pdf methods. The second form, Eq.(2.46), becomes Eq.(2.48) at high Reynolds numbers and this is the form that will be used in the next section (see that which follows Eq.(3.3)) for deriving the first-order CMC equation the pdf method. It is by also used in Sections 8 and 9 for derivation of the second-moment (see that which follows Eq.(8.6)) and doubly-conditional (see which that follows Eq.(9.27)) CMC equations by the pdf approach. At high Reynolds numbers the transport equation for the pdf of a single conserved scalar is simplified into Eq.(2.51) and this is widely used in CMC methods in the form of Eq.(3.15). It is to be noted that, in these equations, some terms involving the diffusion coefficient, D, are neglected when the Reynolds number is large. Simplified rules for determining which terms can be are listed in section 2.3.3. These approximations also neglected are the closely related result of Eq.(2.49), involved in derivation of that is be very useful in modelling differential diffusion effects (see found to Eq.(9.16)). The transport equation for the fine-grained joint pdf. CMC Eq.(2.43) is the preferred starting point for derivation of the the presence of differential diffusion Eq.(9.3)) equation in (see and is deriving also used in an alternative approach to the first-order CMC equation as is presented in section 3.1.2.

formulae for conditional averages, Eq.(2.15) Basic and Eq.(2.18), are for the derivation of the first-order CMC needed equations (Eqs(3.3 and higher-order forms Sections (3.4))and in 8 and 9.2.1. Conditional

of functions, Eq.(2.17), is widely used in the formulation of averaging the decomposition approach to the derivation of the CMC equation (see that preceding Eq.(3.20)) and in the development of primary closure hypotheses and (3.30)). It is also used in exploring the relationship Eqs(3.11)(see between CMC and other models such as frozen and fast chemistry (section 4.3) and flamelets (section 4.4). The warning that differentiation and conditional averaging do not usually commute, as shown in Eq.(2.36), important in formulating the primary closure is hypothesis when deriving the CMC equations by the decomposition approach (see section 3.2.2). Equation (2.36) is also needed in showing the equivalence of the primary closure hypotheses by the various approaches (see section 3.3).

Conserved-scalar pdfs used in applications can be broadly divided into two groups: intermittent pdfs and smooth pdfs. The former involve one or two generalized components at the bounds of the pdf which correspond to the non-turbulent spots in a turbulent flow. As shown in section 2.4 the most common boundary conditions for the pdf are given by $\langle N|\xi=\eta \rangle P(\eta) \rightarrow 0$ as η tends to its maximum and minimum values. In some cases, as shown in section 2.4.2, the zero condition can be also imposed on the derivative These conditions $\partial < N | \xi = \eta > P(\eta) / \partial \eta$. boundary are important in determining conditional scalar dissipation from the transport equation for the the pdf (sections 6.1 conserved-scalar 4.1, 5.6, and 7.2) and the closely related question of preservation of the conservation integrals (sections 4.2, 5.4, and 5.5).

3. THE CMC EQUATION

Section we consider derivations of the In this equation for the conditional mean $Q \equiv \langle Y | \xi = \eta \rangle \equiv \langle Y | \eta \rangle$ and the basic closure hypotheses which lead us to the model for first-order Conditional Moment Closure (CMC). Here the variable Y represents a reactive scalar, that is a scalar which can be involved in chemical reactions. For simplicity, and without loss of generality, we drop the subscript i used in Eq. (1.1). Usually, several reactive scalars Y_1, \dots, Y_n are involved: Y represents any of them and the CMC model can be written for each of the scalars. The variable ξ denotes a conserved scalar for which its transport equation has zero source term. The model considered in this section is an effective tool for non-premixed combustion in turbulent flows with two-stream mixing (usually fuel injected into oxidizer). The best, physically sound choice of the scalar ξ is the mixture fraction which indicates conserved the frozen (without chemical reactions) mass fraction of fuel in the fuel-oxidizer Section the molecular diffusion coefficients are assumed mixture. In this to be the same for both scalars ξ and Y. The transport equations (2.38) can be written for ξ and Y as

$$\rho \frac{\partial \xi}{\partial t} + \rho \mathbf{v} \cdot \nabla \xi - \operatorname{div} \left(\rho D \nabla \xi \right) = 0$$
(3.1)

$$\rho \frac{\partial Y}{\partial t} + \rho \mathbf{v} \cdot \nabla Y - \operatorname{div} \left(\rho D \nabla Y \right) = \rho W$$
(3.2)

It is noted that, in general, W will depend on ξ and this means that Y will not be independent of ξ and $\langle Y|\eta \rangle$ will differ from its unconditional value $\langle Y \rangle$.

are different approaches to the derivation of the first-order There independently suggested by Klimenko⁵⁴ and $Bilger^{10,12}$. as CMC equation different different These approaches use mathematical methods and modelling assumptions but yield the same result for the CMC equation. They give different insights into the nature of the CMC equation and into the assumptions in its formulation. Both primary closure used approaches get a full appreciation of the rigour should be understood to of the of the CMC equation and the transparency of formulation the primary closure assumptions made. In Section 3.1 we follow Klimenko⁵⁴ and use the joint pdf of the reactive and conserved scalar as has been derived in Section 2. In Section 3.2 we follow $Bilger^{10,12}$ and derive the equation by

considering the reactive scalar to be decomposed into its conditional average for the corresponding value of the mixture fraction and a fluctuation from that. This derivation does not heavily the rely on results of Section 2 and may be more accessible to newcomers to CMC. The closure assumptions used in these derivations primary are compared and discussed in Section 3.3.

In flows with heat release it is necessary to consider the energy Section 3.4 CMC equation. presents the first-order equation for the conditional average enthalpy.

Apart from the primary closure hypothesis, the next most important issue for closure of the CMC equation is the closure of the conditional chemical raction Basic considerations average rate term. regarding the conditional average reaction rate and radiation source are outlined term in Section 3.5. Higher-order closure using second conditional moments is left until Section 8. The modeling of other unclosed terms is left to Section 4.

For simplicity, conditional correlations between density and other scalars are neglected in the main treatment of the theory. In situations with fluctuations about the conditional heat release and large means, correlations could be significant. Section these 3.6 outlines how conditional Favre averaging can be used in these problems.

A broader discussion of the physical bases for the closure hypotheses approaches may be found in Section 3.7. used in these This discussion involves the theory of Markov processes and uses concepts which are more complex than the simpler level of explanation adopted Sections in 3.1-3.6.

The Section closes with a summary of the major results obtained.

3.1. The Joint Pdf Method

3.1.1. The unclosed equation

The relationship of the conditional expectation Q, the joint pdf $P(Z,\eta)$ (where Z is the sample space variable for Y) and the pdf $P(\eta)$ is given by Eq.(2.15)

$$Q P(\eta) \equiv \langle Y | \xi = \eta \rangle P(\eta) = \int_{-\infty}^{+\infty} Z P(Z,\eta) dZ$$
(3.3)

We utilize the joint pdf equation (2.48) which is valid for the case of large Re and consider this equation as the equation for $P(Z,\eta)$. We put in Eq.(2.48) $Z=Z_1$, $\eta=Z_2$, $W=W_1$, $W_2=0$, n=2 (*n* is the dimension of the vector $\mathbf{Z}=(Z_1,...,Z_n)$).

$$\frac{\partial \langle \rho | \mathbf{Y} = \mathbf{Z} \rangle P}{\partial t} + \operatorname{div} \left(\langle \rho \mathbf{v} | \mathbf{Y} = \mathbf{Z} \rangle P \right) + \frac{\partial^2}{\partial Z_i \partial Z_j} \left(\langle \rho D(\nabla Y_i \cdot \nabla Y_j) | \mathbf{Y} = \mathbf{Z} \rangle P \right) =$$
$$= - \frac{\partial}{\partial Z_1} \left(\langle W_1 \rho | \mathbf{Y} = \mathbf{Z} \rangle P \right)$$

This equation is multiplied by Z and integrated over all Z. It can be noted that since Z is an independent variable it can be taken inside derivatives with respect to t, x and η . The terms which involve $\partial/\partial Z$ are integrated by parts

$$\int_{-\infty}^{+\infty} Z \frac{\partial^{2}}{\partial Z \partial \eta} \Big(\langle \rho D(\nabla Y \cdot \nabla \xi) | Y = Z, \xi = \eta \rangle P \Big) dZ = \left[Z \frac{\partial}{\partial \eta} \Big(\langle \rho D(\nabla Y \cdot \nabla \xi) | Y = Z, \xi = \eta \rangle P \Big) \right]_{Z = -\infty}^{Z = +\infty} - \frac{\partial}{\partial \eta} \int_{-\infty}^{+\infty} \Big(\langle \rho D(\nabla Y \cdot \nabla \xi) | Y = Z, \xi = \eta \rangle P \Big) dZ = - \frac{\partial}{\partial \eta} \Big(\langle \rho D(\nabla Y \cdot \nabla \xi) | \xi = \eta \rangle P \Big)$$

and

$$\int_{-\infty}^{+\infty} Z \frac{\partial^2}{\partial Z^2} \Big(\langle \rho D(\nabla Y \cdot \nabla Y) | Y = Z, \xi = \eta \rangle P \Big) dZ = \left[Z \frac{\partial}{\partial Z} \Big(\langle \rho D(\nabla Y \cdot \nabla Y) | Y = Z, \xi = \eta \rangle P \Big) \right]_{Z=-\infty}^{Z=+\infty}$$
$$\left[\langle \rho D(\nabla Y \cdot \nabla Y) | Y = Z, \xi = \eta \rangle P \right]_{Z=-\infty}^{Z=+\infty} = 0$$

Note that $P(Z,\eta) \rightarrow 0$ as $Z \rightarrow \pm \infty$. The result of the integration is

$$\frac{\partial \rho_{\eta} Q P(\eta)}{\partial t} + \operatorname{div} \left(\rho_{\eta} < \mathbf{v} Y | \eta > P(\eta) \right) = \rho_{\eta} < W | \eta > P(\eta) + \frac{\partial J_{Y}}{\partial \eta}$$
(3.4)

where

$$J_{\rm Y} \equiv 2\rho_{\eta} < D(\nabla Y \cdot \nabla \xi) |\eta > P(\eta) - \frac{\partial \rho_{\eta} < NY |\eta > P(\eta)}{\partial \eta}$$
(3.5)

Here $\langle \cdot | \eta \rangle$ is short for $\langle \cdot | \xi = \eta \rangle$; $N \equiv D(\nabla \xi)^2$ is the dissipation of the conserved scalar; and subscript " η " also indicates conditional averaging: e.g. $\rho_{\eta} \equiv \langle \rho | \eta \rangle$. Equation (2.15) is repeatedly used in the derivation of Eq.(3.4). Note that the density fluctuations about the conditional mean are not taken into account here. This will be discussed in Sec.3.6.

Equation (3.4) has five independent variables: t, x_1, x_2, x_3 and η . The physical meaning of this equation is quite obvious since there is a of similarity with unconditionally certain degree the averaged scalar transport equation. The second term on the left-hand side of Eq.(3.4)corresponds to the convective terms conditional on η . The first term on right-hand side of Eq.(3.4) represents the conditional expectation of the Transport in physical space by molecular diffusion reactive source term. is neglected since Re is large, but it would be incorrect to say that all molecular diffusion effects are neglected in Eq.(3.4). The new term $\partial J_{\rm Y}/\partial \eta$ determined small-scale diffusion is by processes. The variable J_{Y} in diffusive Eq.(3.5) specifies the net flux of reactive Y the scalar in conserved scalar space.

3.1.2. Alternative derivation of the unclosed equation

The derivation given above for Eqs.(3.4) and (3.5) is not the only one possible. Here we consider an alternative derivation of Eqs.(3.4) and (3.5). In general, both techniques are equivalent but in some cases the alternative derivation can be more convenient. First we derive the governing equation for $\varphi \equiv Y\psi_{\eta}$, where $\psi_{\eta} \equiv \delta(\xi(\mathbf{x},t)-\eta)$, and then average it. The new function ϕ_η can be called the fine-grained density in the x-\eta space. We use Eq.(2.43) as the equation for ψ_{η} ($\eta=Z_1$, $\xi=Y_1$, $W_1=0$, n=1). This equation is multiplied by Y and added to Eq.(3.2) multiplied by ψ_n

$$\frac{\partial \rho \psi_{\eta} Y}{\partial t} + \operatorname{div} \left(\rho \mathbf{v} \psi_{\eta} Y \right) + \frac{\partial \psi_{\eta} Y \operatorname{div} (\rho D \nabla \xi)}{\partial \eta} - \psi_{\eta} \operatorname{div} (\rho D \nabla Y) = \rho \psi_{\eta} W \quad (3.6)$$

We use also Eqs.(2.28) and (2.30) which are written with some minor modifications

$$\operatorname{div}\left(\rho DY \nabla \psi_{\eta}\right) = \frac{\partial^{2}}{\partial \eta^{2}} \left(\psi_{\eta} Y \rho D(\nabla \xi)^{2}\right) - \frac{\partial}{\partial \eta} \left(\psi_{\eta} \operatorname{div}(Y D \rho \nabla \xi)\right)$$
(3.7)
$$= \frac{\partial^{2}}{\partial \eta^{2}} \left(\psi_{\eta} Y \rho D(\nabla \xi)^{2}\right) - \frac{\partial}{\partial \eta} \left(\psi_{\eta} Y \operatorname{div}(\rho D \nabla \xi)\right) - \frac{\partial}{\partial \eta} \left(\psi_{\eta} \rho D(\nabla \xi \cdot \nabla Y)\right)$$

$$\operatorname{div}\left(\psi_{\eta}\rho D\nabla Y\right) = \psi_{\eta}\operatorname{div}\left(\rho D\nabla Y\right) - \frac{\partial}{\partial\eta}\left(\psi_{\eta}\rho D(\nabla\xi\cdot\nabla Y)\right)$$
(3.8)

The two last terms on the left-hand side of Eq.(3.6) are modified using Eqs.(3.7) and (3.8)

$$\frac{\partial \psi_{\eta} \rho Y}{\partial t} + \operatorname{div} \left(\rho \mathbf{v} Y \psi_{\eta} \right) + \frac{\partial^{2}}{\partial \eta^{2}} \left\{ \psi_{\eta} Y \rho D(\nabla \xi)^{2} \right\} - 2 \frac{\partial}{\partial \eta} \left\{ \psi_{\eta} \rho D(\nabla \xi \cdot \nabla Y) \right\} - \operatorname{div} \left(\rho D \nabla (\psi_{\eta} Y) \right) = \psi_{\eta} \rho W$$

$$(3.9)$$

Equation (3.9) is averaged with the use of Eq.(2.18). The term $div \langle \rho D\nabla(\psi_{\eta} Y) \rangle$ is neglected since it is small if Re is large (see Section 2.3.3). The resulting equation is exactly the same as Eqs.(3.4) and (3.5).

3.1.3. Primary closure hypothesis

The flux of the reactive scalar in conserved scalar space, J_Y , is given by Eq.(3.5). This equation is unclosed. We seek a closure for J_Y in the form of a diffusion approximation

$$J_{\rm Y} = AQ + B \frac{\partial Q}{\partial \eta} \tag{3.10}$$

We do not assume any particular form of the drift coefficient A and the diffusion coefficient В but we note that formulas for these coefficients linear properties of turbulent scalar must preserve the transport. This means that A and B are independent of Q. The physical basis of these assumptions is discussed in Section 3.7 where the analogy between particle diffusion in conserved scalar space and a Markov process is established. There is also an analogy with conventional modeling of scalar transport in turbulent flows, where the turbulent flux $\langle vY \rangle$ is approximated by the term $\langle \mathbf{v} \rangle \langle \mathbf{Y} \rangle - D_{\mathbf{t}} \nabla \langle \mathbf{Y} \rangle$. There significant difference: is, however, a the turbulent diffusion coefficient needs further modeling, whereas the $D_{\rm t}$ coefficients A and B are fully determined by a necessary constraint. This if W=0, a=const, b=const and the constraint is: initial and boundary conditions for Y are related to those for ξ by $Y=a+b\xi$, then $Y = a+b\xi$ is a solution of Eq.(3.2) for any velocity field. According to Eq.(2.17) this solution corresponds to $Q=a+b\eta$. By substituting $Y=a+b\xi$ into Eq.(3.5) and taking into account that

$$\frac{\partial(a+b\eta) < N[\eta > P(\eta)\rho_{\eta}}{\partial \eta} = (a+b\eta) \frac{\partial < N[\eta > P(\eta)\rho_{\eta}}{\partial \eta} + b < N[\eta > P(\eta)\rho_{\eta}$$

and that the identities $\langle N(a+b\xi)|\eta\rangle = \langle N|\eta\rangle$ (a+b η) and $\langle D(\nabla(a+b\xi)\cdot\nabla\xi)|\eta\rangle = b\langle N|\eta\rangle$ can be easily derived from Eq.(2.17), we obtain

$$J_{\rm Y} = -(a+b\eta)\frac{\partial\rho_{\eta} < N|\eta > P(\eta)}{\partial\eta} + b\rho_{\eta} < N|\eta > P(\eta)$$
(3.11)

while the substitution of $Q=a+b\eta$ into Eq.(3.10) yields

$$J_{\rm Y} = (a+b\eta)A + bB \tag{3.12}$$

Since Eqs. (3.11) and (3.12) are valid for any arbitrary constants a and b, the coefficients A and B must be determined as the corresponding terms in Eq.(3.11). The closure for J_Y takes the form

$$J_{\rm Y} = - \frac{\partial \rho_{\eta} < N |\eta > P(\eta)}{\partial \eta} Q + \rho_{\eta} < N |\eta > P(\eta) \frac{\partial Q}{\partial \eta}$$
(3.13)

Since the coefficients A and B are independent of Q this closure is, according to assumption (3.10), valid not only for $Q=a+b\eta$ but also for any arbitrary field Q. The substitution of Eq.(3.13) into Eq.(3.4) yields

$$\frac{\partial \rho_{\eta} Q P(\eta)}{\partial t} + \operatorname{div} \left(\rho_{\eta} < \mathbf{v} Y | \eta > P(\eta) \right) = \rho_{\eta} < W | \eta > P(\eta)$$
$$- \frac{\partial^{2} \rho_{\eta} < N | \eta > P(\eta)}{\partial \eta^{2}} Q + \rho_{\eta} < N | \eta > P(\eta) \frac{\partial^{2} Q}{\partial \eta^{2}}$$
(3.14)

This equation is the basic CMC equation which governs the evolution of the conditional expectation Q. It involves the conserved scalar pdf $P(\eta)$ and some conditional expectations $\langle N | \eta \rangle \equiv \langle D(\nabla \xi)^2 | \eta \rangle$, $\langle \mathbf{v}Y | \eta \rangle$ and $\langle W | \eta \rangle$. The pdf equation (2.51) is rewritten using the notation of this section

$$\frac{\partial \rho_{\eta} P(\eta)}{\partial t} + \operatorname{div} \left(\rho_{\eta} < \mathbf{v} | \eta > P(\eta) \right) = - \frac{\partial^2 \rho_{\eta} < N | \eta > P(\eta)}{\partial \eta^2}$$
(3.15)

We have put $\eta = Z_1$ and $W_1 = 0$ and n=1 in Eq.(2.48). The conditional expectations $\langle N | \eta \rangle$, $\langle vY | \eta \rangle$ and $\langle W | \eta \rangle$ need further modeling. This modeling and the basic properties of the CMC equation will be considered later. Equation (3.14) can be written in another equivalent form. Subtracting Q times Eq.(3.15) from Eq.(3.14) we obtain

$$\frac{\partial Q}{\partial t} + \langle \mathbf{v} | \boldsymbol{\eta} \rangle \nabla Q + \frac{\operatorname{div} \left(\rho_{\boldsymbol{\eta}} \langle \mathbf{v}'' Y'' | \boldsymbol{\eta} \rangle P(\boldsymbol{\eta}) \right)}{P(\boldsymbol{\eta}) \rho_{\boldsymbol{\eta}}} - \langle N | \boldsymbol{\eta} \rangle \frac{\partial^2 Q}{\partial \boldsymbol{\eta}^2} = \langle W | \boldsymbol{\eta} \rangle (3.16)$$

where $\mathbf{v}'' \equiv \mathbf{v} \cdot \langle \mathbf{v} | \eta \rangle$ is the velocity fluctuation about its conditional mean. The conserved scalar pdf equation (3.15) is essentially mathematically adjoint to Eq.(3.16). This is not a coincidence since such linkage of the CMC and pdf equations is necessary to preserve conservation integrals. In CMC modeling the CMC and pdf equations must both be satisfied.

3.2. Decomposition Method

In this section we consider an alternative way of deriving the equation for $Q(\eta, \mathbf{x}, t) \equiv \langle Y(\mathbf{x}, t) | \eta \rangle$. The idea of this derivation is based on the decomposition

$$Y(\mathbf{x},t) = Q(\xi(\mathbf{x},t),\mathbf{x},t) + Y''(\mathbf{x},t)$$
(3.17)

where Y'' is the fluctuation with respect to the conditional mean or, more briefly, the conditional fluctuation.

3.2.1. The unclosed equation

Let us apply conditional averaging $\langle \cdot | \eta \rangle$ to Eq.(3.17). According to Eq.(2.17) $\langle Q(\xi, \mathbf{x}, t) | \eta \rangle = Q(\eta, \mathbf{x}, t)$ so that this averaging yields $\langle Y'' | \eta \rangle = 0$. The function $Q(\xi(\mathbf{x}, t), \mathbf{x}, t)$ is a non-random function of variables \mathbf{x} , t and ξ . Variable ξ is a random variable which is also function of \mathbf{x} and t. Let us differentiate Eq.(3.17)

$$\frac{\partial Y}{\partial t} = \frac{\partial Q}{\partial t} + \frac{\partial Q}{\partial \eta} \frac{\partial \xi}{\partial t} + \frac{\partial Y''}{\partial t}$$
(3.18)

$$\nabla Y = \nabla Q + \frac{\partial Q}{\partial \eta} \nabla \xi + \nabla Y''$$
(3.19)

We also consider the transformation of the molecular diffusion term by decomposition (3.17)

$$\operatorname{div}(\rho D\nabla Y) = \operatorname{div}(\rho D\nabla Q) + \frac{\partial Q}{\partial \eta} \operatorname{div}(\rho D\nabla \xi) + \rho D(\nabla \xi)^{2} \frac{\partial^{2} Q}{\partial \eta^{2}} + \rho D\nabla \xi \cdot \nabla \frac{\partial Q}{\partial \eta} + \operatorname{div}(\rho D\nabla Y'')$$
(3.20)

We substitute Eqs.(3.18)-(3.20) into Eq.(3.2) and use Eq.(3.1). Thus we obtain

$$\rho W = \rho \frac{\partial Q}{\partial t} + \rho \mathbf{v} \cdot \nabla Q - \rho N \frac{\partial^2 Q}{\partial \eta^2} - \operatorname{div}(\rho D \nabla Q) - \rho D \nabla \xi \cdot \nabla \frac{\partial Q}{\partial \eta} + \rho \frac{\partial Y''}{\partial t} + \rho \left(\mathbf{v} \cdot \nabla Y'' \right) - \operatorname{div} \left(D \rho \nabla Y'' \right)$$
(3.21)

Taking the conditional expectation of Eq.(3.21), conditional on $\xi(\mathbf{x},t)=\eta$, yields

$$\rho_{\eta} \frac{\partial Q}{\partial t} + \rho_{\eta} \langle \mathbf{v} | \eta \rangle \langle \nabla Q - \rho_{\eta} \langle N | \eta \rangle \frac{\partial^2 Q}{\partial \eta^2} = \rho_{\eta} \langle W | \eta \rangle + e_{Q} + e_{Y}$$
(3.22)

with

$$e_{\rm Q} \equiv \langle \operatorname{div}(\rho D \nabla Q) + \rho D \nabla \xi \cdot \nabla \frac{\partial Q}{\partial \eta} \mid \xi(\mathbf{x}, t) = \eta \rangle$$
(3.23)

$$e_{\mathbf{Y}} \equiv - \langle \rho \frac{\partial Y''}{\partial t} + \rho \mathbf{v} \cdot \nabla Y'' - \operatorname{div} \left(D \rho \nabla Y'' \right) \mid \xi(\mathbf{x}, t) = \eta \rangle$$
(3.24)

Here $N \equiv D(\nabla \xi)^2$, $\rho_{\eta} \equiv \langle \rho | \eta \rangle$. The conditional fluctuations of density are neglected (see Section 3.6 for details). Equation (3.22) represents the unclosed form of the equation for Q.

3.2.2. Primary closure hypothesis

The analysis of Section 2.3.3 indicates that the term e_Q is small when Re number is large. This term can be neglected. Now we need to estimate the last term on the right-hand side of Eq.(3.22), e_Y .

The conditional fluctuations Y'' have been introduced such that $\langle Y'' | \eta \rangle = 0$. Hence $\langle Y'' \rangle = 0$. It follows from Eq.(2.36) that $\langle Y'' | \eta \rangle = 0$ does not necessarily mean that $\langle \nabla Y'' | \eta \rangle = 0$ and $\langle \partial Y'' / \partial t | \eta \rangle = 0$. We can, however, conclude that the unconditional averages of such derivatives are zero: $\langle \nabla Y'' \rangle = \nabla \langle Y'' \rangle = 0$ and $\langle \partial Y'' / \partial t \rangle = \partial \langle Y'' \rangle / \partial t = 0$. This means that

$$\int \langle \nabla Y'' | \eta \rangle P(\eta) d\eta = 0 \tag{3.25}$$

$$\int \langle \partial Y'' / \partial t | \eta \rangle P(\eta) d\eta = 0$$
(3.26)

where the integrals are taken over all η . Terms such as $\langle \nabla Y'' | \eta \rangle$ are not zero but, according to (3.25) and (3.26) their integral contribution is zero. Let us determine the unconditional average value of $e_{\rm Y}$

$$-\int e_{\mathbf{Y}} P(\eta) d\eta = \langle \frac{\partial \rho Y''}{\partial t} + \operatorname{div} \left(\rho \mathbf{v} Y'' \right) - \operatorname{div} \left(\rho D \nabla Y'' \right) \rangle$$
$$= \frac{\partial \langle \rho Y'' \rangle}{\partial t} + \operatorname{div} \left(\langle \rho \mathbf{v} Y'' \rangle \right) - \operatorname{div} \left(\langle \rho D \nabla Y'' \rangle \right) = \operatorname{div} \left(\langle \rho \mathbf{v}'' Y'' \rangle \right)$$
$$= \operatorname{div} \left(\int \rho_{\eta} \langle \mathbf{v}'' Y'' | \eta \rangle P(\eta) d\eta \right) = \int \operatorname{div} \left(\rho_{\eta} \langle \mathbf{v}'' Y'' | \eta \rangle P(\eta) \right) d\eta$$
(3.27)

Some details in this derivation of Eq.(3.27) need comment. The continuity equation multiplied by Y'', that is

$$\left(\frac{\partial \rho}{\partial t} + \operatorname{div}(\rho \mathbf{v})\right) Y'' = 0$$

has been conditionally averaged and added to Eq.(3.24). The conditional fluctuations of density ρ and diffusivity D are neglected so that $\langle \rho Y'' | \eta \rangle = \rho_{\eta} \langle Y'' | \eta \rangle = 0$; hence $\langle \rho Y'' \rangle = 0$. The diffusion term is transformed as

 $\langle \rho D \nabla Y'' \rangle = \nabla \langle \rho D Y'' \rangle - \langle Y'' (\nabla \rho D) \rangle = 0$. The velocity is decomposed as $\mathbf{v} = \langle \mathbf{v} | \eta \rangle + \mathbf{v''}$. Note that conditional fluctuations are different from $\langle \mathbf{v}'' \mathbf{Y}'' \rangle \neq \langle \mathbf{v}' \mathbf{Y}' \rangle$ unconditional fluctuations that where so **v**=<**v**>+**v**' and $Y = \langle Y \rangle + Y'$.

The basic closure hypothesis employed in the decomposition approach is that

$$e_{\mathbf{Y}}P(\mathbf{\eta}) = -\operatorname{div}\left(\rho_{\mathbf{\eta}} < \mathbf{v}'' \mathbf{Y}'' \,|\, \mathbf{\eta} > P(\mathbf{\eta})\right) \tag{3.28}$$

equality of the integrals in Eq.(3.27) is being assumed to be valid The for the conditional expectations inside the integrals. This is, of course, Here we emphasize that this assumption will not not generally true. bring any error in the unconditional averages since integration over the of η , as in Eqs(3.25)-(3.27), eliminates With range any error. the modelling of Eq.(3.28) the resulting equation takes the form

$$\frac{\partial Q}{\partial t} + \langle \mathbf{v} | \boldsymbol{\eta} \rangle \nabla Q + \frac{\operatorname{div} \left(\rho_{\boldsymbol{\eta}} \langle \mathbf{v}'' Y'' | \boldsymbol{\eta} \rangle P(\boldsymbol{\eta}) \right)}{P(\boldsymbol{\eta}) \rho_{\boldsymbol{\eta}}} - \langle N | \boldsymbol{\eta} \rangle \frac{\partial^2 Q}{\partial \boldsymbol{\eta}^2} = \langle W | \boldsymbol{\eta} \rangle (3.29)$$

This is the same as Eq.(3.16).

second and third terms on the left-hand side of Eq.(3.29)The are The third convective terms. term corresponds transport to by the conditional fluctuations. The last term on the left-hand side of Eq.(3.29) corresponds to diffusion in conserved scalar phase space. This term is dissipation Note determined by processes. that this term cannot be neglected even if Re is large. The term on the right-hand side of (3.29) is the conditional expectation of the chemical source term.

3.3. Comparing the Primary Closure Hypotheses

In this section we first examine the Klimenko⁵⁴ closure hypothesis of Eq.(3.13) using Bilger's^{10,12} decomposition Y=Q+Y''. Taking into account Eqs.(2.17) and (3.19) we may rewrite Eq.(3.5) in the form

$$J_{\rm Y} \equiv 2 < N |\eta > P(\eta) \rho_{\eta} \frac{\partial Q}{\partial \eta} - \frac{\partial < N |\eta > P(\eta) \rho_{\eta} Q}{\partial \eta} +$$

$$2 < D(\nabla(Y'') \cdot \nabla\xi) |\eta > P(\eta)\rho_{\eta} - \frac{\partial < NY'' |\eta > P(\eta)\rho_{\eta}}{\partial\eta}$$
(3.30)

The term $D\nabla Q \cdot \nabla \xi$ has been neglected in Eq.(3.30) using the high Re assumption of Section 2.3.3. Using Eq.(3.30) we can rewrite the closure assumption of Eq.(3.13) in the form

$$2 < D(\nabla(Y'') \cdot \nabla\xi) |\eta > P(\eta)\rho_{\eta} - \frac{\partial < NY'' |\eta > P(\eta)\rho_{\eta}}{\partial\eta} = 0$$
(3.31)

Since only $\partial J_{\rm V}/\partial \eta$ appears in the CMC equation, the right-hand side of Eq.(3.31) could in fact be a non-zero constant. From this it appears that lack of correlation between Y and N would be a sufficient condition for the closure to be valid. Such a condition may, however, be too strict. Indeed, consideration in of the balance equation for the second 8.2, 8.3) conditional moments (Sections this term is seen being as an important component of the major source of conditional fluctuations.

Next, let us compare in general the results obtained in Sec.3.1 and Sec.3.2. First we note that Eqs.(3.16) and (3.29) are exactly the same. We forms of the equation also expect that the unclosed can for Q in Eqs.(3.4)-(3.5) Eqs.(3.22)-(3.24) must be equivalent both and in since derived from the scalar forms are transport equations (3.1)and (3.2).obvious but it can be This equivalence is not demonstrated by using Eqs.(2.36) and (3.30). The primary closure hypotheses (3.10) and (3.28)quite different, but they arise from equations seem to be which are and result in identical CMC equations (3.16) and equivalent (3.29),and hence they must be the same. They must also be the same as that in Eq. (3.31).Clarification of these issues and of the question of what sufficient for conditions are necessary and the closure to be valid are matters of ongoing research. Some further discussion of the latter issue may be found in Section 3.7.

3.4. Enthalpy Equation in CMC

The enthalpy h of a mixture of different species is a function of concentrations of the species Y_i and temperature T which is given by the equation

$$h = h(Y_1, ..., Y_n, T) = \sum_{i} Y_i \left((h_0)_i + \int_{T_0}^{T} (C_p)_i dT \right)$$
(3.32)

where $(h_0)_i$ is the enthalpy of formation and $(C_p)_i$ is the specific heat at constant pressure.

For the case of identical diffusion coefficients and low Mach number, the governing equation for the enthalpy takes the form

$$\rho \frac{\partial h}{\partial t} + \rho \left(\mathbf{v} \cdot \nabla h \right) - \operatorname{div} \left(D \rho \nabla h \right) = \frac{\partial p}{\partial t} - W_{\mathrm{R}} \rho$$
(3.33)

where $W_{\rm R}$ is the heat loss rate per unit mass due to radiation. The pressure derivative $\partial p/\partial t$ is often neglected in combustion modelling ⁴⁷. In unsteady systems, such as combustion in a diesel engine, this term is, however, usually important. The form of Eq.(3.33) is quite similar to Eq.(3.2). Hence the CMC equation for $Q_{\rm h} \equiv \langle h | \eta \rangle$ is given by

$$\frac{\partial Q_{\rm h}}{\partial t} + \langle \mathbf{v} | \boldsymbol{\eta} \rangle \cdot \nabla Q_{\rm h} + \frac{\operatorname{div} \left(\rho_{\boldsymbol{\eta}} \langle \mathbf{v}'' h'' | \boldsymbol{\eta} \rangle P(\boldsymbol{\eta}) \right)}{P(\boldsymbol{\eta}) \rho_{\boldsymbol{\eta}}} =$$

$$< N |\eta > \frac{\partial^2 Q_h}{\partial \eta^2} + < \frac{1}{\rho} \frac{\partial p}{\partial t} |\eta > - < W_R |\eta >$$
 (3.34)

This equation is similar to Eqs.(3.16) and (3.29) and does not need special consideration of its properties.

3.5. Note on Averaging of the Source Terms

Solving the conventionally averaged scalar transport equations requires a closure for the average values of the source terms $\langle W_i \rangle$. In this section we follow the analysis of Bilger^{10,12,13}. The chemical source terms for one-step irreversible reactions

$$A + B \rightarrow$$
 Products

$$W = \rho k Y_{\rm A} Y_{\rm B}; \tag{3.35}$$

$$k = A_0 T^\beta \exp\left(-T_a/T\right)$$
(3.36)

where k is the rate constant, A_0 is the frequency factor, β is the temperature exponent, and T_a is the activation temperature. It is quite obvious that W is not a linear function of its arguments. In the general the reaction rate is a non-linear function of case the species mass and the temperature T. Temperature can be determined fractions, Y_1, \dots, Y_n , from Eq.(3.32) as a function of h and Y_1, \dots, Y_n . Thus finally we can write $W_i = W_i(\mathbf{Y}, h)$ where $\mathbf{Y} \equiv (Y_1, \dots, Y_n)$ and the reaction rate is a non-linear function of its arguments. When W_i are not linear functions of Y_i , the unconditional average values of W_i are not functions of the unconditional averages of the mass fractions and enthalpy $\langle W_i(\mathbf{Y},h) \rangle \neq W_i(\langle \mathbf{Y} \rangle,\langle h \rangle)$. The size of the error depends on the size of the fluctuations in the scalars Y_i and *h*. In real combustion processes the source terms are usually non-linear functions of strongly their arguments and the scalar fluctuations are large so that use of the approximation $\langle W_i(\mathbf{Y},\mathbf{h}) \rangle$ = $W_i(\langle \mathbf{Y} \rangle, \langle \mathbf{h} \rangle)$ would cause very large errors. This problem is well-known in combustion science as the problem of averaging the reaction rates. This is a simple mathematical explanation of what is in reality a complex physical problem. For multistep kinetics the reaction rate for a given species often has positive and negative terms which are of similar magnitude. Borghi¹⁸ has shown that it is not possible to obtain closure by modelling the second or even higher order correlations.

In conditional modeling we need to determine the conditional expectation of the source terms $\langle W_i | \eta \rangle$. This in itself does not seem to make problem of averaging easier since $\langle W_i \rangle$ is determined by $\langle W_i | \eta \rangle$ and the pdf, $P(\eta)$. The CMC model, however, provides more information for averaging the source terms because the conditional expectations $Q_i \equiv \langle Y_i | \eta \rangle$ much more detailed characteristics of the reactive scalar fields than are unconditional expectations $\langle Y_i \rangle$. Also, in many the cases conditional order than unconditional fluctuations fluctuations of smaller are $Y''_{\cdot} \ll Y'_{\cdot}$ The $Y''_{i} \equiv Y_{i} - Q_{i}, \qquad Y'_{i} \equiv Y_{i} - \langle Y_{i} \rangle.$ conserved scalar ξ representing where the

mixture fraction gives a measure of the stoichiometry of the mixture and so usually characterizes well the chemical structure of the mixture. The inequality $Y''_{i} \ll Y'_{i}$ will be considered in Section 5 where we consider CMC in turbulent shear flows and in Section 8 where the equation for the conditional variance $G_i \equiv \langle (Y''_i)^2 | \eta \rangle$ is derived. We emphasize here that the conditional depend only on intensities of fluctuations not the configuration of the turbulent flow but also on the nature of the chemical reactions. We would not expect, for example, the conditional fluctuations to be small in zones where combustion is close to extinction. In many cases the conditional fluctuations Y''_{i} can be neglected so that

$$\langle W_{i}(\mathbf{Y},h) | \eta \rangle \approx \langle W_{i}(\mathbf{Q},Q_{h}) | \eta \rangle = W_{i}(\mathbf{Q},Q_{h})$$

$$(3.37)$$

This can be seen for the simple case of Eq.(3.35) by where $\mathbf{Q} \equiv (Q_1, \dots, Q_n)$. series making a Taylor expansion for the reaction rate around the conditional means of the reactive scalars and conditionally averaging the result. To second order accuracy we obtain

$$<\!\!W|\eta> \approx <\!\!\rho|\eta>k(Q_{\rm T}) Q_{\rm A}Q_{\rm B} \left[1 + \frac{<\!Y_{\rm A}'' Y_{\rm B}''|\eta>}{Q_{\rm A}Q_{\rm B}} + \left(\beta + \frac{T_{\rm a}}{Q_{\rm T}}\right) \left(\frac{<\!Y_{\rm A}'' T''|\eta>}{Q_{\rm A}Q_{\rm T}} + \frac{<\!Y_{\rm B}'' T''|\eta>}{Q_{\rm B}' Q_{\rm T}}\right) + \frac{1/2}\left[\beta(\beta-1) + \frac{2(\beta-1)T_{\rm a}}{Q_{\rm T}} + \frac{T_{\rm a}^2}{Q_{\rm T}^2}\right] \leq \!\!\frac{<\!(T'')^2|\eta>}{Q_{\rm T}^2} \right]$$
(3.38)

where

$$k(Q_{\mathrm{T}}) = A_0 Q_{\mathrm{T}}^{\beta} \exp\left(-T_{\mathrm{a}}/Q_{\mathrm{T}}\right), \quad Q_{\mathrm{T}} \equiv \langle T | \eta \rangle \text{ and } T'' \equiv T - Q_{\mathrm{T}}.$$

with density fluctuations neglected Correlations are in this derivation. not be involved if density weighted conditional averaging is They will used as is shown in the next Subsection. It can be seen from Eq.(3.38)that the errors in making a first order closure will be small if the conditional variances of species mass fractions and temperature are small compared with the square of their conditional means. sufficiently In saving note that the conditional correlation coefficients this we are bounded between -1 and +1. Eq.(3.38) can be used to evaluate the adequacy of first moment closures. It is to be noted that where β and T_a/Q_T are much larger than unity the requirement for smallness of the conditional temperature variance becomes particularly stringent. In these cases it may be necessary to make a second order closure for the conditional reaction rate. This can be done using Eq.(3.38)and solutions of appropriately for the conditional variances modelled equations and covariances as are developed in Section 8.

Let us consider closure for the source terms in Eq.(3.34). Closure the conditional average of the radiation source term that appears last for right-hand side of Eq.(3.34) follows on the the same considerations as systems given above for the reaction rate. For those which are $W_{\rm R}$ is a function of the composition and enthalpy and optically thin, estimates the conditional average first-order closure $\langle W_{\rm R}/\eta \rangle$ by using the conditional averages in this function similar to Eq.(3.37). Estimates of the error involved in such first-order closure can be made using A Taylor series expansion similar to that Eq.(3.38). The second term on the of Eq.(3.34) is related to rapid right-hand side global expansion or compression in the combustor such as that occurring due to piston motion diesel engine. Its conditional be in а average can reasonably approximated by using the conventional average for the rate of pressure rise so that

$$<\frac{1}{\rho} \frac{\partial p}{\partial t} |\eta> \approx <\rho |\eta>^{-1} <\frac{\partial p}{\partial t}>$$
(3.39)

Indeed for the diesel engine example, it will be probably be sufficient to take the rate of rise of the average pressure across the whole clearance volume.

3.6. Note on Conditional Favre Averaging

In the derivations of the CMC equation we repeatedly neglected the conditional fluctuations of density as if ρ were a deterministic function of η . For example $\langle Y\rho | \eta \rangle = Q\rho_{\eta}$. This is a reasonable assumption since the density fluctuations with respect to its conditional mean are often not

significant in non-premixed systems which are not near extinction. verv conditional fluctuations (This corresponds to relatively small of chemical Bilger¹⁰ and species and temperature.). We can, however, follow discard this by introducing conditional Favre averaging: assumption we put $\langle \cdot |\eta \rangle \equiv \langle \cdot |\xi = \eta \rangle$ $\langle \cdot | \eta \rangle \equiv \langle \cdot \rho | \xi = \eta \rangle / \rho_{\eta}$ instead of where $\rho_{\eta} \equiv \langle \rho | \xi = \eta \rangle.$ definitions This corresponds to of the conditional new mean $Q \equiv \langle Y \rho | \xi = \eta \rangle / \rho_{\eta}$ and the conditional fluctuations $Y'' \equiv Y - Q.$ Note that $\langle Y'' | \eta \rangle \equiv \langle Y'' \rho | \xi = \eta \rangle / \rho_n = 0$ but the unweighted conditional average $\langle Y''|\xi = \eta \rangle$ is not necessarily zero. The equations derived in Sec.3 are still valid with the new definitions but their derivations do not require neglecting the conditional fluctuations of the density.

3.7. The Physical Basis of CMC

We consider here some physical ideas and hypotheses which provide a basis for the primary closure involved in CMC as has been developed in This discussion mainly follows Klimenko^{54,56} Sections 3.1 - 3.3. and it is particularly related to the closure hypothesis in Eq. (3.10). Note that with ideas sufficient the constraints associated these are but not necessary for the derivation of the CMC. The CMC equations can be derived ways using different versions of the primary in different closure The considered hypothesis. concepts in this section are, probably, most complicated and the section can be omitted at first reading.

3.7.1 Diffusion approximations and the analogy with a Markov process

The focus here is the applicability of the diffusion analogy approximation for $J_{\rm Y}$. We compare the diffusion approximation in Eq.(3.10) and the diffusion approximation of the turbulent scalar flux

$$\langle \mathbf{v}' \mathbf{Y}' \rangle = -D_t \nabla \langle \mathbf{Y} \rangle \tag{3.40}$$

where $\mathbf{v}' \equiv \mathbf{v} \cdot \langle \mathbf{v} \rangle$, $Y' \equiv Y \cdot \langle Y \rangle$. In the case of a homogeneous velocity field, constant density and W=0, considered here as a most simple example of using Eq.(3.40), the average scalar transport equation takes the form

$$\frac{\partial \langle Y \rangle}{\partial t} = \operatorname{div}\left(D_{t}\nabla \langle Y \rangle\right)$$
(3.41)

Following Corrsin³¹, let us assume that the scalar Y is the number density of an enormously large set of infinitesimal particles, which do not affect the flow. A sample particle is chosen with uniform probability for all particles. Its coordinates are marked by subscript index "p". The physical coordinate along a certain direction is denoted in this section as X and the corresponding component of the velocity is denoted by V. We consider the case when all particles are initially located at $X=X_0$ The Taylor's¹³⁴ turbulent diffusion of main result theory is the relation $\langle (\Delta X_{\rm p})^2 \rangle$ between mean square deviation of the sample particle position correlation $K_{v} = \langle V_{p}(t_{1}) \cdot V_{p}(t_{2}) \rangle$ of (where $\Delta X_{\rm p} \equiv X_{\rm p} - X_0$ and the the corresponding component of the particle velocity. It is that

$$<(\Delta X_{\rm p})^2 > = 2 \int_{t_0}^{t} \int_{t_0}^{t_1} K_{\rm v}(t_1, t_2) dt_2 dt_1$$
 (3.42)

If the particle motion can be assumed to be steady-state during the period of time under consideration, the velocity correlation depends only on the time difference $K_v = K_v(\Delta t)$, $\Delta t = t_2 - t_1$. Differentiating Eq.(3.42) twice with respect to *t*, we obtain

$$\frac{d^2}{dt^2} < (\Delta X_p)^2 > = 2K_v(0) + 2\int_{t_0}^t \frac{d}{dt} K_v(t_2 - t) dt_2 = 2K_v(t - t_0)$$
(3.43)

In terms of the mean concentration, $\langle Y \rangle$, the mean square deviation (or dispersion) is determined by

$$\langle (\Delta X_{\rm p})^2 \rangle = \frac{\int_{\infty}^{\infty} (X - X_0)^2 \langle Y \rangle d\mathbf{x}^3}{\int_{\infty}^{\infty} \langle Y \rangle d\mathbf{x}^3}$$
(3.44)

since all particles will, on average, behave similarly. Assuming constant turbulent diffusion coefficient D_t , we multiply Eq.(3.41) by $(X-X_0)^2$ and integrate it over all X. This yields linear dependence of $\langle (\Delta X_p)^2 \rangle$ in time and, according to Eq.(3.43), the particle velocities should be

uncorrelated $K_{v}(\Delta t)=0$. This is true only for values Δt greater than the turbulence integral time scale τ_L . It is quite obvious that the diffusion approximation (3.40) with a constant D_t fails to describe the processes with characteristic time scale less then τ_{I} . This means that the turbulent differently for diffusion coefficient must be chosen different initial conditions imposed on Y even if the velocity fields are the same. This deprives Eq.(3.40) of any useful information since Eq.(3.40) can be considered in the one-dimensional case as being merely the definition of diffusion coefficient $D_t \equiv -\langle V'Y' \rangle / (\partial \langle Y \rangle / \partial X), \quad V' \equiv V - \langle V \rangle$ the turbulent (We leave aside the possibility of there being negative values for D_{t} .)

Our purpose now is to analyze the diffusion approximation of $J_{\rm Y}$ in Let us introduce a new variable ξ_p that is the particle Eq.(3.10). coordinate in conserved scalar space. The value of ξ_p is specified as the instantaneous value ξ in the close vicinity of our chosen small particle. The value ξ_p , itself, and its derivative $\xi'_p \equiv d\xi_p/dt$ are random functions of time. We assume that at initial time t_0 the particle has a coordinate ξ_0 in the conserved scalar space. The deviation from the initial position is designated as $\Delta \xi_p = \xi_p(t) - \xi_0$. Within time scale of the inertial range of assume that the particle motions are statistically the turbulence we $<\xi'_{p}(t_{1})\cdot\xi'_{p}(t_{2})>=K_{\xi}(t_{2}-t_{1}).$ We uniform shall determine the in time: relation between mean square deviation in conserved scalar space $<(\Delta \xi_p)^2 >$ and time $\Delta t = t - t_0$. According to the theory of Kolmogorov⁶³ and Oboukhov¹⁰², belonging the inertial interval time intervals to this relation is in determined by the mean dissipation of turbulent energy $\langle \epsilon_d \rangle$ and the mean scalar dissipation $\langle N \rangle$ where $N \equiv D_{\xi} (\nabla \xi)^2$. It requires the following form of the relation

$$<(\Delta \xi_{\rm p})^2 > = \operatorname{const} \Delta t;$$
 (3.45)

Using (3.43) we easily obtain

$$K_{\xi} = \frac{1}{2} \frac{d^2}{dt^2} < (\Delta \xi_{\rm p})^2 > = 0$$
(3.46)

This means that increments of the particle position inside the inertial time interval are uncorrelated. Note that if ξ_0 is fixed but not randomly chosen with probability $P(\eta)$, conditional averaging $\langle \cdot | \xi = \xi_0 \rangle$ should be

substituted here for unconditional averaging <.>.

Let us consider the qualitative behaviour of correlation $K_{\mathcal{E}}(\Delta t)$ in the full range of the variable Δt . We assume that the Peclet number, Pe. calculated from the conserved scalar molecular diffusivity, D_{ξ} , is of the same order as the Re number calculated from the kinematic viscosity. The integral of function $K_{\xi}(\Delta t)$ determines the diffusion coefficient in conserved scalar space. The correlation K_{ξ} contains a component related to the molecular Brownian motion of the particle. The characteristic time of this correlation is the interval between molecular interactions. the As such time is very small this component is shown in Fig. 10 as a delta-function. Therefore this component is not discussed further in this section so that the particle we consider becomes a fluid particle. Its motion in conserved scalar space is given by the equation

$$\xi'_{\rm p} = {\rm div}(D_{\xi}\rho\nabla\xi)/\rho \approx D_{\xi}\nabla^2\xi; \quad \xi'_{\rm p} \equiv d\xi_{\rm p}/dt$$

This equation represents the Lagrangian time derivative of ξ . The value of $D_{\xi}\nabla^2\xi$ which involves derivatives of the second order is determined by the turbulent fluctuations of smallest scales. Thus the order of ξ'_p can be expressed using the corresponding Kolmogorov scales. These scales are: the length scale $x_{\rm K}$, the time scale $\tau_{\rm K}$ and the conserved scalar scale $\xi_{\rm K}$. Taking into account that $\langle N \rangle \sim \xi_{\rm K}^2 / \tau_{\rm K}$, we obtain

$$K_{\xi}(0) \sim \xi_{\rm K}^2/\tau_{\rm K}^2 \sim \langle N \rangle / \tau_{\rm K} ; \quad \tau_{\rm K} \sim \tau_{\rm L} {\rm Re}^{-1/2}$$
 (3.47)

where Re is the turbulence Reynolds number. But in times longer than τ_{K} the correlation $K_{\xi}(\Delta t)$ tends to zero according to Eq.(3.46) (Fig. 10 solid emphasize that correlation $K_{\xi}(\Delta t)$ It is necessary to line). can have component $K'_{\sharp} \sim <N > /\tau_{L}$ with the typical correlation time $\Delta t \sim \tau_{\rm L}$ (Fig. 10 dotted line). The component K'_{\sharp} does not interfere with Eqs.(3.45) and (3.46) in the main order. The particle motion in conserved scalar space is determined by the universal small-scale turbulent processes. According Kolmogorov⁶³ theory (see also Batchelor⁵), the such processes to are stochastically of non-universal large-scale turbulent independent motions. In our case it means, that process $\xi_{\rm p}(t)$ at time $\tau_{\rm K} \ll \Delta t \ll \tau_{\rm L}$ can be considered as a Markov process with independent increments. This statement with some continuity assumptions leads us to the conclusion of the local Brownian and conserved analogy between scalar space motion. This

Oboukhov's¹⁰³ hypothesis hypothesis shows similarities with the of a properties Markov-process of particle motions in the velocity space. It too optimistic assume that the motion of a particle would be to in conserved scalar space is mathematically identical to the Brownian motion but we can reasonably expect that the diffusion approximation in Eq.(3.10) which corresponds to a Markov diffusion process is much better than the diffusion approximation in Eq.(3.40). The coefficients A and В in be dependent Eq.(3.10) are expected to on the velocity and conserved scalar fields but independent of the reactive scalar field (the scalar Y affect the coefficients A and В only by affecting density). This can preserves the linear properties of the turbulent transport in Eqs.(3.2) and (3.4) for the Markov-process assumptions.

Klimenko⁵⁵ demonstrated that the assumption of Markov-process scalar properties for particle diffusion in conserved corresponds space to Kolmogorov⁶³ original version of the theory. Kolmogorov's⁶⁴ the refinements to the theory may result in a significant deviation of the local transitional pdfs from a Gaussian shape. This seems to be insignificant for the first moment $(Q \equiv \langle Y | \eta \rangle)$ closure, but corrections for second moment closure may be needed.

Chemical reactions cause the appearance and disappearance of the particles. The analysis of this section be applied reactive can to provided the characteristic chemical time, which particles, that can be characteristic time "life", also called the of particle is much greater than Kolmogorov time scale τ_{κ} . We emphasize that we do not draw here a conclusion about the the limits of the CMC applicability and these final matters will be discussed further in the paper.

3.7.2. Hypothesis of local similarity

hypothesis of local similarity of scalar fields is not a needed The of the CMC assumption in the derivation equations but it yields some Here we consider the case when either additional insights. chemical reactions do not occur or they are too weak to affect the small-scale structure of the scalar fields.

If $Y=a+b\xi$ and W=0 in the large scales, this linear dependence would preserved small-scale fluctuations. small-scale be in the Any $a+b\xi$ would quickly disappear. perturbations of So. if it appears that

there is dependence between Y and ξ somewhere in a large-scale region of the turbulent flow, this dependence will be preserved in this region at The kinetic energy and scalar fluctuations transferred smaller scales. are form large scales to small scales. Once dependence between Y and ξ appears it will at anv stage of this transport, be preserved at further stages. The local similarity of scalar fields is understood as the general tendency of the scalar gradients ∇Y and $\nabla \xi$ to become more and more aligned if the large-scale gradients of the scalars are not even aligned. Hence inside a relatively small region one scalar would be a function of another $Y=f_{\rm Y}(\xi)$ and this random function is and determined larger scalar by fluctuations. The implementation of this local similarity of scalar fields and of the independence of large-scale and small-scale fluctuations yields

$$\langle NY|\eta \rangle = \langle N|\eta \rangle \langle Y|\eta \rangle \tag{3.48}$$

$$\langle D(\nabla Y \cdot \nabla \xi) | \eta \rangle = \langle N | \eta \rangle \frac{\partial \langle Y | \eta \rangle}{\partial \eta}$$
(3.49)

We emphasize here that Eqs.(3.48) and (3.49)sufficient but are not (3.48)necessary for derivation of Eq.(3.31). The assumptions and (3.49)given here since they represent a self-consistent logic. The CMC are equation actually requires Eq.(3.31), or more precisely this equation but with a constant on the RHS (but not Eqs.(3.48) and (3.49)) to be valid.

3.8 Summary of the Major Results

In this Section we have presented the derivation of the main CMC given by Eqs (3.14), (3.16) equation with the result and (3.29).The results in Eqs (3.29) and (3.16) are identical and are linked to Eq.(3.14)by the high Reynolds number form of the transport equation for the joint pdf of the mixture fraction, Eq.(3.15). These two forms of the first-order CMC equation are widely used in Sections 4 to 7 that deal with its application. They gover n the spatial and temporal evolution of the conditional expectation $\langle Y_i | \xi = \eta \rangle$ of reactive scalars Y_i conditioned on а value η of the conserved scalar ξ . The CMC equation has fixed the additional independent variable η and several terms this in equation need further modelling. Closure for the chemical source term is considered in section 3.5 and general issues of closure of the other unclosed terms are covered in Section 4. Simplified versions of CMC and further closures will

be considered in Sections 5, 6 and 7.

The enthalpy equation which is needed to find the temperature is considered separately in Section 3.4. In this section the CMC equation is formulated for the enthalpy and the major terms of this equation are similar to those in Eqs. (3.16) and (3.29).

Conditional expectations of the chemical source terms are present in the CMC equations. The most simple approximation for these conditional obtained Eq.(3.37) expectations is in by neglecting the conditional Y″. In other fluctuations words, a first-order conditional moment closure is made for the chemical source terms in the first-order CMC equation. Higher-order closure for these terms is considered in more detail in Section 8. It should be noted that this approximation does take into account the most significant components of the conventional fluctuations. \mathbf{Y}' , from the unconditional average and thus has higher precision than the conventional pseudo-laminar approximations which neglect the effects of Y' entirely.

Conditional fluctuations of density have been generally neglected in the derivations of the CMC equations. This assumption can be avoided by introducing conditional Favre averaging as is discussed in Section 3.6.

The CMC equations are derived here by two alternative techniques: the pdf method and the decomposition method. The decomposition method may be preferred by readers who are familiar with the fast chemistry and the flamelet models. The pdf method, as it follows from its title, provides with the traditional pdf techniques. It is important to emphasise the link basic CMC equations (3.14), (3.16), and (3.29)involve that the certain basic assumptions primary closure hypotheses. Although both derivations vield exactly the same result, the assumptions involved in both identical, least from the philosophical point derivations are not at of view. The pdf derivation is based on a new physical concept - diffusion in conserved scalar space, which is presented in Section 3.7. This concept is directly supported by the Kolmogorov theory of turbulence and can not be found in CMC predecessors - the fast chemistry and the flamelet models. This concept explains why CMC is valid for slow and moderate chemistry and reaction zones which are broad compared with the Kolmogorov length scale. At the same time CMC is still valid for fast chemistry and, with certain reservations, for thin reaction zones. It should be emphasised that the decomposition method does not impose the restriction which is required by the concept of diffusion in conserved scalar space where it was assumed

that the chemical transformation time exceeds the Kolmogorov time scale, τ_{K} . That is why our intention is to inform the reader about different approaches to CMC. If the reader is concerned with practical applications rather then fundamental properties of turbulence, he or she may wish to skip Section 3.7 and focus on comparison of CMC and the fast chemistry and the flamelet models forgetting for a while about their fundamental differences. Such comparative analysis of the models is given in the next section.

4. MAIN FEATURES OF THE CMC EQUATION

In Section 3, different methods of deriving the CMC equation and formulations for the primary closure hypothesis have been presented. They result in the same first-order CMC equation for the conditional averages $Q = \langle Y | \eta \rangle$, where the subscript *i* used in eq. (1.1) has been dropped without loss of generality (see the beginning of Section 3.) From Eqs.(3.14), (3.16) and (3.29) we see that the CMC equation has two equivalent forms

$$\frac{\partial QP(\eta)\rho_{\eta}}{\partial t} + \operatorname{div}\left(\langle \mathbf{v}Y|\eta \rangle P(\eta)\rho_{\eta}\right) = \langle W|\eta \rangle P(\eta)\rho_{\eta} + \frac{\partial}{\partial \eta}\left(\langle N|\eta \rangle P(\eta)\rho_{\eta} \frac{\partial Q}{\partial \eta} - \frac{\partial \langle N|\eta \rangle P(\eta)\rho_{\eta}}{\partial \eta}Q\right)$$
(4.1)

$$\frac{\partial Q}{\partial t} + \langle \mathbf{v} | \eta \rangle \cdot \nabla Q + \frac{\operatorname{div} \left(\langle \mathbf{v}'' Y'' | \eta \rangle P(\eta) \rho_{\eta} \right)}{P(\eta) \rho_{\eta}} - \langle N | \eta \rangle \frac{\partial^2 Q}{\partial \eta^2} = \langle W | \eta \rangle (4.2)$$

The second of these is the more general form of Eq. (1.1) in the present notation. The transformation of Eq.(4.1) into Eq.(4.2) and Eq.(4.2) into Eq.(4.1) can be easily made by using the conserved scalar pdf equation (3.15)

$$\frac{\partial P(\eta)\rho_{\eta}}{\partial t} + \operatorname{div}\left(\langle \mathbf{v} | \eta \rangle P(\eta)\rho_{\eta}\right) = -\frac{\partial^{2} \langle N | \eta \rangle P(\eta)\rho_{\eta}}{\partial \eta^{2}}$$
(4.3)

First-order closure for the conditional average reaction in rate terms of the conditional average species mass fractions and enthalpy has been presented in Section 3.5. Here we are concerned with models for closing the remaining unclosed terms in the CMC equation, investigation of general properties and nature of the boundary conditions its the applicable.

outlined in Section 1.2, solution of the CMC As equation usually in parallel with some method for solution of the velocity proceeds and fraction fields. Information from these solutions needed for mixture is modeling $P(\eta)$, $\langle \mathbf{v} | \eta \rangle$ and other unclosed terms in the CMC equations. In Section 4.1 we outline methods for doing this in the general case.

The CMC equation, (4.1) or (4.2), needs to be solved in a way that is with consistent the pdf transport equation (4.3). In the normal procedure this means that models used for the conditional average dissipation, $\langle N | \eta \rangle$, should show consistency with the given pdf field in As is shown in Section 4.2, this is necessary terms of Eq. (4.3). to conservation integrals. It is found. however, that in preserve errors $\langle N | \eta \rangle$ are only significant where they occur in reaction zones. The conservation integrals also give some constraints the boundary on conditions for Q.

The form of the CMC equation of (4.2) has certain similarities with turbulent combustion models applicable in the fast chemistry limit and to laminar flamelet modeling ^{109,22}. The relationship of CMC (FCL) ^{6,7} methods these other modelling approaches is of some interest. The to to the frozen and fast chemistry limits is explored relationship in Sections 4.3, and to laminar flamelet models in Section 4.4.

Solution of the CMC equation requires specification of the correct initial and boundary conditions for the flow under consideration. In mixing layers with non-reactive fuel and oxidant streams there is little problem with specification of boundary conditions at $\eta = 0$ and 1. There are many subtleties to be considered, however, when the unmixed streams when, as in jets wakes and real combustors, are reacting or the probability of unmixed fluid of one or both streams becomes very small. downstream in a jet diffusion For example. far flame there is little probability of there being fluid with $\xi = 1$ and questions arise as to whether applying a boundary condition at $\eta = l$ is meaningful or valid. These questions are most simply addressed by considering time-dependent homogeneous turbulent flows. In Section 4.5 some results are obtained for the case of homogeneous turbulence which may be of general applicability. In Section 4.6 we summarize the nature of CMC modeling for first order closure.

Equations (4.1)-(4.3)involve five independent variables t, x_1 , x_2 , x_3 , and η . Numerical solution of the equations is greatly eased when the dimensionality of the problem can be significantly reduced. In Sections 5 and 6 we consider the cases when the general CMC equations can be transformed into which simplified and forms can be easily used in practical applications. In Section 7, the even simpler case of homogeneous turbulence is considered.

4.1. Possible Closures of the Remaining Unclosed Terms in the General CMC Equations

Conserved scalar pdf modelling is relatively well-explored area in combustion theory¹¹³. We assume that the function $P(\eta)$ is known throughout and with time. In current practice, a presumed form of the field the pdf^{9,27,47} (such as a clipped Gaussian or beta function) will often be and $P(\eta)$ obtained from solutions for the mean and variance of the used conserved scalar; these being obtained from numerical integration of their modelled transport equations. In fact, any form of detailed information on field the velocity and mixing is acceptable, e.g. experimental measurements or output from large eddy simulations.

Where Favre averaging is used for calculating the velocity and mixing fields, it is usually more convenient to work in terms of the Favre pdf, $\tilde{P}(\eta)$, which is related to the conventional pdf, $P(\eta)$, by

$$\rho_{\eta} P(\eta) = \langle \rho \rangle \widetilde{P}(\eta) \tag{4.4}$$

where $\langle \rho \rangle$ is the unconditionally averaged density. Eq.(4.4) may be used to convert Eqs (4.1)-(4.3) to the Favre pdf form.

The linear approximation for the conditional expectation of velocity is given by

$$\langle \mathbf{v} | \boldsymbol{\eta} \rangle = \langle \mathbf{v} \rangle + \frac{\langle \mathbf{v}' \boldsymbol{\xi}' \rangle}{\langle (\boldsymbol{\xi}')^2 \rangle} \left(\boldsymbol{\eta} - \langle \boldsymbol{\xi} \rangle \right)$$
(4.5)

with similar form applicable to Favre averaging. This simple a approximation is supported by some experimental data summarized in Ref. 72 It is known to be exact if the velocity and conserved scalar are jointly Gaussian, but this condition is not necessary for Eq. (4.5)to be an approximation. The unconditional covariance $\langle v'\xi' \rangle$ adequate usually is available from the modelling used for this quantity in balance the equation for the mean mixture fraction.

The conditional expectation of the scalar dissipation can be found from Eq.(4.3) with the use of the boundary conditions for the product

 $\langle N|\eta \rangle P(\eta)$ established in Section 2.4. With the use of Eq. (4.5) and appropriate integration by parts the double integrals can be reduced to single integrals so that we obtain

$$<\!\!N|\eta\!>\!\!P(\eta) = \frac{\partial I_1(\eta)}{\partial t} + <\!\!\mathbf{v}\!\!>\!\!\nabla I_1(\eta) + + <\!\!\rho\!\!>^{-1} \nabla \cdot \left(<\!\!\rho\!\!>\!<\!\!\mathbf{v}'\xi'\!\!>\!\!I_2(\eta)/\!<\!\!\xi'^2\!\!>\right)$$
(4.6)

where

1

$$I_{n}(\eta) = \int_{\eta} (\eta^{\circ} - \eta)^{n} P(\eta^{\circ}) d\eta^{\circ}$$
(4.7)

These formulae are readily converted to Favre pdfs through the use of Eq. (4.4). For a given presumed form of the pdf, $I_n(\eta) = I_n(\eta; \langle \xi \rangle, \langle \xi'^2 \rangle)$ so that look-up tables can be precomputed. The conditional expectation of the scalar dissipation $\langle N | \eta \rangle$ determined in this way is in agreement with the pdf equation.

experience been gained in As yet no has CMC modelling in multi-dimensional systems. The "brute force" approach outlined above may prove to be too cumbersome or result in negative values of $\langle N|\eta \rangle$ being As will be presented later, errors in obtained through numerical error. the estimation of $\langle N|\eta \rangle$ are only significant for values of η where $\partial^2 O/\partial \eta^2$ is high which is usually where $\eta \approx \eta_s$, the stoichiometric value of the mixture fraction. Usually, values of the unconditional scalar dissipation, <N>, will be available from the flow and mixing calculation. Sufficient accuracy may be achievable through the modelling

$$\langle N | \eta \rangle = \langle N \rangle F_{N} \left(\eta; \langle \xi \rangle, \langle \xi'^{2} \rangle \right)$$

$$(4.8)$$

with the function $F_{\rm N}$ being determined from limited surveys of the flow using Eqs (4.6), (4.7) above, or from experimental data. Some experimental data is available for flows without heat release^{45,49,92} while Starner *et* al^{128} give some results obtained in turbulent jet diffusion flames.

Closure for the term $\langle \mathbf{v}'' \mathbf{Y}'' | \eta \rangle$ can be formulated by analogy with Eq.(3.40)

$$\langle \mathbf{v}'' Y'' | \eta \rangle = -D_t \nabla Q \tag{4.9}$$

approximation (3.40) Since the diffusion is commonly adopted. Eq.(4.9) seems to be a plausible approximation. With the use of Eq. (4.9) the common transport equation for $\langle Y \rangle$ is obtained by integrating Eq. (4.1) over η space; Eq. (3.40) being incorporated in it. It should be noted that a transport equation is not well established for reactive such scalars. For the limiting case of premixed combustion it is known that the gradient (3.40)seriously transport assumption can be in error with the experimental diffusivities often being negative. We are not aware of any data that directly confirms disapproves Eq.(4.9) nonpremixed or for systems. Suffice it to say that this modelling should be treated with many nonpremixed flame situations gradients of O caution. In are quite Eq. (4.9) can be used to give an estimate of the magnitude of this small. it will often be found to be entirely negligible. Exceptions term: will where gradients Q expected stabilization occur strong in are in as regions, such as in a lifted turbulent jet diffusion flame.

The closure of the conditional average chemical reaction rate and radiation loss source terms is discussed in Section 3.5. With the (3.37), (4.5), (4.9)and proper boundary conditions approximations for Q (outlined later in Section 4.6) Eq.(4.2) can be solved. In many cases, as discussed in Sections 5 and 6, we can use the CMC model in a simplified form and avoid the additional assumptions (4.5) and (4.9).

4.2. Conservation Integrals

more The conditional expectation $Q \equiv \langle Y | \eta \rangle$ is a much detailed the characteristic of reactive scalar field than the unconditional expectation *<Y>*. The transport equation for $\langle Y \rangle$ can be obtained by integrating the CMC equation (4.1) over all η (The conserved scalar pdf $P(\eta)$ is bounded by values η_{min} and η_{max}). The CMC equations are fully consistent 54,59,60 with different types of conserved scalar pdf as is shown in this section.

4.2.1. Smooth pdfs with fixed bounds

Here we consider conserved scalar pdfs which do not have any
generalized function components, such as delta functions at η_{min} and η_{max} . The beta function described in Eq.(1.6) and Fig. 3 is an example of this The pdf bounds η_{min} and η_{max} are assumed to be constant. We type of pdf. integrate Eq.(4.1) over the interval $\eta_1 \leq \eta \leq \eta_2$ where $\eta_{\min} < \eta_1 < \eta_2 < \eta_{\max}$. Assuming and taking the limit $\epsilon \rightarrow 0$ $(\eta_1 \rightarrow \eta_{\min})$ and $\eta_2 = \eta_{max} - \varepsilon$ $\eta_1 = \eta_{\min} + \varepsilon$ $\eta_2 \rightarrow \eta_{max}$) we obtain

$$\frac{\partial \langle Y \rho \rangle}{\partial t} + \operatorname{div}\left(\langle \mathbf{v} Y \rho \rangle\right) - \langle W \rho \rangle = \\ = \left[\langle N | \eta \rangle P(\eta) \rho_{\eta} \frac{\partial Q}{\partial \eta} - \frac{\partial \langle N | \eta \rangle P(\eta) \rho_{\eta}}{\partial \eta} Q\right]_{\min}^{\max} = 0$$
(4.10)

The notation used here is that introduced in Eq.(2.56). We have also used the boundary conditions given in Eqs.(2.61) and (2.62). Equation (4.10) is consistent with the average scalar transport equation. Because of the high Reynolds number assumption, transport by molecular diffusion is neglected in the CMC and pdf equations so that the corresponding term does not appear in Eq.(4.10) Note that Eq.(4.10) is valid for any arbitrary values of $[Q]_{\min}$, $[Q]_{\max}$, $[\partial Q/\partial \eta]_{\min}$ and $[\partial Q/\partial \eta]_{\max}$. Thus, in the case of smooth boundary determined pdfs. the conditions by the conservation are not Eq.(4.2) integral. The boundaries represent special point of а since Eq.(2.63) indicates that $\langle N | \eta \rangle \rightarrow \theta$ as $\eta \rightarrow \eta_m$. Hence the boundary values of Qare restricted by Eq.(4.2) itself, so that

$$\left[\frac{\partial Q}{\partial t} + \langle \mathbf{v} | \boldsymbol{\eta} \rangle \nabla Q + \frac{\operatorname{div} \left(\langle \mathbf{v}'' Y'' | \boldsymbol{\eta} \rangle P(\boldsymbol{\eta}) \rho_{\boldsymbol{\eta}} \right)}{P(\boldsymbol{\eta}) \rho_{\boldsymbol{\eta}}} - \langle W | \boldsymbol{\eta} \rangle \right]_{\mathrm{m}} = 0 \quad (4.11)$$

This boundary fluid must shows that if the is reacting there be corresponding temporal and spatial variations in the boundary values for Q.

4.2.2. Intermittent pdfs

The intermittent pdfs specified in Eq.(2.64)) involve delta functions at $\eta = \eta_{min}$ and $\eta = \eta_{max}$ which correspond to the uncontaminated, vortex-free regions of a turbulent flow and η_{min} and η_{max} are constants. The clipped Gaussian pdf described in Eqs (1.3)-(1.5) and Fig. 2 is an example of this

type of pdf. Equation (4.10) is not completely valid for an intermittent pdf. The integration over $\eta_1 \le \eta \le \eta_2$ where $\eta_1 = \eta_{min} + \varepsilon$, $\eta_2 = \eta_{max} - \varepsilon$ and $\varepsilon \rightarrow 0$ does not involve the delta functions at $\eta = \eta_{min}$ and $\eta = \eta_{max}$. Hence the integral of Eq.(4.1) should be written as

$$\frac{\partial \langle Y\rho | t \rangle \gamma_{t}}{\partial t} + \operatorname{div}\left(\langle vY\rho | t \rangle \gamma_{t}\right) - \langle W\rho | t \rangle \gamma_{t} = \\ = \left[\langle N | \eta \rangle P(\eta) \rho_{\eta} \frac{\partial Q}{\partial \eta}\right]_{\min}^{\max} - \left[\frac{\partial \langle N | \eta \rangle P(\eta) \rho_{\eta}}{\partial \eta} Q\right]_{\min}^{\max}$$
(4.12)

where the condition t represents being in the turbulent region and γ_t is the intermittency factor (see Eq.(2.64)). The $\eta_{\min} < \xi < \eta_{\max}$ boundary conditions in Eq.(2.68)-(2.71) indicate that the first term on the RHS of Eq.(4.12) is zero and that the second term can have a non-zero This corresponds to the entraiment of the scalar Y from value. the regions $Y=Y_{\min}$, uncontaminated. vortex-free (where $Y = Y_{max}$) into the contaminated, turbulent region.

It is reasonable to assume that the conditional expectation Q does not have any discontinuity at the vortex-free/turbulent bounds

$$\left[\begin{array}{c} Q \end{array} \right]_{\mathrm{m}} \equiv \lim_{\eta \to \eta_{\mathrm{m}}} \left(\begin{array}{c} Q \end{array} \right) = Y_{\mathrm{m}} \tag{4.13}$$

We remind the reader that index "m" is used for either of the indexes "min" or "max". In order to demonstrate the conservation properties we need to consider a scalar Y° which takes zero values in the vortex-free regions so that entraiment from these regions does not involve entrainment of Y° . We introduce Y° by the equation

$$Y^{\circ} = Y + a(\xi - \eta_{\min}) + b(\xi - \eta_{\max}); \quad a = \frac{-Y_{\max x}}{\eta_{\max} - \eta_{\min}}; \quad b = \frac{Y_{\min n}}{\eta_{\max} - \eta_{\min}}$$
(4.14)

The scalar Y° satisfies Eq.(3.2) hence $Q^{\circ} \equiv \langle Y^{\circ} | \eta \rangle$ satisfies Eq.(4.1). Since Y° is a linear combination of Y and ξ , it is sufficient to demonstrate the conservative properties for Y° . The conservative properties of the conserved scalar are preserved by the pdf equation (4.3). Since $Y^{\circ}_{m}=0$ it is obvious that $[Q^{\circ}]_{m}=0$ and $\langle FY^{\circ} | t \rangle \gamma_{t}=\langle FY \rangle$ for any F. Equation (4.12) takes the form

$$\frac{\partial \langle Y^{\circ} \rho \rangle}{\partial t} + \operatorname{div} \left(\langle \mathbf{v} Y^{\circ} \rho \rangle \right) - \langle W \rho \rangle =$$

$$= \left[\langle N | \eta \rangle P(\eta) \rho_{\eta} \frac{\partial Q^{\circ}}{\partial \eta} \right]_{\min}^{\max} - \left[\frac{\partial \langle N | \eta \rangle P(\eta) \rho_{\eta}}{\partial \eta} Q^{\circ} \right]_{\min}^{\max} = 0 \quad (4.15)$$

This proves that the CMC equation preserves the conservation integrals for the intermittent pdfs provided the boundary conditions in Eq.(4.13) are satisfied. At the same time Eq. (4.15) can be also considered as the proof of the boundary conditions in Eq.(4.13)

4.2.3. Errors arising from the conditional scalar dissipation

It has been emphasised earlier that the modeled values used for the conditional average of the scalar dissipation, $\langle N | \eta \rangle$ in integrating the CMC equation (4.2) must be fully consistent with the pdf field so that the transport equation and the boundary conditions (2.61),(2.62)pdf are satisfied. Otherwise, errors will arise in the predictions for 0 and in particular conservation integrals will not be satisfied. Here we seek to clarify the nature of the errors involved.

Let us assume that the actual conditional average scalar dissipation used in solving Eq. (4.2) is $\langle N|\eta \rangle + \Delta N(\eta)$, where $\langle N|\eta \rangle$ is the correct (4.3)value that satisfies Eq. and the boundary conditions (2.61), (2.62)or (2.68)-(2.71) for the given pdf field and $\Delta N(\eta)$ is the error. It is easy to show that the conservation integrals of Eqs (4.10) and (4.15) will leave an error term $\langle E_N \rangle$ on the RHS where

$$\langle E_{\rm N} \rangle = \int_{\eta_{\rm min}}^{\eta_{\rm max}} \rho_{\eta} \Delta N(\eta) P(\eta) \frac{\partial^2 Q}{\partial \eta^2} d\eta \qquad (4.16)$$

We will call this the "false chemical source term". This is because it leaves unchanged combinations of species that form conserved scalars, Z, provided those conserved scalars have initial and boundary conditions that satisfy the normal requirement that they are linear functions of η . Thus

$$\langle E_{\mathrm{N},Z} \rangle = \int_{\eta_{\mathrm{min}}}^{\eta_{\mathrm{max}}} \rho_{\eta} \Delta N(\eta) P(\eta) \frac{\partial^2 Z}{\partial \eta^2} d\eta \qquad (4.17)$$

where

$$Z \equiv \sum_{i} \mu_{i} Y_{i} = \sum_{i} \mu_{i} Q_{i} \quad \text{and} \quad \sum_{i} \mu_{i} \rho W_{i} = 0$$

(Note that the symbolc use of Z here is different to that in Section 2.) This shows that, as far as the conservation of $\langle Y \rangle$ for any species is concerned, any error introduced by error in $\langle N | \eta \rangle$ will be accompanied by complimentary errors in other species from which a conserved scalar can be defined. There should thus be no evidence of the error arising from this source in element and other conserved scalar balances.

For many nonpremixed systems most species react only in a narrow region in conserved scalar space around stoichiometric, and it is only there are significant second derivatives, $\partial^2 Q / \partial \eta^2$. It there that is seen. stoichiometric, then. that it is only there, that is around that the conditional average of the scalar the dissipation, $\Delta N(\eta)$, errors in need conservation of *<Y>*. For reactant and product to be small to ensure species, there is usually a monotonic change in gradient $\partial Q/\partial \eta$ around stoichiometric and the error can then be estimated as

$$\langle E_{\rm N} \rangle \approx \left(\rho_{\eta} \Delta N(\eta) P(\eta) \right)_{\eta_{\rm s}} \left[\frac{\partial Q}{\partial \eta} \right]^+$$

$$(4.18)$$

where the subscripts "+" and "-" refer to the gradients on the high and low η side of stoichiometric, $\eta = \eta_s$.

For the unconditional average enthalpy there will be a corresponding false radiation source term, $\langle E_{N,h} \rangle$

$$\langle E_{\rm N,h} \rangle = \int_{\eta_{\rm min}}^{\eta_{\rm max}} \rho_{\eta} \Delta N(\eta) P(\eta) \frac{\partial^2 Q_{\rm h}}{\partial \eta^2} d\eta \qquad (4.19)$$

Here there is no corresponding conserved scalar. Radiation losses ensure that $\partial^2 Q_h / \partial \eta^2$ is positive for a broad range of η around stoichiometric and positive values of the error $\Delta N(\eta)$ in this region will result in an overprediction of enthalpy and hence temperature.

4.3. Frozen and Fast Chemistry Limits

The frozen limit corresponds to the situation when chemical reactions do not occur (that is fuel and oxidizer are mixed in turbulent flow but do not react). In this case a reactive scalar is a linear function of the conserved scalar, $Y=a+b\xi$, which corresponds to $Q=a+b\eta$. The constants a and b determined by the values of reactive scalar before are mixing. Condition $Y=a+b\xi$ has been used in the derivation of the CMC equation in Section 3.1.2. Hence the CMC model must be consistent with $Y=a+b\xi$. This can be checked directly by substituting $Q=a+b\eta$, Y''=0 and $\langle W|\eta \rangle =0$ into identity obtained demonstrates that the CMC Eq.(4.2). The model is consistent with the frozen limit.

We consider next the case of fast chemical reaction which is opposite to the case just considered. When chemistry is fast, the reactive scalars tends to their equilibrium values: $Y \rightarrow Y_e(\xi)$ as $Da \rightarrow \infty$, where the Damkohler number is defined as $Da = \tau_L/\tau_c$, τ_L being the integral time scale of turbulence and τ_c a characteristic chemical time scale. The equilibrium concentration Y_e is then a function of the mixture fraction, ξ , and following Bilger⁷ we obtain

$$-N \frac{\partial^2 Y_{\rm e}}{\partial \xi^2} = W \tag{4.20}$$

This applies instantaneously at every point in the field. In order to make comparison with CMC we obtain the conditionally averaged version of this equation. Taking in to account that Y_e is a deterministic function of ξ , we apply Eq.(2.17). This yields $\langle Y_e(\xi) | \eta \rangle = Y_e(\eta)$. We obtain

$$-\langle N|\eta \rangle \frac{\partial^2 Y_e}{\partial \eta^2} = \langle W|\eta \rangle$$
(4.21)

This equation can be derived from Eq.(4.2) by taking the limit $Q \rightarrow Y_e(\eta)$ and $Y'' \rightarrow 0$. The CMC model is thus consistent with the fast chemistry limit (FCL). The solution of the CMC equation will automatically tend to this

limit when the Da number is sufficiently large.

It was concluded in Section 3.5.2 that the CMC model is correct when it describes processes with τ_c greater than the Kolmogorov scale τ_K . In the limit of fast chemistry, $\tau_c \ll \tau_K$, we have just shown that the fast chemistry limit is absolutely correctly described by the CMC model. The validity of CMC in between these limits is still an open question.

4.4. Laminar Flamelet Model

The stationary laminar flamelet model (SLFM), for which different versions have been suggested by $Kuznetsov^{71}$ and $Peters^{109}$, can be written in the form

$$-N_{\rm s} \frac{\partial^2 Y}{\partial \xi^2} = W(Y) \tag{4.22}$$

In spite of having quite a similar form to Eq.(4.20), these models have differences which are outlined further here. For a recent review of SLFM see Bray & Peters²².

FCL is a global model which covers all (or maybe a significant part of) the turbulent field. SLFM is a local model which specifies reaction processes inside very thin reaction zones. Strictly, SLFM is only applicable when the thickness of the reaction zone ℓ_{z} is much smaller than the Kolmogorov length scale $\ell_{\rm K}$. The conserved scalar dissipation $D(\nabla\xi)^2$ inside the reaction zone is then uniform and is denoted by N_s . Unlike N in Eq.(4.20) where N can vary markedly throughout the reaction zone, N_s in Eq.(4.22) is effectively a random parameter. In FCL $Y_{e}(\xi)$ is given. In SLFM Y is to be determined as the solution of Eq.(4.22). This solution requires boundary conditions at the bounds of the reaction zone $\xi = \eta_s - \eta_z$ and $\xi = \eta_s + \eta_z$ where $\xi = \eta_s$ is stoichiometric surface determining the location the reaction zone and $2\eta_z$ is the width of the reaction zone in conserved These conditions are usually specified as the equilibrium scalar space. conditions

$$Y=Y_e$$
 at $\xi=\eta_s-\eta_z$ and $Y=Y_e$ at $\xi=\eta_s+\eta_z$ (4.23)

 $Y=Y(\xi,N_s)$ The solution of Eq.(4.22) is thus function of the a instantaneous values of the conserved scalar and the instantaneous value of the scalar dissipation at stoichiometric. SLFM implicitly requires fulfilled. large Da number to have conditions in Eq.(4.23) In practice Eq.(4.22) with then. can be solved in the interval $\eta_{\min} \leq \xi \leq \eta_{\max}$ the boundary conditions

$$Y=Y_{\min}$$
 at $\xi=\eta_{\min}$ and $Y=Y_{\max}$ at $\xi=\eta_{\max}$ (4.24)

The solution at the outer edges of the reaction zone is normally very close to $Y_{\rm e}$. Well inside the reaction zone the deviations are larger since the derivative $\partial^2 Y_e / \partial \xi^2$ is usually large at $\xi = \eta_s$. In SLFM a flamelet library is generated for a range of $N_{\rm s}$, $Y(\xi, N_s),$ and unconditional averages can then be obtained by weighting this by the joint pdf of ξ and $N_{\rm s}$.

The CMC model has some similarities with the SLFM model but these models are not identical. If we conditionally average Eq.(4.22) we cannot split the correlation $\langle N_s \partial^2 Y / \partial \xi^2 | \eta \rangle$ since Y depends on N_s . We can demonstrate, however, that CMC, FCL and SLFM are in agreement integrally in the reaction zone. We integrate Eq.(4.22) between the limits $\xi = \eta_s - \eta_z$ and $\xi = \eta_s + \eta_z$ and utilize the boundary conditions (4.23)

$$N_{\rm s} \left[\frac{\partial Y}{\partial \xi} \right]_{\eta_{\rm s} - \eta_{\rm z}}^{\eta_{\rm s} + \eta_{\rm z}} = N_{\rm s} \left[\frac{\partial Y_{\rm e}}{\partial \xi} \right]_{\eta_{\rm s} - \eta_{\rm z}}^{\eta_{\rm s} + \eta_{\rm z}} = -\int_{\eta_{\rm s} - \eta_{\rm z}}^{\eta_{\rm s} + \eta_{\rm z}} W d\xi$$
(4.25)

Conditional averaging of Eq.(4.25) yields

$$-\langle N|\eta_{s}\rangle \left[\frac{\partial Y_{e}}{\partial \eta}\right]_{\eta_{s}-\eta_{z}}^{\eta_{s}+\eta_{z}} = \int_{\eta_{s}-\eta_{z}}^{\eta_{s}+\eta_{z}} \langle W|\eta\rangle d\eta \qquad (4.26)$$

Equation (4.26) coincides with the integral of Eq.(4.21) over the interval $\eta_s - \eta_z \le \eta \le \eta_s + \eta_z$. These equations are in agreement with the limit of the CMC equation at Da $\rightarrow\infty$. Note that inside the reaction zone, as it assumed in SLFM, $\langle N | \eta \rangle = \langle N | \eta_s \rangle$.

If the reaction zone is wider than the Kolmogorov length scale, the scalar disipation will vary significantly and randomly through the reaction zone so that strictly SLFM modelling is no longer applicable. In practice the dissipation averaged across the reaction zone N_z is often used instead

of N_s in Eq.(4.22) as suggested by Kuznetsov & Sabelnikov⁷². Often the value of N_z is also assumed to be a constant. The fluctuations of N_z are significantly smaller than the fluctuations of N_s . In many practical applications SLFM is used in the form

$$-\langle N | \eta_{s} \rangle \frac{\partial^{2} Y}{\partial \xi^{2}} = W(Y)$$

$$(4.27)$$

that is the fluctuations of N are neglected (sometimes it is also assumed that $\langle N | \eta_s \rangle = \langle N \rangle$ in Eq.(4.27)). Conditional averaging of Eq.(4.27) yields

$$-\langle N|\eta_{s} > \frac{\partial^{2} Q}{\partial \eta^{2}} = \langle W|\eta > = W_{\eta}(Q)$$
(4.28)

The difference between this equation and Eq.(4.21) is that Q is obtained as the solution of Eq.(4.28) and for simple one-step chemistry and finite Damkohler number Q is close to Y_e but does not coincide with $Y_{\rm e}$. For especially for hydrocarbon combustion, solutions for Q complex chemistry, be from full equilibrium can markedly different even at quite high Damkohler number. This is particularly so the rich side on of stoichiometric where peaks in CO are less than one third of those obtained flamelet calculations from equilibrium calculations. Such give much data⁸² improved predictions of experimental and have led to their applications. widespread use in practical Such success should not be allowed to overshadow the fact that the use of Eq.(4.28) is not soundly based.

Equation (4.28) can be also obtained from Eq.(4.2) by neglecting the convective terms and assuming that $\langle N|\eta \rangle = \langle N|\eta_s \rangle$. It is seen that the form of SLFM using Eq.(4.28) is a special case of the CMC model.

Various formulations of unsteady laminar flamelet models have been *et al*¹¹¹consider unsteady flamelets Mauss in a jet proposed. diffusion flame in a Lagrangian manner. From the flow and mixing caculation they deduce the variation of the average scalar dissipation with time. This is used in an unsteady laminar counterflow flamelet caculation to get the timeor space-dependent composition as a function of the mixture fraction. This function is then weighted by the local pdf to get al ¹¹⁵ consider unconditional averages. Pitsch et unsteady flamelets in introduce the concept of 'Representative diesel engines. They Interactive

Flamelets' which are applied in domains "..with statistically average quantities of flamelet parameters". In applications similar both of these of unsteady flamelets the equations being solved similar the are to that would be solved using CMC equations modeling. There are subtle In CMC the modeling assumptions are differences, however. much more transparently defined and the specification of the scalar dissipation and conditional velocity is precise. For the diesel engine problem CMC would retain the terms involving the spatial gradients of Q; these are neglected in the unsteady flamelet formulation. Furthermore, in CMC the requirement that the reaction zones be thin with respect to the Kolmogorov length scale is not necessary.

CMC, FCL and flamelet models have many similarities and belong to a category of non-premixed combustion models that use the concept of a conserved scalar and its dissipation. In general it can be seen that the CMC model is a more advanced model in which the temporal and spatial evolution of chemical processes are taken properly into account. This evolution is neglected in the SLFM and FC models. CMC is a more soundly based approach than concepts of partially-premixed and unsteady flamelets The SLFM, used in the form of Eq.(4.22), can which have been proposed. have some advantages in consideration of local processes when fluctuations of the conserved scalar dissipation are important. Local extinction is an example of such processes.

Clear delineation of the regimes of validity of CMC and SLFM awaits For $\theta < \tau_c < \tau_K$ (one strict condition for validity further investigation. of SLFM) CMC may not be valid even though it is valid at the fast chemistry limit, $\tau_c/\tau_K = 0$. This seems likely to be so for one-step irreversible chemistry and perhaps for multistep chemistry involving irreversible reactions. reversible reactions involving For the term the scalar dissipation becomes of second order at high Damkohler number, the balance being between the forward and backward rates, and the first order solution is that for equilibrium, i.e. FCL. In such cases the width of the reaction zone, ℓ_z , may be broader than the Kolmogorov length scale, $\ell_{\rm K}$, It seems that if this is so, both CMC and SLFM may even though $\tau_c < \tau_K$. strictly valid and may not give the correct results for the not be departure from the first order solution, which is given by chemical equilibrium.

4.5. Some Further Results for Homogeneous Turbulence

we consider some further properties of the CMC equation which Here than the properties considered in previous sections. more complicated are In order avoid complications we assume that the turbulence is to homogeneous but the results obtained could have general significance. more In homogeneous turbulence with uniform and constant density Eqs.(4.1)-(4.3) take the form

$$\frac{\partial QP(\eta)}{\partial t} = \langle W|\eta \rangle P(\eta) + \frac{\partial}{\partial \eta} \left(\langle N|\eta \rangle P(\eta) \frac{\partial Q}{\partial \eta} - \frac{\partial \langle N|\eta \rangle P(\eta)}{\partial \eta} Q \right)$$
(4.29)
$$\frac{\partial Q}{\partial t} - \langle N|\eta \rangle \frac{\partial^2 Q}{\partial \eta^2} = \langle W|\eta \rangle$$
(4.30)

$$\frac{\partial P(\eta)}{\partial t} = -\frac{\partial^2 \langle N | \eta \rangle P(\eta)}{\partial \eta^2}$$
(4.31)

Eq.(4.29) is a combination of Eqs.(4.30) and (4.31)and can be easily derived from them. If the conserved scalar pdf $P(\eta)$ is specified and if $\langle W(\xi, Y) | \eta \rangle$ is approximated by $W(\eta, Q)$, the system of Eqs.(4.30) and (4.31) is closed. The conditional scalar dissipation $\langle N|\eta \rangle$ can be determined from Eq.(4.31). The boundary conditions determined by Eqs.(2.57) and (2.58) for smooth pdfs or by Eqs.(2.68) and (2.69) for intermittent pdfs should be integrate Eq.(4.31) twice over η . The boundary conditions used to in Eqs.(2.57) and (2.58) seem to be overdetermined. In fact they are not. The pdf is not an arbitrary function. It must satisfy certain normalization restrictions (for example, the constraint: a pdf integral all η is always unity). It is possible to select any over two boundary conditions from Eqs.(2.57) and (2.58) and the other two must be fulfilled choice should such minimize automatically. In practice the be as to numerical integration errors. Using boundary conditions Eq.(2.68) the in and (2.69) or in Eq.(2.62) we obtain

$$\langle N|\eta \rangle = \frac{1}{P(\eta)} \begin{pmatrix} \eta & \eta_{\max} \\ \int & \eta_{\max} \\ \eta_{\min} \\ \eta_{\min}$$

The conditional scalar dissipation determined by Eq.(4.32) is utilized to solve Eq.(4.30) for the conditional expectation of a reactive scalar.

4.5.1. Low probability effects

The pdf $P(\eta)$ is not explicitly involved in Eq.(4.30) and any η in the interval $\eta_{min} < \eta < \eta_{max}$ has equal significance for Eq.(4.30). Some values of probability $P(\eta)$ and should not may have very small be $\eta_{min} < \eta < \eta_{max}$ important. Intuitively, these ranges of η with small probability should not affect the solution of Eq.(4.30) in the high-probability regions. On the face of it, this seems to be a contradiction, but in fact it is not. conditional In order to demonstrate this we assume that the scalar dissipation is approximated by

$$\langle N | \eta \rangle = \langle N \rangle \equiv N_{a}(t)$$
 (4.33)

with This approximation is consistent there being Gaussian pdf a The satisfying Eq.(4.31). approximation in Eq.(4.33) is not an approximation which can be always used for Eq.(4.30): it is used here as a simple example for the demonstration of some quite general properties of $\theta(t) \equiv \langle (\xi')^2 \rangle$ the CMC equations. The unconditional variance ξ'≡ξ-<ξ> satisfies the equation 115,135

$$\frac{d\theta}{dt} = -2N_{\rm a}(t) \tag{4.34}$$

We introduce a new independent variable

$$\frac{d\tau}{dt} = N_{a}(t) \quad \text{or} \quad \tau = \tau_{0} + \int_{t_{0}}^{t} N_{a}(t^{\circ}) dt^{\circ}$$
(4.35)

Equations (4.30) and (4.31) then take the form

$$\frac{\partial Q}{\partial \tau} = \frac{\partial^2 Q}{\partial \eta^2} - \frac{\langle W | \eta \rangle}{N_a}$$
(4.36)

$$\frac{\partial P(\eta)}{\partial \tau} = -\frac{\partial^2 P(\eta)}{\partial \eta^2}$$
(4.37)

The variable τ can be thought of as the "diffusion time". This variable determines the solutions of Eqs.(4.36) and (4.37). Combining Eq.(4.34) and Eq.(4.35) we obtain

$$2(\tau - \tau_0) = \theta_0 - \theta \tag{4.38}$$

where $\theta_0 \equiv \theta(t_0)$. The conserved scalar fluctuations disappear in a sufficiently long time interval $\theta \rightarrow 0$, $\tau - \tau_0 \rightarrow \theta_0/2$ as $t \rightarrow \infty$ and

$$P(\eta) \rightarrow \delta(\eta \rightarrow \infty)$$
 as $t \rightarrow \infty$. (4.39)

Equation (4.36) is parabolic, and from the theory of such equations it is known that any changes to Q at $t=t_1$, $\eta=\eta_1$ affect Q for all η at any $t>t_1$. In practice, however, only the limited region $\eta_1-\Delta\eta<\eta<\eta_1+\Delta\eta$, where

$$\Delta \eta \sim (\tau - \tau_1)^{1/2}$$

and $\tau_1 \equiv \tau(t_1)$ is significantly affected. Since

$$\tau - \tau_1 = \frac{\theta_1 - \theta}{2} \leq \frac{\theta_1}{2}$$

by analogy with Eq. (4.38), this region is bounded for any $t > t_1$

$$\Delta \eta \leq \operatorname{const}(\theta_1)^{1/2}$$

This means that the CMC equations can be solved for quite a large range of η , but the solution in ranges of η which have a small probability $P(\eta)$ cannot significantly affect the solution in the high probability range of η . It also means that at large times, when the range of the pdf is remote from the initial bounds of η , solution can proceed over a much narrower range of η with arbitrary values of $\langle N | \eta \rangle$ and boundary conditions for Q being used at values of η greater than about four standard deviations from the mean. This result is also likely to apply to high values of η in the far downstream regions of jets and wakes, where it is cumbersome to compute all the way to a mixture fraction of unity and the numerical

solution of the pdf transport equation gives unreliable values for the conditional scalar dissipation at high values of η .

4.5.2. Boundary conditions

The observations in the text immediately preceding this bring up the question of what are the general effects of moving boundary conditions on Q and $\langle N | \eta \rangle$

The most complicated case is the case of а smooth pdf with This could happen for species when the non-constant bounds η_{\min} and η_{\max} . unmixed fluid is undergoing reaction or is being fed to the system with a varying For enthalpy it might also arise time composition. from compression or expansion of the unmixed fluid such as happens in a diesel First we engine during injection and ignition. check if the reactive species conservation integral is preserved in this case. Equation (4.29) is integrated over the interval $\eta_1 \leq \eta \leq \eta_2$ where $\eta_{\min} < \eta_1 < \eta_2 < \eta_{\max}$. The last two terms on the right-hand side of Eq.(4.29) are integrated by parts

$$\int_{\eta_{1}}^{\eta_{2}} \frac{\partial QP(\eta)}{\partial t} d\eta = \frac{\partial}{\partial t} \int_{\eta_{1}}^{\eta_{2}} QP(\eta) d\eta - \left[\frac{\partial \eta_{i}}{\partial t} QP(\eta)\right]_{\eta_{1}}^{\eta_{2}} =$$
$$= \int_{\eta_{1}}^{\eta_{2}} \langle W|\eta \rangle P(\eta) d\eta + \left[\langle N|\eta \rangle P(\eta) \frac{\partial Q}{\partial \eta} - \frac{\partial \langle N|\eta \rangle P(\eta)}{\partial \eta} Q\right]_{\eta_{1}}^{\eta_{2}}$$
(4.40)

where η_i is for either η_1 or η_2 . Assuming $\eta_1 = \eta_{min} + \varepsilon$ and $\eta_2 = \eta_{max} - \varepsilon$ and taking the limit $\varepsilon \rightarrow 0$ we obtain

$$\frac{\partial \langle Y \rangle}{\partial t} - \langle W \rangle = \left[\langle N | \eta \rangle P(\eta) \frac{\partial Q}{\partial \eta} + J_{\rm P}^{\circ} Q \right]_{\rm min}^{\rm max} = 0$$
(4.41)

where

$$J_{\rm P}^{\circ} \equiv P(\eta) \frac{\partial \eta_{\rm m}}{\partial t} - \frac{\partial \langle N | \eta \rangle P(\eta)}{\partial \eta}$$
(4.42)

We use here the boundary conditions in Eqs.(2.57) and (2.58) which

indicate that $J_{P}^{\circ} \rightarrow 0$ and $\langle N | \eta \rangle P(\eta) \rightarrow 0$ as $\eta \rightarrow \eta_{m}$. It is easy to see that Eq.(4.41) preserves the conservation integral for any finite values of $[\partial Q/\partial \eta]_{\rm m}$. In the case of non-homogeneous turbulence $[Q]_{\rm m}$ and and and η_{max} the conservation integral also non-constant η_{min} is preserved provided $P(\eta) < \mathbf{v}'' Y'' | \eta > \nabla \eta_m \rightarrow 0$ as $\eta \rightarrow \eta_m$. This condition is usually fulfilled in practical cases.

Preserving the reactive species conservation integral determines boundary conditions (4.13) in the case of intermittent pdfs but it seems from the above that in the case of smooth pdfs the boundary conditions remain undetermined. This needs further consideration. The boundary values $[Q]_{min}$ and $[Q]_{max}$ are restricted by the CMC equation itself since η_{min} and η_{max} are special points of this equation. We introduce a new independent variable $\eta_{+} \equiv \eta - \eta_{m}$ in Eq.(4.29). If we consider the boundary conditions at η_{min} then $\eta_{m} = \eta_{min}$ and $\eta_{+} \ge 0$. If we consider the boundary conditions at η_{max} then $\eta_{m} = \eta_{max}$ and $\eta_{-} \le 0$. Taking into account the differentiation chain

$$\left(\frac{\partial}{\partial t}\right)_{\eta=\text{const}} = \left(\frac{\partial}{\partial t}\right)_{\eta_{+}=\text{const}} - \frac{d\eta_{\text{m}}}{dt} \frac{\partial}{\partial \eta_{+}}$$
(4.43)

we obtain

$$\frac{\partial QP(\eta)}{\partial t} - \langle W|\eta \rangle P(\eta) = \frac{\partial}{\partial \eta_{+}} \left(J_{P}^{\circ}Q + \langle N|\eta \rangle P(\eta) \frac{\partial Q}{\partial \eta_{+}} \right) = \\ = \frac{\partial J_{P}^{\circ}}{\partial \eta_{+}} Q + \frac{\partial \langle N|\eta \rangle P(\eta)}{\partial \eta_{+}} \frac{\partial Q}{\partial \eta_{+}} + J_{P}^{\circ} \frac{\partial Q}{\partial \eta_{+}} + \langle N|\eta \rangle P(\eta) \frac{\partial^{2}Q}{\partial \eta_{+}^{2}} \quad (4.44)$$
$$\frac{\partial P(\eta)}{\partial t} = \frac{\partial J_{P}^{\circ}}{\partial \eta_{+}} \qquad (4.45)$$

where J_P° is introduced in Eq.(4.42). We use Eq.(4.45) to cancel the first term on the right-hand side of Eq.(4.44). According to Eqs.(2.57) and (2.58), $J_P^{\circ} \rightarrow 0$ and $\langle N | \eta \rangle P(\eta) \rightarrow 0$ as $\eta_+ \rightarrow 0$. We assume that $[Q]_m \neq 0$. This assumption is not restrictive since we always can introduce a new scalar $Y^{\circ} = Y + a$ (*a*=const) which conditional expectation satisfies Eq.(4.30). Hence, in the vicinity of $\eta_+ = 0$, we can neglect the two last terms on the right-hand side of Eq.(4.44)

$$\left[\left(\frac{\partial Q}{\partial t} \right)_{\eta_{+}} - \langle W | \eta \rangle - \frac{I}{P(\eta)} \frac{\partial \langle N | \eta \rangle P(\eta)}{\partial \eta_{+}} \frac{\partial Q}{\partial \eta_{+}} \right]_{m} = 0$$
(4.46)

where $[\cdot]_m$ is for the limit of (\cdot) at $\eta \rightarrow \eta_m$. Taking into account that $[J_P^\circ]_m = 0$ and using Eqs. (4.42) and (4.43) we obtain

$$\left[\left(\frac{\partial Q}{\partial t} \right)_{\eta_{+}} - \frac{\partial \eta_{m}}{\partial t} \frac{\partial Q}{\partial \eta_{+}} - \langle W | \eta \rangle \right]_{m} = \left[\left(\frac{\partial Q}{\partial t} \right)_{\eta} - \langle W | \eta \rangle \right]_{m} = 0 \quad (4.47)$$

coincides with The derivative $dQ_{\rm m}/dt$ (where $Q_{\rm m}(t) \equiv [Q]_{\rm m}$) limit of the $[(\partial Q/\partial t)_{\eta}]_{\rm m}$. Equation (4.47)determines the boundary derivative conditions for Q at $\eta = \eta_{\min}$ and $\eta = \eta_{\max}$. Note that Eq.(4.47) can be formally derived from Eq.(4.30) by putting $[\langle N|\eta \rangle]_m = 0$. This is in agreement with Eqs.(2.60) and (2.63) and can be used to obtain the boundary conditions in the case of non-homogeneous turbulence.

4.6 Overview

The CMC equation, (4.1) or (4.2), is to be solved over the space and domain interest subject to initial and boundary conditions time of problem being studied. Before moving on to consider appropriate for the simplified forms of the equations and more advanced modelling questions it seems appropriate to pause and review the nature of CMC modeling.

The modeling assumptions involved in deriving Eqn (4.1) and (4.2) are diffusion the molecular level is Fickian with that mass at all Lewis unity, Reynolds numbers are moderate, at least, and that the numbers primary closure assumption of Section 3.3 is valid. The first two practice in turbulent assumptions are normal combustion modelling. The effects of differential diffusion can be incorporated into the CMC method as is discussed in Section 9. In Section 3.7, theoretical support for the of the primary closure assumption is validity given but ultimately validation comes from studies involving Direct Numerical Simulation (DNS) and successful applications of CMC in a wide range of reacting flows. This evidence is presented in Sections 5 and 7. It is believed that the primary closure assumption is valid for cases well removed the from the fast flows showing quite chemistry limit and in large conditional average conditional average. fluctuations about the Further work is needed to firmly, particularly in flows with ignition and establish this extinction

occurring.

Other modeling assumptions are of a secondary nature. The closure of the conditional chemical reaction rate $\langle W | \eta \rangle$ is discussed in Section 3.5 and will often be possible at the first moment level as given by Eq. this closure to be valid, conditional (3.34).For average fluctuations of mass fractions and temperature about their conditional means must species be small, particularly for reactions having high activation energies. In Section made 8 we summarise the progress so far on modeling the conditional average variance and covariance balance equations and with second order closure for the chemical reaction rates. Such modeling is helping to elucidate the conditions under which first order closure will be valid. At this stage, no simple criteria can be recommended.

Modeling of the conditional scalar dissipation $\langle N | \eta \rangle$ is discussed in Section 4.1 and errors associated with the poor estimation of it is discussed in Section 4.2. This question is discussed further in Sections 5 and 7. Some flows are quite sensitive to the scalar dissipation and others are not. The situation depends quite subtly on the flow and the chemistry and it is not possible at this stage to recommend simple criteria. Models for $\langle \mathbf{v} | \eta \rangle$ and $\langle \mathbf{v}'' Y'' | \eta \rangle$ are also discussed in Section 4.1. At this stage little experience has been gained in validation of these models. It can be noted here that modeling of the mixing at this first-order closure level is confined to conserved scalars for both mixing at the molecular level, as in $\langle N | \eta \rangle$, and at the turbulent flux level, as in $\langle v | \eta \rangle$ and $\langle v'' Y'' | \eta \rangle$. Mixing of both these sorts for reactive scalars is known to be fraught with difficulty since the chemistry can have a major influence on setting concentration gradients and countergradient turbulent fluxes can also be combustion²¹ This is most evident in turbulent premixed but obtained. will also be important in nonpremixed systems. Modeling for the molecular with mixing terms is a persistent problem the Monte-Carlo/joint pdf method ¹¹⁴ and the role of the chemistry in the determination of the mixing rate has not yet been satisfactorily treated. The fact first that order CMC only considers conserved scalars is a distinct advantage of the method. It is noted also that conserved scalars are only needed for the of the flow and field. Modeling of modeling mixing reactive scalar however, in second order closure CMC dissipation terms does appear, and this is discussed in Section 8.

In applications of CMC to individual problems care is needed in the specification of the initial and boundary conditions for Q. Examples will

be found in Sections 5 to 7. Often the two boundary conditions needed for Q in η space are taken as those in the unmixed fluid at $\eta = 0$ and 1. This is usually found to be satisfactory even though there may be a total lack of one or both of the unmixed fluids in parts of the space time domain that the findings in homogeneous flows being calculated. It appears of Section 4.5 are applicable here. In such places it matters little what values are taken for the boundary condition as its effects will not be felt in the range of η of practical interest.

It seems, then, that CMC has a very sound basis in theory and that the modeling assumptions made are expressible mathematically and can be individually checked. This transparency of the modeling is a distinct advantage for CMC over models of a more conceptual kind such as the linear eddy model 53 or laminar flamelet models used beyond the scope of their strict applicability.

5. CMC IN TURBULENT SLENDER LAYER FLOWS

The general CMC equation (4.2) remains rather complicated for general term $\langle \mathbf{v}'' \mathbf{Y}'' | \eta \rangle$ is not easy to The close satisfactorily and the use. closure of Eq.(4.5) for $\langle v | \eta \rangle$ is not well established. Of more practical the large number of independent variables involved. Here we concern is consider some techniques that transform the general CMC equation (4.2)well-founded models which are much simpler and easier to use. The into techniques are applicable to turbulent shear flows and other thin layers such non-bouyant plumes and scalar mixing layers in homogeneous as turbulence. For such flows the CMC equations can be significantly simplified. Many of the comparisons of CMC predictions with experiment made for flows of this type using these simplifications, have been and hence much of the evidence for the validity of CMC modeling rests on the further simplifying assumptions. Accordingly, validity of these we present in some detail here the theoretical basis for these simplifications, practical correct concerns about the modeling of the conditional scalar dissipation, and some comparison of predictions with experiment.

and Bilger^{10,12} suggested two different forms of the Klimenko⁵⁴ CMC these flows. At first equation for slender layer these equations were considered to be approximate equations based plausible on some Bilger^{10,12} supported assumptions. his assumptions by comparison with experimental measurements. Later Klimenko⁵⁷ showed that both forms of the simplified CMC shear flow equation are correct asymptotic limits of the CMC equation, having similarities with the traditional general some boundary layer analysis. The structure of the expansions is, however, equations for conventional quite different from the traditional means in start our consideration of slender layer flows from slender layers. We asymptotic the main results of this analysis and in the following two Bilger ^{10,12} "simple" method proposed by sections present the and the Klimenko⁵⁴. The method proposed by Integral method is the Integral prferred method where assurance of accuracy is of prime importance.

The Integral method requires evaluation of a weighted integral of the flow. conditional average scalar dissipation across the This can be obtained from the pdf transport equation after suitable integrations across the flow and in mixture fraction space. Once again, subtleties are introduced for consideration of either intermittent or smooth pdfs. These are explored in Section 5.4. The nature of the pdf boundaries are also

important in considering the preservation of conservation integrals. as 4.2 already considered in Section for the non-Integral case. Α brief outline of this problem together with methods for assessing the errors are given in Section 5.5. Both of these sections can be omitted on the first reading.

Evaluation of the conditional average scalar dissipation is not always straight forward and small numerical inaccuracies can result in which are, of course, physically impossible. negative values Experimental measurements, results for self-similar flows and approximate methods that have been used in the literature are reviewed in Section 5.6.

Measurements in jet diffusion flames³ indicate that there are small variations in conditional average temperature, cross-stream and that these could be important in predicting nitric oxide formation. In Section 5.7 possibility of predicting these from higher the order terms in the asymptotic expansion is outlined.

CMC predictions and comparison with experiment are presented in Section 5.8 for a reacting scalar mixing layer, turbulent jet diffusion flames and for round plumes in grid turbulence.

5.1. Basics of the Asymptotic Analysis

Slender layer flows are characterized by small ratio l_2/l_1 where l_2 transverse and longitudinal characteristic length and l_1 are scales. In section the longitudinal components are denoted by x_1 and v_1 ; this the transverse components are denoted by x_2 and v_2 . We introduce a small parameter $\varepsilon^2 = l_2/l_1$ which is common for all boundary layer asymptotic similar methods. The analysis we consider here is to the traditional which layer methods simplify the unconditionally averaged boundary (see Hinze⁴⁴). Navier-Stokes and scalar transport equations in shear flows Thus in estimates of all variables in the CMC equation we follow Hinze's estimates. The asymptotic analysis is quite complicated and we consider a brief review of its results. According to Klimenko⁵⁷ here only and Klimenko & Bilger⁶⁰ the conditional expectation Q in shear flows is given by the series

$$Q = Q_0(x_1, \eta) + \varepsilon Q_1(x_1, \eta) + \varepsilon^2 Q_2\left(x_1, \eta, \frac{\eta - \langle \xi \rangle}{\varepsilon}\right) + \dots$$
(5.1)

The second term in this series, Q_1 , can be used⁶⁰ to obtain higher order

corrections for the CMC shear flow equation. As follows from the expansion (5.1) the conditional expectations, Q, are a weak function of transverse coordinate This seems unexpected since the x_2 . the unconditional mean, $\langle Y \rangle$, is a strong function of x_2 ; but this is predicted by the asymptotic analysis and is supported by experimental data^{10,12} as is seen in Fig. 1 A nother supplementary result of the analysis is the representation of the conserved scalar pdf in the form of the series

$$P(\eta') = \frac{a_1}{\langle N | \eta' \rangle} \exp \left(-a_2 \int_0^{\eta'} \frac{\langle v_2 | \eta' \rangle}{\langle N | \eta' \rangle} d\eta \right) + \dots$$
(5.2)

where $\eta' \equiv \eta - \langle \xi \rangle$ and a_1 , a_2 do not depend on η' . The asymptotic analysis shows that $a_2 \gg I$ and that the pdf $P(\eta)$ has a strong dependence on x_2 .

5.2. Simple Method

Assuming that Q does not depend on x_2 , Bilger^{10,12} neglected the transport across the flow and suggested that in turbulent shear flows Q satisfies the equation

$$U^{\circ} \frac{\partial Q_0}{\partial x_1} - N^{\circ} \frac{\partial^2 Q_0}{\partial \eta^2} = W(\eta, Q)$$
(5.3)

The index "₀" indicating that Q_0 is the main order approximation of Q. The coefficients U° and N° are given by

$$U^{\circ} = U_{a} \Big(x_{1}, x_{2}^{\circ}(x_{1}, \eta) \Big); \qquad N^{\circ} = N_{a} \Big(x_{1}, x_{2}^{\circ}(x_{1}, \eta) \Big); \tag{5.4}$$

where x_2° is the solution of the equation $\xi_a(x_1, x_2^{\circ}) = \eta$ and the new functions introduced here are

$$U_{a}(x_{1},x_{2}) \equiv \langle v_{1} \rangle; \quad N_{a}(x_{1},x_{2}) \equiv \langle N \rangle; \quad \xi_{a}(x_{1},x_{2}) \equiv \langle \xi \rangle$$

The values of U° and N° are specified as the corresponding mean values taken at $x_2=x_2^{\circ}$ which is usually close to the most probable location of the isopleth $\xi(t,\mathbf{x})=\eta$: more precisely, at $x_2=x_2^{\circ}$ the mean value $\langle \xi \rangle$ coincides with η . For jets, wakes and plumes the mean mixture fraction on the

centreline of the flow is much less than unity in downstream regions. A problem with this method is the assignment of U° and N° for values of η greater than the centreline mean. The conditional expectation of the source term is given by the approximation in Eq.(3.37). The asymptotic analysis⁵⁷ indicates that Eq.(5.3) is the correct asymptotic limit of the CMC equation as $\varepsilon \rightarrow 0$ everywhere except at the centerline of the flow.

5.3. Integral Method

Assuming steady-state flow we integrate Eqs.(4.1) and (4.3) across the flow

$$\frac{\partial}{\partial x_{1}} \left\{ \langle v_{1}Y|\eta \rangle P(\eta)\rho_{\eta} \right\} - \left\{ \langle W|\eta \rangle P(\eta)\rho_{\eta} \right\} =$$

$$= \frac{\partial}{\partial \eta} \left\{ \langle N|\eta \rangle P(\eta)\rho_{\eta} \frac{\partial Q}{\partial \eta} - \frac{\partial \langle N|\eta \rangle P(\eta)\rho_{\eta}}{\partial \eta}Q \right\} =$$

$$= \frac{\partial}{\partial \eta} \left\{ 2\langle N|\eta \rangle P(\eta)\rho_{\eta} \frac{\partial Q}{\partial \eta} \right\} - \frac{\partial^{2}}{\partial \eta^{2}} \left\{ \langle N|\eta \rangle P(\eta)\rho_{\eta}Q \right\}$$
(5.5)

$$\frac{\partial}{\partial x_1} \left\{ < \mathbf{v}_1 | \boldsymbol{\eta} > P(\boldsymbol{\eta}) \boldsymbol{\rho}_{\boldsymbol{\eta}} \right\} = - \frac{\partial^2}{\partial \boldsymbol{\eta}^2} \left\{ < N | \boldsymbol{\eta} > P(\boldsymbol{\eta}) \boldsymbol{\rho}_{\boldsymbol{\eta}} \right\}$$
(5.6)

where the curly brackets denote integrals across the flow

$$\left\{ \cdot \right\}_{\mathrm{R}} \equiv \int (\cdot) dx_2; \quad \left\{ \cdot \right\}_{\mathrm{R}} \equiv \int \int (\cdot) dx_2 dx_3; \quad \left\{ \cdot \right\} \equiv \lim_{\mathrm{R} \to \infty} \left\{ \cdot \right\}_{\mathrm{R}}$$
(5.7)
$$|x_2| \leq R \qquad \qquad x_2^2 + x_3^2 \leq R^2$$

The first formula in Eq.(5.7) is used for two-dimensional flows and the second formula is used for three-dimensional flows. Equations (5.5) and (5.6) are valid for any chosen η which belongs to the open interval $\eta_{\min} < \eta < \eta_{\max}$, where η_{\min} and η_{\max} denote the pdf bounds which are assumed to be constants. We consider the case when $\xi \rightarrow \eta_{\min}$ and $P(\eta) \rightarrow \delta(\eta - \eta_{\min})$ as $R \rightarrow \infty$ where $R \equiv |x_2|$ in two-dimensional flows or $R^2 \equiv x_2^2 + x_3^2$ in three-dimensional flows. The value of η_{\min} which corresponds to ξ far from the turbulent flow is a constant. The pdf $P(\eta)$ rapidly tends to zero as $R \rightarrow \infty$ for any fixed $\eta > \eta_{\min}$. Hence the integrals in Eqs.(5.5) and (5.6) are finite for

 $\eta > \eta_{\min}$. In two-dimensional flows (such as a mixing layer) another flow configuration is possible: $\xi \rightarrow \eta_{\min}$ as $x_2 \rightarrow -\infty$ and $\xi \rightarrow \eta_{\max}$ as $x_2 \rightarrow +\infty$. In this case (which is not specifically discussed in this paper) the bound η_{\max} should be considered in a similar way to the bound η_{\min} . The integrals $\{\cdot P(\eta)\}$ can take zero, finite or infinite values at $\eta = \eta_{\min}$ depending on the pdf model used.

Now we take into account that $Q \approx Q_0$ does not depend on the coordinates across the flow and put Q_0 in Eq.(5.5) outside of the integration brackets. For example, the most restrictive term in Eq.(5.5) is modified so that

$$2\frac{\partial}{\partial\eta}\left\{ \langle N|\eta \rangle P(\eta)\rho_{\eta}\frac{\partial Q}{\partial\eta} \right\} = 2\frac{\partial}{\partial\eta} \left\{ \langle N|\eta \rangle P(\eta) \right\} \left(\frac{\partial Q_{0}}{\partial\eta}\rho_{\eta} + O(\varepsilon) \right) \right\}$$
(5.8)

The conditional expectation of the density, ρ_{η} , is considered to be a function of η and Q_0 . According to the approximation of Eq.(3.37), $\langle W|\eta \rangle$ can also be removed outside the integration brackets. Turbulent transport in the longitudinal direction by conditional fluctuations is neglected so that

$$\langle \mathbf{v}_1 Y | \eta \rangle \approx \langle \mathbf{v}_1 | \eta \rangle Q_0 \approx \langle \mathbf{v}_1 \rangle Q_0$$

$$\tag{5.9}$$

This is a common assumption in turbulent shear flows. Approximation (5.9) is supported by the asymptotic analysis. We introduce the new notation

$$P^{*} = \left\{ P(\eta) \right\}; \quad U^{*} = \frac{\left\{ < v_{1} \mid \eta > P(\eta) \right\}}{\left\{ P(\eta) \right\}}; \quad N^{*} = \frac{\left\{ < N \mid \eta > P(\eta) \right\}}{\left\{ P(\eta) \right\}}$$
(5.10)

and write Eqs.(5.5) and (5.6) in the form

$$\frac{\partial}{\partial x_1} \left(U^* Q_0 P^* \rho_{\eta} \right) - W(Q_0) P^* \rho_{\eta} = \frac{\partial}{\partial \eta} \left(N^* P^* \rho_{\eta} \frac{\partial Q_0}{\partial \eta} - \frac{\partial N^* P^* \rho_{\eta}}{\partial \eta} Q_0 \right)$$
(5.11)
$$\frac{\partial}{\partial x_1} \left(U^* P^* \rho_{\eta} \right) = -\frac{\partial^2}{\partial \eta^2} \left(N^* P^* \rho_{\eta} \right)$$
(5.12)

Another form of the CMC equation for shear flows is derived from Eqs.(5.11) and (5.12)

$$U^* \frac{\partial Q_0}{\partial x_1} - N^* \frac{\partial^2 Q_0}{\partial \eta^2} = W(Q_0)$$
(5.13)

similar to Eq.(5.3) but these equations have different Equation (5.13) is formulas for their coefficients. Equation (5.3)is easier in practical use than Eq.(5.13) since solving of Eq.(5.3)does not require any pdf modeling. Equation (5.13) is expected to be close to the correct near centerline and, as is proved in the next subsections, it preserves Equation (5.12)conservation integrals. is an exact integral of the pdf equation. Equations (5.3), (5.11)and (5.13)are approximate equations which utilize the approximation of Q in Eq.(5.1). Equations (5.3), (5.11)and (5.13) are valid for turbulent shear flows which are characterized by of the characteristic scales a small ratio across and along the flow. (5.13) effectively replaces the coordinate across Equation the flow x_2 by the new coordinates related to moving surfaces ξ=const. Diffusion in scalar space is determined universal small-scale conserved by fluctuations for which the conditional dissipation is the measure.

The coefficients of Eq.(5.13) are specified provided conserved the Indeed, the values of P^* and U^* can readily be scalar pdf $P(\eta)$ is known. determined from Eq.(5.10) using the approximation of Eq.(5.9). The coefficient N^* cannot be determined from Eq.(5.10), however, since $\langle N | \eta \rangle$ is unknown. The best way is to determine N^* by integration of Eq.(5.12). This requires the specification of the boundary conditions for the product N^*P^* . These are considered next.

5.4. Boundary Conditions for the Pdf Integrals

face of it, determining the boundary conditions for On the the pdf integrals across the flow should be similar finding the boundary to conditions for the pdfs. This would involve the following

$$\lim_{\eta \to \eta_{\rm m}} P^* \equiv \lim_{\eta \to \eta_{\rm m}} \lim_{R \to \infty} \left\{ P \right\}_{\rm R} = \lim_{R \to \infty} \lim_{\eta \to \eta_{\rm m}} \left\{ P(\eta) \right\}_{\rm R} = \lim_{R \to \infty} \left\{ \lim_{\eta \to \eta_{\rm m}} P(\eta) \right\}_{\rm R} (5.14)$$

where $\xi \rightarrow \eta_m$ as $R \rightarrow \infty$. On more careful consideration, however, it appears that Eq.(5.14) is not always valid since the interchange of the order of and $R \rightarrow \infty$ requires special conditions. We note that the the limits $\eta \rightarrow \eta_m$ which integration over integration $\{\cdot\},$ is an an infinite interval. involves the limit operation $R \rightarrow \infty$ as is indicated in Eq.(5.7). Normally in engineering applications, differentiating, integrating and taking limits are considered as interchangeable operations and we have followed this practice elsewhere. There is, however, an exception in the case of Eq.(5.14). Interchanging the order of the limits and $R \rightarrow \infty$ needs η→η_m special consideration. We can demonstrate that Eq.(5.14) is valid for intermittent pdfs but the order of the limits $\eta \rightarrow \eta_{\min}$ and $R \rightarrow \infty$ cannot be interchanged for smooth pdfs. In intermittent turbulent flows the pdf is determined by Eq.(2.64) and the intermittency factor γ_t (and γ_{max}) rapidly tend to zero as $R \rightarrow \infty$. This means that if R is large the pdf is small for all η which belong to the open interval $\eta_{min} < \eta < \eta_{max}$. Almost all of the probability is accumulated in the delta function $\gamma_{\min}\delta(\eta-\eta_{\min})$ and does affect the integral for $\eta_{min} < \eta < \eta_{max}$. Thus in numerical calculations not can reasonably cut off our integrals somewhere at large R. The we situation with smooth pdfs is totally different. These pdfs are large at $\eta \sim <\xi >$ even if R is large. Since $<\xi > \rightarrow 0$ as $R \rightarrow \infty$, this does not affect the vicinity of η_{max} and the asymptotes $\eta \rightarrow \eta_{max}$ of the integrals across the flow present no difficulties. The asymptote of the integral P^* of a smooth pdf $P(\eta)$ at $\eta \rightarrow \eta_{\min}$ is different from the asymptote of the pdf $P(\eta)$ itself. Let us consider the moment integral

$$M_{\alpha} \equiv \langle (\xi - \eta_{\min})^{\alpha} \rangle = \int_{\eta_{\min}}^{\eta_{\max}} (\eta - \eta_{\min})^{\alpha} P(\eta) d\eta \quad (5.15)$$

It is evident that $(\eta - \eta_{\min})^{\alpha} \rightarrow I$ as $\alpha \rightarrow 0$ everywhere except at $\eta = \eta_{\min}$ where $(\eta - \eta_{\min})^{\alpha} = 0$. This is why $M_{\alpha} \rightarrow I$ as $\alpha \rightarrow 0$ for smooth pdfs and $M_{\alpha} \rightarrow \gamma_t + \gamma_{\max} \eta_{\max}^{\alpha}$ as $\alpha \rightarrow 0$ for intermittent pdfs. If $\alpha > 0$ is fixed, M_{α} rapidly tends to zero as $R \rightarrow \infty$ for both types of the pdfs since $(\xi - \eta_{\min})^{\alpha} \leq \exp(-\alpha aR)$ for large R and constant α . The integral

$$\left\{ M_{\alpha} \right\} = \left\{ \begin{array}{c} \eta_{\max} \\ \int (\eta - \eta_{\min})^{\alpha} P(\eta) d\eta \\ \eta_{\min} \end{array} \right\} = \left\{ \begin{array}{c} \eta_{\max} \\ - \int (\eta - \eta_{\min})^{\alpha} P^* d\eta \\ \eta_{\min} \end{array} \right. (5.16)$$

exists for any $\alpha > 0$ but, if $P(\eta)$ is smooth, $\{M_a\} \rightarrow \infty$ as $\alpha \rightarrow 0$. This can be valid only if

$$P^* \sim \frac{1}{\eta - \eta_{\min}}$$
 as $\eta \to \eta_{\min}$ (5.17)

asymptote of the conditional dissipation at The be also $\eta \rightarrow \eta_{\min}$ can The formula relating surface to volume assessed. ratio for an isopleth surface^{8,114} ξ=const can be integrated across flow yield the to the isopleth surface area per unit length in the x_1 direction: it is given by $S_n = (N^*/D)^{1/2} P^*$. In planar two-dimensional flows at large distance from the turbulent region S~const. This gives

$$N^* \sim (\eta - \eta_{\min})^2$$
 as $\eta \rightarrow \eta_{\min}$ (5.18)

For the smooth pdfs the asymptote of P^* is always determined by Eq.(5.17) irrespective of the asymptote of $P(\eta)$ at $\eta \rightarrow \eta_{\min}$, while for intermittent pdfs the asymptotes of P^* and $P(\eta)$ at $\eta \rightarrow \eta_{\min}$ are similar. We emphasize that Eqs.(5.17) and (5.18) are only approximate asymptotes. The asymptote of N^* at $\eta \rightarrow \eta_{\max}$ is similar to the asymptote for $\langle N | \eta \rangle$ given in Eq.(2.63) $(\eta_{\max}=\text{const})$ which also mirrors Eq.(5.18).

The purpose of the rest of this section is to derive and prove the boundary conditions for the product N^*P^* . This condition enables us to solve Eq.(5.18) for N^*P^*

5.4.1. Intermittent pdfs

In the of intermittent pdfs, Eqs.(2.68) (2.69)case and can, according our previous analysis, be generalized for the integrals to across the flow so that

$$N^*P^* \to 0$$
 as $\eta \to \eta_m$ (5.19)

5.4.2. Smooth pdfs

In order to investigate the boundary conditions for the integrals of smooth pdfs we consider the integral of Eq.(2.53) across the flow

$$\frac{\partial}{\partial x_1} \left\{ \langle v_1 \rho F \rangle \right\} = - \left\{ \langle \rho N F'' \rangle \right\}$$
(5.20)

As stated earlier, stationary flow is assumed. The arbitrary good function $F(\eta)$ is chosen so that $F(\eta_{\min})=0$ that is $F \rightarrow 0$ as $R \rightarrow \infty$. This ensures that the integral on the left-hand side of Eq.(5.20) exists. The integral on the right-hand side of Eq.(5.20) exists since in the flow $\xi \rightarrow \eta_{\min}$ and $N \rightarrow 0$ as $R \rightarrow \infty$. We multiply Eq.(5.12) by $F(\eta)$ and integrate it over the interval $\eta_1 \leq \eta \leq \eta_2$ where $\eta_1 = \eta_{\min} + \varepsilon$ and $\eta_2 = \eta_{\max} - \varepsilon$ and ε is small. The integral on the right-hand side is integrated by parts. Taking the limit $\varepsilon \rightarrow 0$ we obtain

$$\frac{\partial}{\partial x_1} \left\{ \langle v_1 \rho F \rangle \right\} - \left\{ \langle \rho N F'' \rangle \right\} = \left[\frac{\partial}{\partial \eta} \left(N^* P^* \rho_\eta \right) F \right]_{\max} + \left[N^* P^* \rho_\eta F' \right]_{\min}^{\max}$$
(5.21)

Matching Eq.(5.21) and Eq.(5.20) for arbitrary F_{max} , F'_{max} and F'_{min} yields the boundary conditions for the pdf integrals across the flow

$$N^*P^* \to 0$$
 as $\eta \to \eta_{\min}$ (5.22)

$$N^*P^* \to 0$$
 as $\eta \to \eta_{\max}$ (5.23)

$$\frac{\partial}{\partial \eta} \left(N^* P^* \right) \to 0 \quad \text{as} \quad \eta \to \eta_{\text{max}}$$
 (5.24)

If we assume that η_{max} is a function of x_1 , the revised version of Eq.(5.24) includes some additional terms⁶². Since $F(\eta_{\min})=0$ Eq.(5.21) does not specify the boundary conditions for the derivative of $N^{*}P^{*}$ at $\eta \rightarrow \eta_{\min}$. Considering smooth pdfs with constant bounds, one might expect that Eq.(2.61) can be generalized for the integrals across the flow to yield $\partial(N^*P^*)/\partial\eta \rightarrow 0$ as $\eta \rightarrow \eta_{\min}$. This equation is not valid, however, because of the reasons discussed above: Equations (5.17)and (5.18)yield $N^*P^* \sim (\eta - \eta_{\min})$ as $\eta \rightarrow \eta_{\min}$ and the derivative has a finite value.

5.4.3 Summary of the boundary conditions

The boundary condition $N^{\uparrow}P^{\uparrow} \rightarrow 0$ as η tends to its limiting values valid cases considered in this are for all section. and η_{min} η_{max} Integration across the flow may, however, change the rate how the product $N^{*}P^{*}$ tends to zero. Thus, certain care is required when the asymptote of the product $N_n P(\eta)$ is integrated across the flow.

The asymptotical behavior of N^* and P^* must be taken into account when the integration of $P(\eta)$ across the flow is conducted numerically. Practically, intermittent pdfs are safer to use since for these pdfs it is easier to perform integration across the flow. The correct treatment of the boundary conditions is important also for preserving the conservation integrals.

5.5. Conservation Integrals

The integral form of the CMC equation for shear flows. Eq. N^{*} (5.13), preserves conservation integrals provided that the values of and P^* used are consistent with Eq.(5.12). The CMC equation should be always used in conjunction with its adjoint equation, given here by the equation (5.12). The conservation integrals across the flow integral pdf are finite provided $Q(\eta_{\min})=0$ where $\xi \rightarrow \eta_{\min}$ as $R \rightarrow \infty$. If we consider the case $Q(\eta_{\text{max}})=0$ intermittent pdfs, also require that avoid of we to the reactive scalar Y from the non-turbulent transport of the region at $\eta = \eta_{max}$. It is apparent that these integrals will not be finite for species Y non-zero flow. This does not restrict with in the outer our since we always can introduce a new scalar Yand consideration of them its conditional expectation Q° similar that used in Eq.(4.14). Integrating Eq.(5.11) between the limits $\eta_{\min} + \varepsilon$ and $\eta_{\max} - \varepsilon$ and letting ε tend to zero we obtain

$$\frac{\partial}{\partial x_1} \left\{ \langle v_1 \rho Y \rangle \right\} - \left\{ \langle \rho W \rangle \right\} = \left[Q \frac{\partial}{\partial \eta} \left(N^* P^* \rho_\eta \right) \right]_{\min}^{\max} + \left[N^* P^* \rho_\eta \frac{\partial Q}{\partial \eta} \right]_{\min}^{\max} = 0 \quad (5.25)$$

used the boundary conditions given in Eqs.(5.19), We have (5.22)and also, (5.23).It is required that $[Q]_{\min} = Q(\eta_{\min}) = 0$ and for intermittent This specifies the boundary conditions pdfs, that $[Q]_{\max} = Q(\eta_{\max}) = 0.$ for

the limits of Q at $\eta \rightarrow \eta_m$. These conditions are quite obvious. For smooth pdfs the boundary condition for Q at $\eta \rightarrow \eta_{max}$ is not determined. According to the analysis of Sec.4.5.2 (note that $[N^*]_{max}=0$), this boundary condition is given by

$$\left[U^* \frac{\partial Q}{\partial x_1} - W(Q)\right]_{\max} = 0$$
(5.26)

5.5.1. Errors arising from inconsistent scalar dissipation

It has been emphasised that the conditional scalar dissipation used in solving Eq.(5.13) must be fully consistent with that which satisfies Eq.(5.12) for a given pdf field. If this is not the case then the conservation integrals will not be satisfied. We seek here to quantify the errors involved. The results parallel those for the general case given in Section 4.2.3.

Let us assume that the value of N^* actually used in Eq.(5.13) is $N^* + \Delta N^*$ where N^* is the value that satisfies Eq.(5.12). It is easy to show that Eq.(5.25) then becomes

$$\frac{\partial}{\partial x_1} \left\{ \langle v_1 \rho Y_i \rangle \right\} - \left\{ \langle \rho W_i \rangle \right\} = \int_{\eta_{\min}}^{\eta_{\max}} \rho_\eta \Delta N(\eta)^* P^* \frac{\partial^2 Q_{0,i}}{\partial \eta^2} d\eta \equiv E_N^*$$
(5.27)

We will call the error, $E_{\rm N}^*$, the "integrated false chemistry source term". because it is evident that weighting Eq. (5.27) by suitable This is coefficients, μ_i , and summing over appropriate species to form a conserved scalar, η_* , will eliminate this term if the aforesaid boundary conditions This is because all such conserved scalars should satisfied. be are initially (i.e. at the upstream start of the calculation) linear functions of η and will then accordingly remain so, since

$$E_{\rm N}^{*} = \mu_{\rm i} \int_{\eta_{\rm min}} \rho_{\eta} \Delta N(\eta)^{*} P^{*} \frac{\partial^{2} Q_{0,\rm i}}{\partial \eta^{2}} d\eta = \int_{\eta_{\rm min}} \rho_{\eta} \Delta N(\eta)^{*} P^{*} \frac{\partial^{2} \eta_{*}}{\partial \eta^{2}} d\eta = 0 \qquad (5.28)$$

where $\xi_* \equiv \mu_i Y_i$, $\eta_* \equiv \langle \xi_* | \eta \rangle = \mu_i Q_{0,i}$, $\rho \mu_i W_i = 0$ and sum is taken over

repeated indices.

At high Damkohler number, the mass fractions of reactant and product species tend to be linear functions of η outside of a narrow reaction zone centred at $\eta = \eta_s$. For such species the error due to the false chemistry term, E_N^* , is readily shown to be

$$E_{\rm N}^* = \left(\rho_{\rm \eta} \Delta N^* P^*\right)_{\rm \eta_s} \left[\frac{\partial Q_0}{\partial {\rm \eta}}\right]_{\rm -}^+$$
(5.29)

where the subscripts "+" and "-" signify evaluation of the slope at values of η greater and than η_s . For intermediate and radical species, less similar formulae can be derived if their dependence of Q_0 on η is treated piecewise linearly. Alternatively, error be evaluated the can from Eq.(5.27)

For the enthalpy there will result a similar integrated false radiation loss term

$$E_{\rm N,h}^* = \int_{\eta_{\rm min}} \rho_{\eta} \Delta N^* P^* \frac{\partial^2 Q_{0,h}}{\partial \eta^2} d\eta$$
(5.30)

5.6. Evaluation of the Conditional Scalar Dissipation

we consider evaluation of the this section conditional average In N^{*} , scalar dissipation integrated across the flow. as defined in Eq.(5.10). For the stationary slender layer flows under consideration here, N^* can in principle be evaluated from the pdf transport equation, integrated across the flow, Eq.(5.12). Using the boundary conditions for N^*P^* at $\eta = 0$ and 1 as set out in Section 5.4, Eq.(5.12) may be integrated by parts to yield

$$\rho_{\eta}N^{*}P^{*} = \frac{\partial}{\partial x} \left[\int_{\eta}^{I} \rho_{\eta}U^{*}P^{*}(\eta')(\eta-\eta')d\eta' \right] =$$

$$= \frac{\partial}{\partial x} \left\{ \overline{\rho} \widetilde{u} \int_{\eta}^{I} \widetilde{P}(\eta', r)(\eta - \eta') d\eta' \right\}$$
(5.31)

second version involves working with the Favre pdf, $\tilde{P}(\eta)$, where the flows defined in Eq.(4.4). For combusting density-weighted or Favre usually used and the first and second moments of the mixture averaging is and $(\xi'')^2$, ξ are solved for in the mixing fraction, calculation. А presumed form for $\tilde{P}(\eta)$ is assumed so that $\tilde{P}(\eta)$ can be evaluated at each the flow and Eq.(5.31) is evaluated numerically. It should be point across noted that the approximation of setting the conditional average axial velocity component to the unconditional Favre average axial velocity component $\tilde{u} = \langle \rho v_1 \rangle / \langle \rho \rangle$ has been used - an assumption equivalent to that We shall call evaluation of N^* from Eq.(5.31) made in Eq. (5.9). the direct method. In practice the inaccuracies in numerical integration and differentiation can lead to significant errors, even to the extent that unphysical negative values are obtained. It is evident that more robust methods are needed.

For flows without heat release, self-similar mixing fields are possible such as in the classical mixing layer, scalar mixing layer, jets For these flows, self similar-solutions and small deficit wakes. for the N[°]. conditional dissipation, result and be can evaluated without approximation from experimental data on the pdfs. These results are of general interest since they give examples of the effects of flow geometry on the form of N^* and allow general solutions of the CMC equation to be obtained for various forms of the chemical source term. In the following sections general considerations are given for self-similar flows, and simplified for jets these are further and small-deficit wakes. Results are then obtained for the small-deficit wake, using the experimental data for pdfs of LaRue and Libby 74 .

of particular In combustion a flow interest is the turbulent iet This flow is not self similar owing to the heat release, diffusion flame. due to the presence of a co-flow and/or buoyancy. A and often, also, robust method is needed for these flows. A method based on the assumption of local self-similarity appears to be robust and accurate. It is presented in Section 5.6.3.

In Section 5.6.4 we comment briefly on other methods for obtaining N^* .

5.6.1. Results for self-similar flows

In this section we consider some examples of the functions P^* and N^* which are consistent with Eq.(5.12), and following Klimenko et al 62 , we concentrate on self-similar solutions. The integrals P^* , U^* and $N^{\hat{}}$ are functions of x_1 and η . Self-similarity in the flows of interest ensures that they can be scaled to be functions of just a normalized conserved scalar variable $\eta \equiv \eta/(\eta_c - \eta_{min})$ where η_c is a characteristic value for the x_1 . Furthermore, scalar η at a particular power-law scaling can be expected so that

$$\eta_{c} \equiv \eta_{R} X^{-\beta \eta}$$

$$U^{*} = U_{R} \hat{U}(\hat{\eta}) X^{-\beta_{u}}$$

$$N^{*} = (U_{R} \eta_{R}^{2} / \ell_{R}) \hat{N}(\hat{\eta}) X^{-\beta_{N}}$$

$$P^{*} = \ell_{R}^{r} \hat{P}(\hat{\eta}) X^{\beta_{p}} / \eta_{c}$$

where $X \equiv x_1/\ell_R$, and U_R , ℓ_R and η_R are reference values for velocity, length and the scalar chosen for each particular flow. For two-dimensional (planar) flows r=1, while for three-dimensional (axisymmetric) flows r=2. The functions \hat{U} , \hat{N} and \hat{P} are non-dimensional functions of $\hat{\eta}$ but not of X. Substituting the self-similar forms into equation (5.12) we obtain

$$\lambda^{\circ} \hat{U} \hat{P} + \beta_{\eta} \frac{d}{d\eta} \left(\hat{\eta} \hat{U} \hat{P} \right) + \frac{d^2}{d\eta^2} \left(\hat{N} \hat{P} \right) = 0$$
(5.32)

with

$$\beta_{\rm N} = \beta_{\rm u} + 2\beta_{\eta} + 1$$
; $\lambda^{\circ} \equiv \beta_{\rm p} - \beta_{\rm u}$

Values of β_N , β_u , β_p , β_η can be determined for each particular type of flow and some examples are shown in Table 1. For homogeneous turbulence, wakes and scalar mixing layers, the value of \hat{U} is a constant. The self-similar solution for $P(\eta)$ and $\langle N|\eta \rangle$ in homogeneous turbulence suggested by Sinai and Yakhot¹¹⁹ is included in (5.32) as a special case $\lambda^{\circ}=0.$

Turbulent jets and wakes with $\eta_{min}=0$ are now considered further. Let us integrate (5.32) over $\hat{\eta}$ between the limits $\hat{\eta} > \hat{\eta}_{min}$ and $\hat{\eta}_{max}$ (the bounds which correspond to η_{min} and η_{max}). We have

$$\hat{\eta}_{\text{m a x}} \\ \lambda \int_{\hat{\eta}} \hat{U} P d \hat{\eta}^{\circ} - \hat{\eta} \hat{U} P - \frac{1}{\beta_{\eta}} \frac{d}{d \hat{\eta}} \left(\hat{N} P \right) = 0$$
(5.33)

where $\lambda \equiv \lambda^{\circ} / \beta_{\eta}$ Note that in self-similar flows the variable $\mathring{\eta}$ must be chosen so that $\mathring{\eta}_{\min} = 0$ and $\mathring{\eta}_{\max}$ is either infinite or dependent on X. If $\mathring{\eta}_{\max} = \infty$, the integration constant has zero value, as the function \mathring{P} tends exponentially to zero at $\mathring{\eta} \rightarrow \infty$. In the case of a finite value of $\mathring{\eta}_{\max}$ Eq.(5.33) is still valid⁶². Further integration between the limits $\mathring{\eta}$ and $\mathring{\eta}_{\max}$, taking into account the boundary condition $\mathring{NP} \rightarrow 0$ at $\mathring{\eta} \rightarrow \mathring{\eta}_{\max}$ in Eq.(5.23) yields

$$\frac{\hat{NP}}{\beta_{\eta}} = \int_{\hat{\eta}}^{\hat{\eta}_{max}} \left(\lambda\hat{\eta} + (1-\lambda)\hat{\eta}^{\circ}\right) \hat{U}(\hat{\eta}^{\circ})\hat{P}(\hat{\eta}^{\circ})d\hat{\eta}^{\circ}$$
(5.34)

The evident inequality $\hat{P} \ge 0$ leads us to the conclusion that the boundary condition $\hat{NP} \rightarrow 0$ at $\hat{\eta} \rightarrow \hat{\eta}_{max} = 0$ in Eq.(5.22) can be satisfied only if $\lambda = 1$. This is an eigenvalue of the problem under consideration and it is in agreement with Table 1. Equation (5.34) takes the form

$$\frac{\hat{N}\hat{P}}{\hat{\beta}_{\eta}} = \hat{\eta}\int_{\hat{\eta}}^{\hat{n}_{max}} \hat{U}(\hat{\eta}^{\circ})\hat{P}(\hat{\eta}^{\circ})d\hat{\eta}^{\circ}$$
(5.35)

This method of obtaining N^* in terms of P^* has direct application for CMC modeling. The direct problem (that is expressing \hat{P} in terms of \hat{N} and \hat{U}) also has an analytical solution in this case

$$\hat{P}(\hat{\eta}) = \text{const} \quad \hat{\underline{\eta}}_{N(\hat{\eta})} \exp\left(-\beta_{\eta} \int_{0}^{\hat{\eta}} \hat{\underline{\eta}}_{N(\hat{\eta})} \cdot \frac{\hat{U}(\hat{\eta})}{N(\hat{\eta})} d\hat{\eta}^{\circ}\right)$$
(5.36)

The structure of equation (5.36) has some similarities with the pdf solutions suggested by Sinai and Yakhot¹¹⁹ and with Eq.(5.2).

5.6.2. Results for the planar wake

Figure 11 shows results for the pdf's in the wake of a heated circular cylinder as obtained by LaRue and Libby⁷⁴. The pdf's have been scaled so that $\eta \equiv \Delta T / \Delta T_c$, where ΔT is the temperature increase above that of the external flow and $\Delta T_{\rm c}$ is the mean temperature excess on the centerline (it is assumed that $\Delta h = C_p \Delta T$, $C_p = \text{const}$). The data⁷⁴ are shown here with the associated with uncontaminated external fluid intermittent spike omitted. This has been subtracted according to the method of Bilger, Antonia and Sreenivasan¹⁵. The symbol $\hat{P}^{\circ}(\hat{\eta})$ is used to indicate that these are the pdf's at a point in the wake and not those integrated across the wake as in Eq. (5.10). The pdf's are shown at various distances from the centerline y normalized by the length scale for the width of the wake $l_c \equiv (xd_0)^{1/2}$ where x is stream-wise distance.

Figure 12 shows values of $P(\eta)$ obtained by integrating the data of Fig. 11 across the flow. Here r=1 so that Eq.(5.10) takes the form

$$P^{*} = (xd_{0})^{1/2} \hat{P}(\hat{\eta}) / \Delta T_{c} = 2 \frac{(xd_{0})^{1/2}}{\Delta T_{c}} \int_{0}^{\infty} \hat{P}^{\circ}(\hat{\eta}) d(y/l_{c})$$
(5.37)

The reference length of the flow has been chosen as the cylinder diameter $\ell_{\rm R}=d_0$ although there are arguments to say that it should be $C_{\rm D}d_0$, where $C_{\rm D}$ is the drag coefficient of the cylinder. The other reference values are taken as $\eta_{\rm R}=l_c\Delta T_c/d_0$ and $U_{\rm R}=U_{\rm e}$. It is seen that there is a peak in $\hat{P}(\hat{\eta})$ for values of the scalar just less than the mean on the centerline. A further peak is associated with fluid that is close to the scalar value of the external flow.

For scalars other than temperature P^* can be evaluated by replacing

 $\Delta T_{\rm c}$ by the mean value of the scalar on the centerline (zero in external flow). Figure 13 shows values of $N(\eta)$ obtained by integration of the $P(\eta)$ of Fig. 12 using Eq.(5.35). Since for this flow $\beta_{\rm N}=2$ and $\beta_{\eta}=1/2$ we have

$$N^* = \frac{U_e}{x} \left(\Delta T_c\right)^2 \hat{N}(\hat{\eta})$$
(5.38)

again this may be evaluated for wakes of scalars other Once than temperature by using the mean value of the scalar on the centerline (zero in external flow) in place of ΔT_c . With the $x^{-1/2}$ dependence of this centerline mean it is seen that $N^* \sim x^2$ in a planar wake as indicated in Table 1. The shape of $\hat{N}(\hat{\eta})$ is interesting. It indicates that N^* is highest for material that has a scalar value of about half of the mean centerline value and that material of low scalar values or high scalar values has much smaller N^* . This is consistent with the notion that material near the extremes of concentration must have low gradients so that the bounds are not exceeded. This is unlike the result in homogeneous turbulence with a linear mean scalar profile where the point values of $\langle N|\eta \rangle$ are lowest near η equals mean and rise away from this local mean^{45,146}. When these $\langle N | \eta \rangle$ profiles are integrated across the flow to give N^* and $N(\eta)$, however, the result will be a much less pronounced dependence on $\hat{\eta}$. It is noted that the two-dimensional wake will not be self-similar if there is heat release and these results will not apply directly to such a case. They do. however, give an indication of the form that can be expected for the N^* and P^* profiles in cases with heat release.

5.6.3. Method for local self similarity in jet flames

Kronenburg *et al*⁶⁹ have considered the evaluation of the scalar dissipation from Eq.(5.31) for jet flames. They introduce a scaled value of η , $\hat{\eta} = \eta/\xi_c$ where ξ_c is the Favre averaged value of the mixture fraction on the centreline. They show that Eq.(5.31) may be rewritten as

$$\rho_{\eta}N^{*}P^{*} = \left\{\overline{\rho}N_{\eta}\widetilde{P}_{\eta}\right\} = -\frac{\partial \ln(\xi_{c})}{\partial x} \left\{\overline{\rho}\widetilde{u}\eta\int_{\eta}^{T}\widetilde{P}(\eta^{\circ})d\eta^{\circ}\right\} +$$

$$+ \frac{\partial}{\partial x} \left| \bigwedge_{\eta}^{\wedge} \left\{ \overline{\rho} \widetilde{u} \xi_{c} \int_{\eta}^{\varepsilon^{-1}} \widetilde{P}(\eta^{\circ})(\eta^{\circ} \eta^{\circ}) d\eta^{\circ} \right\}$$
(5.39)

In this equation $\widetilde{P}(\hat{\eta}^{\circ})$ is the Favre pdf scaled to $\hat{\eta}$ and evaluated at the dummy value $\hat{\eta}^{\circ}$. There is no approximation involved in Eq.(5.39). In the region well downstream of the nozzle, $\xi_c \ll l$ and there is no contribution to the pdf integral in the second term on the right hand side from values of $\hat{\eta}^{\circ}$ near $1/\xi_c$. Under these conditions this last term on the RHS will be zero if the flow is locally self similar. This can easily be shown⁶⁹ by scaling the radial profiles by their centerline values and the jet half radius: the double integration over the flow and in scaled mixture fraction space is then for x independent functions and so this double integral can be taken outside the x derivative. The combination of scaling left inside the x derivative is proportional to the values total flux of mixture fraction and this is essentially constant since mean mixture fraction is conserved and the contribution to the total flux of the term involving the axial turbulent flux has already been neglected (see Eqs (5.31), (5.9) and (4.5)).

Neglecting the second term on the RHS of Eq.(5.39) gives

$$\rho_{\eta}N^{*}P^{*} = \left\{\overline{\rho}N_{\eta}\widetilde{P}_{\eta}\right\} = -\frac{\partial \ln(\xi_{c})}{\partial x} \left\{\overline{\rho}\widetilde{u}\eta\int_{\eta}^{I}\widetilde{P}(\eta^{\circ})d\eta^{\circ}\right\}$$
(5.40)

We shall call the evaluation of N^* using this equation the *quasi-self* similarity method. It is seen that positive values of N^* are guaranteed since the mean mixture fraction on the centerline falls with increasing x. Furthermore, the pdf integral will be easy to evaluate well downstream of the nozzle, as there will be no peak near $\eta = I$.

Kronenburg *et al*⁶⁹ have evaluated the accuracy of this quasi-self similarity method for modeling a turbulent jet diffusion flame of hydrogen in air. They find that the method is accurate for $x/d_j > 30$, where d_j is the nozzle jet diameter. Beyond $x/d_j = 120$ the error begins to rise and accuracy can be improved by evaluating the second term in Eq.(5.39) as a correction term. For $x/d_j < 30$, the quasi-self similarity method was found to give results at least as good as those obtainable with the direct method.

Smith *et al*¹²⁴ modeled a turbulent jet diffusion flame of hydrogen in air. For N^* they used a locally homogeneous flow assumption essentially incorporating the method developed for homogeneous flows by Girimaji⁴¹. Further details may be found in Smith¹²¹. The method is robust, always giving positive values of N^* , and the errors involved are probably quite minor for the flow studied.

Problems involved in evaluating N^{*} by the direct method of Eq.(5.31) can be severe when using the Beta function as the presumed form for the mixture fraction pdf, Eq.(1.5). Kronenburg *et al*⁶⁹ use a logarithmic transformation of the mixture fraction suggested by Swaminathan and find that the numerical integrations involved are more robust. Unphysical negative values of N^{*} are still sometimes found, however.

experimental measurement Direct of conditionally averaged scalar dissipation has been made in homogeneous turbulence with a mean scalar gradient by Jayesh and Warhaft⁴⁵ and in other uniform density shear flows by Kailsanath, et al^{49} and Mi et al^{92} . Values of $N(\eta)$ are reported for various positions in the flows. Starner *et al*¹²⁸ report measurements of N^* as a function of mixture fraction in a turbulent jet diffusion flame of Their results are shown in Fig. 14 for $x/d_i = 25$ at air-diluted methane. four different jet Reynolds numbers obtained by varying the jet velocity. The Favre average mixture fraction on the centreline, ξ_c , for these data are close to 0.69 for the two lower Reynolds numbers and to 0.63 at the two higher Reynolds numbers. The results do not show the expected increase of N° with jet velocity, possibly due to the effect of the coflow on spreading rates. The results show a much slower increase of N with η near $\eta = 0$ than is shown for the uniform density wake data in Fig. 13 The sudden peaks in the range $\eta = 0.65 - 0.85$ possibly arise from low number statistics on the jet centerline.

It may be feasible to use such experimental data to relate N° to the unconditional dissipation $\langle N \rangle$ integrated in some way across the flow. It is not clear at this stage whether this sort of modelling would preserve consistency with the pdf transport equation. The errors in **Q** may, however, be small and worth the reduction in computational effort required to evaluate N^{*} from Eq.(5.31) or approximations to it.
5.7. Corrections of Higher Order

Equations (5.3) and (5.13) are approximations which effectively use the properties of turbulent shear flows. We may need, in some cases, to assess the practical precision of these approximations. For this purpose, Klimenko and Bilger⁶⁰ found the equation for the next term $Q_{\pm} \equiv \epsilon Q_1$ in (5.1). In general, this term is expected expansion to be small. The derivation of these corrections requires many steps and is not given here. We present only the main results obtained in Ref. 60 This section can be omitted at first reading. The main idea of the derivation is to retain the terms of the next order in Eq.(5.8) and evaluate them using the asymptotic expansions. The final equation for the corrections is given by

$$U^{*} \frac{\partial Q_{+}}{\partial x_{1}} - N^{*} \frac{\partial^{2} Q_{+}}{\partial \eta^{2}} =$$

$$= \frac{2}{P^{*}(I+\alpha_{t})} \frac{\partial}{\partial \eta} \left(P^{**} \left(U^{**} \frac{\partial Q_{0}}{\partial x_{1}} - N^{**} \frac{\partial^{2} Q_{0}}{\partial \eta^{2}} - W(\eta, Q_{0}) \right) \right)$$
(5.41)

where

$$P^{**} = \left\{ <\xi > P(\eta) \right\}; \quad U^{**} = \frac{\left\{ <\xi > P(\eta) \right\}}{\left\{ <\xi > P(\eta) \right\}}; \quad N^{**} = \frac{\left\{ <\xi > P(\eta) \right\}}{\left\{ <\xi > P(\eta) \right\}}$$

and Q_0 satisfies Eq.(5.13). It is assumed in Eq.(5.41) that the flux diffusion conditional is approximated by the approximation $< v_2'' Y'' | \eta > = -D_t \partial Q / \partial x_2$, and that the value $\alpha_t \equiv D_t (\partial < \xi > / \partial y)^2 / < N >$ is assumed to be constant for the purpose of this estimation. It is easy to see that if $\langle v_1 | \eta \rangle$ and $\langle N | \eta \rangle$ do not depend on x_2 and x_3 then $U^{**} = U^*$, $N^{**} = N^*$ and the side of Eq.(5.41) is zero. In this case, term on the right-hand the with solution of Eq.(5.41) the boundary conditions $Q_{+}(\eta_{\min})=0$ and $Q_{+}(\eta_{\text{max}})=0$ is $Q_{+}=0$. No correction is needed. If $\langle v_{1}|\eta \rangle$ and $\langle N|\eta \rangle$ depend on x_2 then the term on the right-hand side of Eq.(5.41) is non-zero. Such values of Q_{+} indicate certain error in the CMC shear flow non-zero

equation. It is expected that, under normal conditions, the value of Q_+ is much smaller than Q_0 .

data in turbulent jet flames³ indicate that small variations Recent conditional averages of temperature in the and species occur mass the flow. These variations should be predictable by fractions across the term in Q_2 in Eq.(5.1). At this stage, the investigation of the variations of Q across the flow are not completed, but some progress with it is given in Ref 60

5.8. Results for Some CMC Predictions

In this section we review some applications of CMC to turbulent shear flows and other turbulent slender layer flows for which the methods of Section 5 are applicable. In most cases the calculations have not been carried out using the full rigour for the calculation of the conditional scalar dissipation recommended here. Even so the predictions made show generally good agreement with experimental data.

5.8.1. Reacting scalar mixing layer

Figure 15 shows a schematic of a reacting scalar mixing layer as used in the experiments of Bilger $et \ al^{16}$. Upstream of the turbulence grid the streams are separated by a splitter plate and contain small amounts of nitric oxide (stream 1) and ozone (stream 2) uniformly mixed in air. The concentrations used were of the order of *l* part per million (ppm) on a molar basis and so the heat release and associated density change are negligible. For these reactants the rate of reaction is proportional to the reactant concentrations with product of the the rate constant being 0.37 ppm⁻¹s⁻¹at 20°C. Downstream of the grid the mean velocity U $= \langle v_1 \rangle$ uniform and constant and the turbulence decaying, is is somewhat uniform its standard deviations anisotropic and in in the cross-stream (x_2, x_3) directions. Mixing of the two streams is confined to a narrow but growing region in the x_2 direction. with a characteristic thickness $\ell_2 = \ell_2(x_1)$ which is defined as the distance between the x_2 coordinates where the mean mixture fraction is 0.1 and 0.9. Values of $\ell_2/x_1 \sim 0.3$ in

the region where measurements were made but $d\ell_2/dx_1$ is much lower than this.

Figure 16 shows measurements of the conditional average $Q_1 = \langle Y_1 | \eta \rangle$ normalised by the unmixed value $Y_{1,1}$ at various positions across the layer. It is seen that in accord with the asymptotic analysis of Section 5.1, there is no significant dependence of $Q_1(\eta)$ on x_2 .

CMC modeling of the reacting scalar mixing layer has been carried out¹² using somewhat crude assumptions about the conditional average of the scalar dissipation which amount to taking N^* in Eq. (5.13) independent of η

$$N^* = AU/x_1$$

where $A \sim 0.04$ is a constant for this flow. Since there is no variation in the mean streamwise velocity we have

$$U^* = U$$

Normalization of the reactant mass fractions, Y_{i} ,

$$\hat{Q}_{i} = \frac{Y_{i}}{M_{i}(Y_{1,1}/M_{1} + Y_{2,2}/M_{2})}$$

where M_i is the molecular weight of species *i*, leads to the relation

$$\hat{Q}_2 = \hat{Q}_1 - \eta + \eta_s$$

and the normalized CMC equation

$$\frac{\partial \hat{Q}_1}{\partial \hat{x}_1} = \hat{Q}_1 \left(\hat{Q}_1 - \eta + \eta_s \right) + \frac{1}{2} A \hat{x}_1^{-1} \frac{\partial^2 \hat{Q}_1}{\partial \hat{x}_1^2}$$
(5.42)

Here

$$\eta_{\rm s} \equiv \frac{Y_{2,2}}{M_2(Y_{1,1}/M_1 + Y_{2,2}/M_2)}$$

is the stoichiometric mixture fraction and

$$\hat{x}_1 \equiv N_{\rm D} x_1 / M$$

where M is the mesh size of the turbulence generating grid and N_D is a Damkohler number defined

$$N_{\rm D} \equiv kM_{\rm a}(Y_1/M_{1,1} + Y_2/M_{2,2})M/U$$

Here M_a is the molecular weight of air and k is the rate constant. Eq.(5.42) has only two parameters, A and η_s and \hat{Q}_1 is a function of the two independent variables \hat{x}_1 and η . Boundary conditions for \hat{Q}_1 were taken as

$$\hat{Q}_1 = \eta(1 - \eta_s), \text{ at } \hat{x}_1 = 0;$$
$$\hat{Q}_1 = 0, \text{ for } \eta = 0;$$
$$\hat{Q}_1 = 0, \text{ for } \eta = 0;$$

and

$$\hat{Q}_1 = l - \eta_s$$
, for $\eta = l$.

Eq. (5.42) is readily integrated numerically. Figure 17 shows solutions obtained for A = 0.06 and $\eta_s = 0.5$. Solutions at $x_1 = 0$ and ∞ correspond to the frozen and fast chemistry limits as discussed in Section 4.3. It is seen that the solutions always lie between these two limits and that the fast chemistry limit is approached for high Damkohler numbers and large distance downstream from the turbulence generating grid. At the fast chemistry limit the solution is bilinear with the break point on the η axis at $\eta = \eta_s$. Variation of the parameter η_s varies the solutions accordingly.

The effect of the parameter A is shown in Fig. 18 where solutions for $\eta = 0.5$ are shown as a function of \hat{x}_1 . A = 0 corresponds to the so-called reaction-dominated limit obtained by neglecting the mixing term in Eq. (5.13).At this limit there is a balance between the convective and reactive terms in Eq. (5.13). It is seen that A = 0.03 gives a very good fit to the experimental data¹⁶. This corresponds to N^* being equal to the flow estimated unconditional dissipation the centreline for on this On this basis, data from other scalar mixing layers gives experiment. values of A as high as 0.06. It is seen that at realistic values of A the results are substantially above that for the reaction-dominated limit. In

fact all three terms in Eq. (5.42) are of the same order for realistic values of A.

With A = 0.03 it was stated¹⁶ that the solution of Eq. (5.42) at $\hat{x}_1 = 8.9$ if plotted on Fig. 16 would accurately pass through the data points shown. In the light of error estimates for N^* made in Section 5.4.3, this seems a little surprising since no allowance was made in the predictions for the dependence of N^* on η . Li and Bilger⁷⁶ carried out calculations with a model for the scalar dissipation which incorporated a realistic dependence of $\langle N | \eta \rangle$ on η and found only small variations in the results.

5.8.2. Turbulent jet diffusion flames

CMC predictions for kinetically limited combustion including radicals formation have been carried out^{69,121,123,124,125} for turbulent jet and NO diffusion flames of hydrogen in air for the conditions of the scalar and Carter^{2,3}. Velocity measurements of Barlow measurements are also available for these conditions³⁸. CMC predictions have also been made¹²⁵ for the helium-diluted hydrogen flames of Barlow and Carter^{2,3} and also¹²¹ for the H_2 -CO flame of Drake³³. In general the predictions agree well with experiment. We shall present in some detail the methods and results of Smith *et al*¹²⁴, supplemented by some recent calculations⁶⁹, and then make brief summaries of the results obtained in other studies.

Smith *et al*¹²⁴ solve Eq. (5.13) for species using a first-order closure for the conditional reaction rate (Eq. 3.36) using both a "full" mechanism (employing 8 species and 21 reactions for H₂ oxidation and a further 4 species and 5 reactions for NO formation kinetics) and a two-step reduced mechanism that assumes partial equilibrium for the main radicals and steady state for N atom. An equation for the conditional average enthalpy, similar in form to Eq. (5.13) was solved using a first order closure for the radiation source term. The instantaneous source term was assumed to be given by the model of Kuznetsov and Sabelnikov⁷², see Eq. (3.33):

$$W_{\rm R} = - 4\sigma_{\rm o}\beta_{\rm H2O}p_{\rm H2O}(T^4 - T_{\rm b}^4)$$
(5.43)

with the emissivity function, β_{H2O} , being given by

$$\beta_{\rm H2O} = 2.0E-05 - 6.4E-09 T (Pa^{-1}m^{-1})$$
 (5.44)

where T is temperature in Kelvin, subscript b refers to the background value, $p_{\rm H2O}$ is the partial pressure of H₂O in atm., and σ_0 is the Stefan The kinetics and thermodynamic properties were handled Boltzman constant. using standard CHEMKIN II⁵⁰ subroutines and conditional average temperature was obtained from the conditional average enthalpy and composition using algorithms, by neglecting these that is, any contribution from conditional fluctuations. Use of this conditional obtain conditional average temperature to the average radiation source term by first-order closure as is done for the chemical source term in Eq.(3.36) involves neglect of the conditional fluctuations in the sight this problematical due temperature. At first seems to be to the fourth-power dependence evident in Eq.(5,43). This was found to be compensated in large measure by the temperature effects on the emissivity given in Eq.(5.44), as is explained below.

The CMC equations were solved in parallel with a computational fluid dynamics (CFD) solution for the Favre-averaged velocity and mean, ξ , and $(\xi'')^2$ the fraction. variance, of mixture А Reynolds stress turbulence model was used and a parabolic-like equation solver used to step the calculation in the x_1 direction. The CFD code passed information on the velocity and mixture fraction to the CMC subroutine where U^* and N^* were LSODE⁴³ (5.13) integrated using an solver evaluated and Eq. for stiff differential equations. This solver its ordinary has own step-size control and in general many sub steps are taken in the CMC routine for each step taken in the CFD calculation. The CMC subroutine returns the conditional average density, ρ_n , to the CFD code where it is used to calculate the unconditional average density, $\langle \rho \rangle$ at each point across the flow by weighting with the local pdf evaluated from ξ and $(\xi'')^2$ assuming a function form for the pdf. It is noted that since the pdf derived beta from the Favre-averaged moments of the mixture fraction is a Favre pdf, $\tilde{P}(\eta)$, the mean density must be obtained from

$$\langle \rho \rangle = \left(\int \rho_{\eta}^{-1} \tilde{P}(\eta) \ d\eta \right)^{-1}$$
 (5.45)

The CFD code used 50 radial node points and the CMC code 50 in mixture

fraction space, a much closer spacing being used around stoichiometric, η = η_s = 0.0285.

Boundary conditions for the flow were those of the experiment: a jet diameter of 3.75mm, average jet velocity of 300m/s with a profile and turbulence kinetic energy for fully developed pipe flow at the Reynolds number of 10,000, a coflow velocity of 1.0m/s with a turbulence level estimated at 5%. For the CMC calculation the composition is that of air (23% O₂, balance N₂ by mass fraction) at $\eta = 0$ and pure hydrogen at $\eta = 1$. Ignition of the flow is assured by assuming that the composition is that adiabatic equilibrium for the first 5 diameters: results for downstream were found to be insensitive to the axial location where this transition occurs. (Blowoff occurs if it is taken too far upstream.)

At preset intervals in x_1 the code writes out the dependent variables from both the CFD and CMC parts of the code together with unconditional averages (conventional and Favre) of the species and temperature at selected points across the flow. These are evaluated from

$$\widetilde{Y} = \int_{0}^{1} Q(\eta) \widetilde{P}(\eta) \ d\eta$$

$$\widetilde{Y} = \langle \rho \rangle \int_{0}^{1} \left\{ Q(\eta) / \rho_{\eta} \right\} \widetilde{P}(\eta) \ d\eta$$
(5.46)
(5.47)

equations with similar applying to temperature. (Conditional average is obtained from the conditional averages for temperature enthalpy and species via the CHEMKIN II subroutine: it is recognised that this is not exact due to the nonlinearity involved, but the errors estimated from the levels present in the conditional fluctuation experimental less data are than 1%.) The code also puts out profiles of the velocity and turbulence field and integral checks for momentum and flow rate of mixture fraction.

Of first interest in the results, always, is the quality of the prediction of the velocity and mixing fields. Since this flame is close to equilibrium and mean density is the only coupling that occurs from the CMC that can affect the CFD, one can expect that any deficiencies arise from inadequacies in the turbulence model. Comparisons for the flow field are not available. A "flame length" to stoichiometric (where ξ on the axis

of 163 diameters was predicted. The experimenters equals n.) quote a 180 visible flame length of diameters, but a flame length to data^{2,3} their stoichiometric of 128 diameters can be inferred from on averaged mixture Thus the turbulence model Favre fraction. underpredicts the rate of mixing. This should be kept in mind when the predictions for reactive species are being considered.

Comparisons for the reactive species are best made in terms of the conditional predictions versus measurements for the averages plotted fraction. against mixture This is because inadequacies in the CFD will have less effect on the conditional averages will predictions than in unconditional averages plotted against radius since these will appear be strongly affected by errors in the prediction of the mean and variance mixture fraction. In general the predictions are excellent for the of the major species and the OH radical. We will focus first on the most difficult species predict: nitric oxide, NO. Figure 19 to shows predictions for the conditional average of NO compared with the experimental data at $x_1/\ell_f = 0.5$ and 1.0, where ℓ_f is the inferred visible flame length in each case. Such comparisons are better than comparisons at the same values of x_1 , since relative position in the flame is important. be kept in mind, however, It should that the transit times are probably longer in the predictions due to the longer flame length, although this will be compensated to some extent by the velocities being higher. It is seen that CMC gives good predictions in fuel rich mixtures in the middle of the flame but overpredicts NO for lean and stoichiometric mixtures here and at the end of the flame. Figure 20 shows predictions closer to the nozzle. It is seen that NO is grossly underpredicted at $\ell_{\rm f}/8$ but the tendency to overpredict has overtaken this trend by $\ell_f/4$.

The CMC predictions of conditionally averaged temperature compared with the laser measurements are very good near the end of the flame (not shown here) being within about 30 K. Nearer the nozzle the temperatures significantly underpredicted as can be seen in Fig. 21 Temperature are predictions near the end of the flame are sensitive to radiant losses but or the model value for scalar to the kinetics dissipation: the fluid not equilibrium. nozzle is close Near the temperature predictions there to insensitive radiation losses but are quite sensitive to the kinetic are to the three-body recombination Differential rate constants for reactions. also probably significant. diffusion effects are One might also expect results would be sensitive near the nozzle the to the conditional that

average scalar dissipation, particularly as η_s is much lower than the mean ξ in the middle of the shear layer and hence the shape of N^* will be particularly important. These effects are considered in more detail below.

The underprediction of temperature near the nozzle could explain the underprediction of NO in this region. The activation temperature for the controlling step in the formation mechanism is 38,100 K. This means that both errors in the mean and temperature fluctuations (Eq. 3.38) will have significant effects on the mean rate of formation of NO. At $x_1 = \ell_f/8$ the peak conditional mean temperature is underpredicted by about 160 K in 2200 K (see Fig. 21): this will give a reaction rate which is low by a factor of 4. Closer to the nozzle the error could be even larger. Figure 7 of Barlow and Carter³ indicates that the corresponding conditional rms of the fluctuations is about 100 K. temperature and calibration measurements indicate that little of this is shot noise associated with the low number photo-electrons gathered high of Rayleigh at temperatures. Equation (3.38)indicates that neglect of temperature fluctuations of this size causes an underestimation of the reaction rate by a further factor of 1.3. The correlation between T and O-atom fluctuations can be expected to decrease this somewhat. In any event, it appears that the NO production rate will be underpredicted by an amount more than sufficient to explain the low predictions for NO concentration at $x_1 = \ell_f/8$.

Figure 22 shows the effect of radiation loss and main reaction kinetics on the NO predictions. It is seen that an increase in the radiation loss by about 25% would be needed to bring the NO down to values without allowing for the residence time effect. measured Fluctuations in temperature about the conditional mean could alter the loss from the first order closure used radiation for Eqs (5.43), (5.44).We have

$$< W_{\rm R} |\eta> \approx W_{\rm R}(Q_{\rm T}, Q_{\rm H2O}) \left(1 + a_1 \frac{< T''^2 |\eta>}{Q_{\rm T}^2} + a_2 \frac{< T''Y''_{\rm H2O} |\eta>}{Q_{\rm T}Q_{\rm H2O}} \right)$$
(5.48)
ere

where

$$a_1 \equiv 2(3 - 5\alpha Q_{\rm T})/(1 - \alpha Q_{\rm T})$$

$$a_2 \equiv (4 - 5\alpha Q_{\rm T})/(1 - \alpha Q_{\rm T})$$

and

$$\alpha \equiv 3.2E-04$$

For the measured temperature fluctuations these corrections are less than one per cent, assuming that the rms/mean for the H_2O fluctuations are of the same order as the values measured for temperature.

of the The effects kinetic mechanism shown in Fig. 22 are The two-step reduced mechanism uses the same rates for significant. the recombination reactions but assumes that H-atom is given by the partial equilibrium relation. This results in higher recombination rates being higher temperatures and lower O-atom concentrations. predicted with The for al^{124} NO. Smith et also net result is higher predictions show comparisons with a Monte-Carlo PDF calculation that used a two-step kinetics model and adiabatic conditions. The predictions made for NO were higher than for those from CMC but not by as much indicated in Fig. 22.

 $et al^{125}$ have studied this undiluted Smith H_2 flame and the helium-diluted flames of Barlow and Carter^{2,3}. The CFD code was tuned to correct stoichiometric flame length. Much improved predictions give the for NO and temperature are found for all three flames. The major cause of improvement was inclusion of the water vapour present in the air stream, a factor neglected in the earlier study.

Kronenburg *et al*⁶⁹ have investigated the effect of the modelling of conditional scalar dissipation on the results for the undiluted the flame. As expected the results in the near field are quite sensitive to the model used: NO predictions at $x_1 = \ell_f/8$ are nearly doubled when N is computed directly from the pdf transport equation rather than using a simpler model based on Girmaji's⁴¹ method for homogeneous turbulence. In the near field there is a diffusive-reactive balance with the first term on the LHS of Eq. (5.13) being much smaller. Downstream, where most of the NO is produced, the convective and reactive terms are in balance and the results the end of the flame are insensitive the model used for scalar to Kronenburg dissipation and even to artificial inreases in its magnitude. et al^{69} also increasing found that the rate constants for the recombination reactions raised the predicted temperatures as expected. NO predictions in the near field were unchanged, however, but were lowered the end of the flame. It appears toward that the effects of reducing the effects of increased temperature in O-atom levels dominate over the far field but balance out in the near field. The standard rate constants the recombination reactions give the best predictions of OH levels for throughout the flame.

In summary, it appears that CMC predictions in the near field will be

improved by improving the modelling of scalar dissipation. by order closure for the effects of fluctuations incorporating a second on the temperature, O-atom concentration and the NO formation rate, and by effects of differential diffusion. Predictions modelling the in the far satisfactory and are not likely to be sensitive field are now these to refinements needed for the near field.

Smith¹²¹ presents results of calculations for the H₂-CO turbulent jet flame data of Drake³³. In general good agreement is diffusion obtained evidence of differential diffusion although there is effects not Roomina¹¹⁶ modeling. also incorporated in the finds differential diffusion effects to be significant in CMC predictions of jet flames of H₂-CO₂ mixtures and also methanol^{116,117}. Such effects are not found to be of great significance ¹¹⁶ in computation of the partially premixed methane air diffusion flame of Barlow and Frank⁴. Good predictions are found for major species and temperature using the full GRI mechanism⁴⁰ involving all 49 species and 277 reactions. Predictions for OH are also very good. For NO the predictions are somewhat high on the lean side but are very good on the rich side where strong effects of the reburn kinetics for NO are evident. These methane calculations took about 130 hours on a DEC 3000/400 calculation work station. Shorter times should be possible when improvements have been made to the numerical methods used for solving the stiff equations involved.

5.8.3. Round plumes in grid turbulence

Figure 23 shows schematically the set up for the experiments of Brown Bilger²³ for a reacting axisymmetric plume in grid turbulence. The and mean velocity of the flow is uniform and constant, while the turbulence A point source of nitric oxide diluted to 515 ppm is decays downstream. three grid mesh lengths downstream of the turbulence grid and loated enters the flow with the same mean velocity. The main flow contains about 1 ppm of ozone. Here $\Gamma_i \equiv Y_i/M_i$. The streams mix and react as they flow downstream. Measurements were made at $x_1/M = 9$ to 17 where the plume is describable by turbulent diffusion. The plume width there spread is less than $0.3x_1$ and so may be treated as a slender layer with the analysis of Section 5.3 being applicable.

Conditional average statistics for this plume and CMC modelling for

the conditional mean values are presented in Brown and Bilger^{24,25}. It is found that there is a small variation of the conditional averages across the flow with a range of about \pm 5% of the conditional mean of the NO concentration. In the CMC modelling conditional the average scalar estimated experimental dissipation was from the measurements of the conserved scalar variance using the strategy of Section 5.2 assuming a value of the time scale ratio (for decay of turbulence kinetic energy to that for scalar variance) to be independent of radius. Values for $\eta > \langle \xi \rangle$ on the centreline were computed from the centreline variance. Results for a moderate Damkohler number case are shown in Fig. 24 Here $Q_i \equiv \langle Y_i | \eta \rangle$. It is seen that agreement is excellent. The reaction dominated limit shown is obtained by setting $\langle N | \eta \rangle = 0$. It was found that the predictions were relatively insensitive to the values taken for $\langle N | \eta \rangle$ in the CMC equation: changes by a factor of 2 only change the predictions by about 10%. This the mixing term in Eq. (5.3) is less significant means that than the convection term, particularly far downstream. In this flow the scalar dissipation decreases with distance, x_1 , raised to the -3.6 power, almost as strongly as it does in a jet.

6. CMC FOR SIMPLIFIED TURBULENT REACTORS

Here we consider analogues of the Perfectly Stirred Reactor (PSR) and the Plug Flow Reactor (PFR) for conditions under which the flow within the reactor is incompletely mixed in terms of the mixture fraction. The first is the Incompletely Stirred Reactor (ISR), an analogue of of these the PSR, in which it is assumed that the conditional averages of the reactive are uniform within the reactor even scalars though the mixture fraction field is not. Its limiting case is a PSR with incompletely mixed inlets, which is still in fact a PSR. The ISR may prove to be a useful model of reactors as the turbine primary zone in which such gas strong recirculation is present and the Damkohler numbers are moderate.

The second is the Dilution Flow Reactor (DFR) which is an analogue of the Plug Flow Reactor (PFR): the reactive scalars are assumed to be uniform across the flow, as found to leading order for turbulent slender layer flows. The DFR may prove to be a useful model for the dilution zone of a gas turbine combustor and for NOx reburn zones of furnaces.

In both cases it is emphasised that the conditions under which such modelling will be valid are not yet clearly defined. It can be expected, however, that these models will be more informative for such flows than PSR and PFR models at a modest increase in computational cost. They may prove to be useful for parametric studies.

6.1 The Incompletely Stirred Reactor

In this section we examine the application of CMC methods to a class of flows that define what we shall term an Incompletely Stirred Reactor (ISR). The ISR is a generalisation of the well-known Perfectly Stirred Reactor (PSR). In the PSR low temperature reactants enter the reactor and immediately mix with the reacting mixture within the reactor volume. The composition and temperature within the PSR are uniform and are the same as in the outlet flow. The PSR concept has been a useful one and has been used as a preliminary design tool for combustors in which the conditions uniformity fact, far from being realised. Generalisations of are, in of the PSR which allow for incompleteness of the mixing within the reactor additions should prove to be useful to the armory of the combustor designer. The Partially Stirred Reactor (PaSR) concept has been developed

from the Monte-Carlo pdf method and has been applied to premixed^{29,30} and nonpremixed²⁸ systems. Plausible predictions of NO and CO emissions and extinction and ignition phenomena are obtained that show a dependence on insights beyond what is mixing rates that give obtained from PSR There is no direct link, however, between calculations. the mixing rates assumed and the flow field that exists in the combustor. The ISR concept provides such a link for nonpremixed systems and may prove to be useful for these reasons.

described Section As has been in 5, great simplifications are flows where it is found possible in slender layer that there is little variation across the flow of conditional averages of the reactive scalars, strong variations in even though there are very the mixing field as cross-stream profiles of $\langle \xi \rangle$, $\langle \xi'^2 \rangle$ represented by the and *P*(n). This idea generalised to strongly recirculating flows: if is here the recirculation strong enough flow is and other characteristics are appropriate, the conditional averages of the reactive scalars could be essentially uniform within the reactor even though the mixing field represented by $\langle \xi \rangle$, $\langle \xi'^2 \rangle$ and $P(\eta)$ is far from uniform.

Figure 25 shows a schematic of an axisymmetric combustor which may be candidate for ISR analysis of the recirculation a suitable zone. The figure defines the cylindrical coordinate system (x,r). A central primary of partially premixed fuel and air enters without swirl iet along the annulus of secondary air enters axially combustor axis. A thin at high velocity without swirl at a large distance from the centreline but not the wall of the combustor. Figures 26 - 28 show quite at the mean streamlines computed for the flow together with the Favre mean, $f \equiv$ $<\rho\xi''^{2}>/<\rho>.$ and Favre variance of the mixture fraction, $g \equiv$ $<\rho\xi>/<\rho>$, where $\xi'' \equiv \xi - f$. For the partial premixture chosen for the primary jet fraction stream, the stoichiometric value of the mixture is η、 = 0.3. These computations have been made using a standard k-epsilon model for the turbulence and using a fast chemistry assumption for the combustion⁹⁴. The mean flow pattern shows three toroidal vortices, with the large downstream the stoichiometric of most interest to It is seen that one being us. goes right through the middle of contour of mean mixture fraction this recirculation zone. Since most of the reactions of interest to us occur stoichiometric, it is evident that there is no obvious near reason for spatial variations in conditional there to be strong averages of temperature and reactive species mass fractions for values of n near

stoichiometric. It is also seen that the incoming primary fuel-air stream with combustion products, rather than with cold secondary mixes air and hence should not give rise to local premixing without ignition or to local extinction Furthermore, the secondary events. air stream mixes with recirculated combustion products rather than with cold fuel so that there should be no unignited pockets of mixture as occur where a ported flame is lifted and not stabilised right at the burner lip. Conditional variances the conditional mean for species and temperature, at about point in any the flow may not be so large as to make a first moment closure for the conditional reaction rates greatly inaccurate. It is obvious that the strong variations of mixture fraction, spatial and temporal, make this combustor far from being a PSR. It is also evident that reaction will not be confined to a thin flame front and laminar flamelet modelling for the valid^{9,22}. will not be This is combustion because the width of the reaction zone in mixture fraction with space is comparable the rms fluctuations in the mixture fraction.

ISR modelling is not generally applicable to all recirculating flows and indeed may only be valid for a narrow range of tightly constrained combustor designs. Even so, this class of combustors may have practical significance. Like the simply supported beam, ISRs may find favour with designers for the simple reason that they are easily analysable and that combustor geometry and flow parameters are directly linked to performance through CFD and CMC analysis.

ISR Various versions of the concept have been presented in Ref. ^{94,95,96,121,122}. Here we present the formal definition of ISR an and the derivation of its governing equations. We apply the method to the Figs. 25-28 combustor illustrated in and discuss the validity of this effects of application and design operating the and parameters on combustor performance.

6.1.1. Definition and governing equations

an incompletely stirred reactor (ISR) to be a region of We define bounded within simple boundaries, within which the conditional flow. of temperature and species mass fractions show little spatial averages and the conditional variances about conditional variation these averages **Ouantification** small. of "little" and "small" in this definition are is

such as is necessary to allow a first or second order closure for the conditional average reaction rate terms in the CMC equations. By "*simple boundaries*" we mean inlet boundaries across which fluxes of species and enthalpy are given, walls which are non-reactive and impervious to flow and across which heat transfer is readily evaluated, and outlet boundaries across which fluxes of enthalpy and mixture fraction are known, or available from CFD calculations.

For clarity of presentation we restrict ourselves to statistically stationary flow, although unsteady ISRs are also possible. We begin with the CMC equation of Eq. (4.1) in the the statistically stationary form

$$\operatorname{div}\left(\overline{\rho} < \mathbf{v}Y | \eta > \widetilde{P}(\eta)\right) = \overline{\rho} < W | \eta > \widetilde{P}(\eta)$$
$$- \frac{\partial^2 \overline{\rho} < N | \eta > \widetilde{P}(\eta)}{\partial \eta^2} Q + \overline{\rho}_{\eta} < N | \eta > \widetilde{P}(\eta) \frac{\partial^2 Q}{\partial \eta^2}$$
(6.1)

where the last term on the RHS of Eq. (4.1) has been differentiated through and the conversion to Favre pdfs of Eq. (4.4) has been made. Integrating this over the ISR or "core" volume, *V*, and using the flux divergence theorem leads to

$$\int_{A_{out}} \overline{\rho} < \mathbf{v} Y |\eta > \widetilde{P}(\eta) \cdot d\mathbf{A} - \int_{A_{in}} \overline{\rho} < \mathbf{v} Y |\eta > \widetilde{P}(\eta) \cdot d\mathbf{A} = \int_{V} \overline{\rho} < W |\eta > \widetilde{P}(\eta) \ dV$$
$$- \int_{V} \frac{\partial^{2} \overline{\rho} < N |\eta > \widetilde{P}(\eta)}{\partial \eta^{2}} Q \ dV + \int_{V} \overline{\rho} < N |\eta > \widetilde{P}(\eta) \frac{\partial^{2} Q}{\partial \eta^{2}} \ dV$$
(6.2)

In this equation conditional correlations between the species and velocity have been ignored, it being assumed that they are small compared with the mean convective flux. Since, according to the definition of the ISR. all conditional reactive scalar statistics are uniform inside the core, they can be moved outside the integrals. We use the definitions:

$$P_{\rm b}^* \equiv \frac{1}{\dot{m}} \int_{A_{\rm b}} \overline{\rho} < \mathbf{v} | \eta > \widetilde{P}(\eta) \cdot d\mathbf{A}$$
(6.3)

$$P^{**} \equiv \frac{1}{\rho^{**}V} \int_{V} \overline{\rho} \widetilde{P}(\eta) \, dV \tag{6.4}$$

$$\rho^{**} \equiv \frac{1}{V} \int_{V} \overline{\rho} \, dV \tag{6.5}$$

$$N^{**} \equiv \frac{1}{\rho^{**} P^{**} V} \int_{V} \overline{\rho} \widetilde{P}(\eta) \langle N | \eta \rangle dV$$
(6.6)

where \dot{m} is the mass flow rate through the reactor and subscript "b" refers to inlet or outlet. Eq. (6.2) can then be written

$$Q P_{\text{out}}^{*} - Q_{\text{in}} P_{\text{in}}^{*} = = \tau_{\text{r}} \left(P^{**} < W | \eta > + P^{**} N^{**} \frac{\partial^{2} Q}{\partial \eta^{2}} - Q \frac{\partial^{2} P^{**} N^{**}}{\partial \eta^{2}} \right)$$
(6.7)

where the residence time $\tau_{\rm r}$ is given by

$$\tau_{\rm r} = \rho^{**} V / \dot{m} \tag{6.8}$$

According to the previous discussion we assume in Eq.(6.7) that 1) Q does not have significant variations within the volume V so that Qcan be placed outside the integration sign and 2) the outlet value of the conditional expectation Q coincides with the value Q within the volume V: $Q_{\text{out}}=Q$. Integrating the Favre form of the pdf transport equation (4.3) and using the above definitions yields

$$P_{\text{out}}^* - P_{\text{in}}^* = -\tau_r \frac{\partial^2 P^{**} N^{**}}{\partial \eta^2}$$
 (6.9)

Subtracting Q times this from Eq.(6.7) yields

$$\left(Q - Q_{\rm in}\right)P_{\rm in}^* = \tau_{\rm r}\left(P^{**} \langle W|\eta \rangle + P^{**}N^{**}\frac{\partial^2 Q}{\partial \eta^2}\right)$$
(6.10)

The volume weighted conditional scalar dissipation, N^{**} may be found by double integration of Eq. (6.9) with respect to η

$$N^{**} = - \frac{l}{\tau_{\rm r} P^{**}} \int_{0}^{\eta} \int_{0}^{\eta'} \left(P^{*}_{\rm out}(\eta'') - P^{*}_{\rm in}(\eta'') \right) d\eta'' d\eta'$$
(6.11)

Use has been made of the boundary conditions of Eqs (2.61),(2.62) and (2.68)-(2.71), with the stationary flow assumption.

Equations (6.10) and (6.11) are the ISR equations for generalised inlet conditions where P_{in}^* has a broad distribution indicative of partial premixing. In this case Q_{in} is defined

$$Q_{\rm in} = \frac{I}{P_{\rm in}^*} \int_{A_{\rm in}} \overline{\rho} \widetilde{P}(\eta) < \mathbf{v} Y | \eta > d\mathbf{A}$$
(6.12)

With N^{**} obtained from Eq.(6.11), Eq. (6.10) is integrated with boundary conditions at $\eta = 0, 1$ given by the inlet values, Q_{in} , at these bounds.

Of greater interest is the case when there are two inlet streams, each having uniform composition and enthalpy, so that P_{in}^* is comprised of two delta functions, one at $\eta = 0$ and the other at $\eta = 1$. In this case the ISR equations reduce to

$$N^{**} \frac{\partial^2 Q}{\partial \eta^2} + \langle W | \eta \rangle = 0$$
(6.13)

$$N^{**} = \frac{1}{\tau_{\rm r} P^{**}} \left[(1 - \xi_{\rm a})\eta - \int_{0}^{\eta} (\eta - \eta') P^{*}_{\rm out}(\eta') d\eta' \right]$$
(6.14)

In Eq.(6.14) use has been made of integration by parts in reducing the double integral to a single integral. Also ξ_a is the fully mixed value of the mixture fraction for the inlet and outlet flows

$$\xi_{a} = \int_{0}^{I} \eta P_{in}^{*} d\eta = \int_{0}^{I} \eta P_{out}^{*} d\eta$$
(6.15)

For flows in which there is no conduction or convective heat transfer at the walls an equation similar to Eq. (6.13) will apply for the conditional average of the standardized enthalpy. The source term will now be the conditionally averaged heat transfer gain by radiation. Extra terms will be needed for inclusion of heat transfer by convection/conduction at the walls.

The boundary conditions to be used in solving Eq. (6.13) are

$$Q = \left(Q_{in}\right)_{\eta=0} \quad \text{for } \eta = 0$$

$$Q = \left(Q_{in}\right)_{\eta=1} \quad \text{for } \eta = 1 \quad (6.16)$$

The inlet streams may be partially premixed (uniformly) mixtures of fuel and air. They may even be reacting as long as the Q_{in} values are taken at the boundary assumed in evaluating the integrals of Eqs (6.3)-(6.6).

The ISR becomes a PSR in the limit of the mixing in the core becoming so strong that both P^{**} and P_{out}^{*} become delta functions centred at ξ_a . This can be shown by substituting Eq. (6.11) in Eq. (6.10) and integrating over mixture fraction space. Integrating the second derivative of Q by parts yields

$$Y - \int_{0}^{I} Q_{\rm in} P_{\rm in}^* d\eta = W \tau_{\rm r}$$
(6.17)

where Y is the species mass fraction that is uniform throughout the reactor and equal to its outlet value; W is its rate of formation by chemical reaction. If the inlet stream(s) are fully premixed we obtain the usual PSR formula:

$$Y - Y_{in} = W\tau_r$$

It is evident hat a PSR does not necessarily need to have its inlet flows fully premixed. If the mixing within the reactor is strong enough the essential features of the PSR are present and the slightly modified formula of Eq. (6.17) applies.

6.1.2. Application

Mobini⁹⁴ has modelled the head end of the combustor of Fig. 25 as an

ISR, taking the outlet area to be the plane normal to the axis as defined From Fig. 26 this is seen as being at the end of by $x = x_{\text{max}} = 0.053m$. This seems appropriate the recirculation zone. an place, although turbulent flux contribution to the outlet integral should strictly the be included for points near the axis where the axial velocity is small. The flow and mixing calculations shown in Figs 26–28 were made with a primary fuel-air flow rate of 0.76 g/s at an equivalence ratio of 4 with methane as the fuel and a secondary air flow rate of 4.3 g/s. Inlet flows were at 300K and the combustor pressure is 1.0 atm.

Figures 29 and 30 show the core, P^{**} , and outlet, P^{*}_{out} , pdfs and the core-averaged conditional scalar dissipation, N^{**} , computed from the CFD output assuming that the mixture fraction Favre pdf at any point in the pdf, P^{**} , is flow has a beta-function form. It is seen that the core quadrimodal with spikes at $\eta = 0, 1$ associated with the potential cores of the inlet jets, a peak at $\eta \simeq 0.04$ associated with the recirculation zone at the outer corner, and a main peak at $\eta \simeq 0.11$ associated with the outer flow near the combustor wall which has a high radius and relatively high These latter two peaks are responsible for the bumps in the N^{**} density. The N^{**} significantly of η . profile has a profile at these values found for laminar counterflow flames, different shape to that with the steep gradient near $\eta = \eta_s = 0.3$ being of possible advantage, as discussed later.

identical The ISR equation (6.13)is to the equation solved for and is solved laminar counterflow flames using a standard two point equation systems 51 . stiff Note boundary value problem solver suitable for that the ISR determined values of N^{**} Mobini⁹⁴ are used, however. has the full Miller-Bowman obtained solutions using mechanism, including all prompt kinetics for the NO. Solutions took approximately 2hrs on a 1995 vintage advanced work station (DEC Alpha 3000-700). Figure 31 shows the obtained for conditional average values of the prediction mass fractions of the major species. while Fig. 32 shows predictions for the conditionally averaged temperature and mass fraction of nitric oxide. The residence time obtained from the volume average of the unconditional mean The major species and temperature are similar to those density was 8.6ms. that would be obtained for a laminar flamelet calculation with the same value of conditional scalar dissipation at stoichiometric, except that the peak CO is somewhat higher. The peak conditional average temperature at 2000K is well above that at which local extinctions would be expected to

occur. The conditional average nitric oxide mass fractions are somewhat higher than would be desired for a rich-burn quick-quench combustor and yield an average value at exit of 41ppm. Such average values at exit can easily obtained by weighting by P_{out}^*

$$\left(\langle Y_{i}\rangle\right)_{out} \equiv \frac{\dot{m}_{i,out}}{\dot{m}} = \int_{0}^{l} Q_{i}(\eta)P_{out}^{*}d\eta \qquad (6.18)$$

flow rate of species *i* at ISR is the mass the exit. where $\dot{m}_{\rm i,out}$ Figure 29 shows that P_{out}^* is quite narrow, so that this quadrature is easy to estimate by eye. It is apparent that the NO levels could be decreased by increasing the mass flow rates of the primary and secondary streams, bringing the ISR residence time down and decreasing the peak temperature. The pressure loss in the secondary air stream would become severe. however, and the CO emissions from the ISR would increase. The latter would perhaps not be too much of a problem as the CO will continue to burn out in the flow downstream of the ISR while little further NO will be The flow downstream of the ISR essentially meets the criteria produced. for slender layer flows presented in Section 5 and the methods put forward there could be used to predict further burn out of the CO using the Dilution Flow Reactor model presented in section 6.2. Parametric studies of the effect of residence time and other variables have been carried out ⁹⁴, but for a different system.

Unconditional averages of temperature and species mass fraction can be estimated at every point within the combustor by weighting the local pdf, $P(\eta; x, r)$, conditional averages by the obtained from predicted the local values of the mean and variance of the mixture fraction and the presumed form of the pdf. Favre averages or conventional averages can be obtained. Usually it is the Favre pdf $\tilde{P}(\eta;x,r)$ that is available and so the formulae are then

$$\widetilde{Y}_{i} = \int_{0}^{I} Q_{i}(\eta) \widetilde{P}(\eta; x, r) d\eta$$

$$\overline{Y}_{i} = \overline{\rho}(x, r) \int_{0}^{I} \rho_{\eta}^{-1} Q_{i}(\eta) \widetilde{P}(\eta; x, r) d\eta$$
(6.20)

similar formulae applying for temperature. Mobini⁹⁴ gives contour with species plots for several and temperature obtained in this way. studies can be made for ISRs with differing characteristics Parametric Since N^{**} is the only thing that without resorting to CFD calculations. chemistry for given inlet composition (see Eqs 6.13, 6.16)), affects the the effects of different mixing patterns and residence times can be explored by parametric variation of the shape of the core, P^{**} , and outlet, P_{out}^* , pdfs and of the residence time, τ_r . Consequent effects on core average conditional scalar dissipation, N^{**} , can be calculated from Eq. (6.14), and solutions for Q obtained from solution of Eq. (6.13) Emissions from the ISR can be evaluated from Eq. (6.18). As a result of such parametric studies it should be possible to find the desirable shapes of P^{**} and P^{*}_{out} and acceptable value of τ_r to meet emission goals. CFD design studies can then be carried out in attempts to meet these desired pdf shapes and residence time. Smith and Bilger¹²² and Mobini et al^{95} have carried out such parametric studies. General findings are that the outlet pdf, P_{out}^* , has little influence on the results provided it is not too broad in mixture fraction space¹²². This is because the numerator for the RHS of Eq. (6.14) has an almost triangular shape with zero values at $\eta =$ 0,1 and an apex of $\xi_a(1-\xi_a)$ at $\eta = \xi_a$: broadening P_{out}^* merely rounds off the corner of the apex. Narrowing P^{**} causes N^{**} to shoot up to high values at η values away from that where P^{**} has its peak: this is how the ISR approaches the PSR limit.

6.1.3. Validity of ISR modelling

Criteria for determining the validity of ISR modelling are as yet not clearly defined. For simple chemistry with an identifiable chemical time scale, τ_c , a suitable criterion may be

$$\tau_{\rm rc} \ll \tau_{\rm c} \ll \tau_{\rm r} \tag{6.21}$$

where τ_{rc} is a suitably defined recirculation time scale. In strongly recirculating flows τ_{rc} will be a measure of the time for a fluid particle to make one orbit of the recirculation zone, many such orbits being made, on average, before the fluid particle exits the reaction zone. If $\tau_{rc} \ll \tau_c$ then it can be expected that there will be little spatial variation of

the conditional averages, Q, within the reactor volume. The requirement that $\tau_c \ll \tau_r$ seems appropriate to ensure that there is no likelihood of extinction events within the reactor volume. For complex chemistry the situation is more complicated and further analysis is needed. A full CMC analysis of the reactor shown in Figs 25-28 is possible and should throw much light on the requirements for validity of ISR modeling.

Gough et al⁴² have made measurements in a combustor very similar to Flow conditions were slightly different than those that shown in Fig. 25 discussed in Section 6.1.2 due to the fact that for those conditions the CFD predictions are inaccurate and the fuel jet penetrates the The outer annulus air velocity was increased recirculation zone. SO that the fuel jet was no longer penetrating. Measurements made with sampling a thermocouple at the exit of the recirculation probes and zone are compared with revised ISR model predictions in Fig. 33. It is seen that the predictions show quite good agreement with experiment although NO is overpredicted somewhat and CO is underpredicted. These results give some encouragement that the ISR model can at least be used for giving the right in parametric studies. Work is in progress to verify this. The trends may also be helpful in getting starting solutions in fully-elliptic model multi-dimensional calculations.

6.2. The Dilution Flow Reactor

The Dilution Flow Reactor (DFR) is a generalisation of the well-known Plug Flow Reactor (PFR)¹³⁹. In the PFR, the flow is assumed well mixed in a cross-stream direction so that profiles of velocity and composition are Composition gradients in the streamwise direction are assumed to uniform. SO that longitudinal molecular and turbulent transport be small can be neglected. The problem is steady and one-dimensional with chemical by longitudinal convection. reaction being balanced Dilution by mass readily added addition can be to the problem but is usually not considered.

In the DFR, dilution of the flow is specifically catered for, as is nonuniformity of the mixture fraction across the flow. The assumption is made that the conditional averages of reactive scalars do not vary across

flow and that fluctuations about these conditional averages are small the enough for closure of the source terms to be made at the first conditional moment level. Figure 34 shows a schematic of such a reactor. The duct may be of varying cross-sectional area, but for simplicity it is assumed that its centreline is straight. In the analysis that follows, the flow through the walls of the duct is assumed to be inward and either of mixture fraction zero or unity. In gas turbine combustors the diluent is air, while in furnaces with NOx reburn the inflow is first fuel (not really a In NOx reburn diluent!) and later air. systems the fuel injection generates NH₂ and other radicals that can reduce the NOx generated in the upstream regions. The chemistry is complex and very sensitive to stoichiometry so that incorporating the mixing effects is very important. It is possible to derive the model for inflows at intermediate mixture fractions, but for simplicity we will assume that the inflow is at $\xi = 0$ or 1.

Integrating Eq.(4.1) and (4.3) across the flow with the assumption that $Q = Q(\eta, x)$ with $x = x_1$ (the streamwise direction) yields

$$\frac{\partial \rho_{\eta} P^{*} U^{*} Q}{\partial x} - \dot{m}'_{fu}(x) \delta(1-\eta) Q - \dot{m}'_{ox}(x) \delta(\eta) Q =$$

$$< W |\eta > \rho_{\eta} P^{*} + \frac{\partial}{\partial \eta} \left[\rho_{\eta} P^{*} N^{*} \frac{\partial Q}{\partial \eta} - \frac{\partial \rho_{\eta} P^{*} N^{*} Q}{\partial \eta} \right] \quad (6.21)$$

$$\frac{\partial \rho_{\eta} P^{*} U^{*}}{\partial x} - \dot{m}'_{fu}(x) \delta(1-\eta) - \dot{m}'_{ox}(x) \delta(\eta) = - \frac{\partial^{2} \rho_{\eta} P^{*} N^{*}}{\partial \eta^{2}} \quad (6.22)$$

where $\dot{m'}_{fu}(x)$ and $\dot{m'}_{ox}(x)$ are the mass inflow rates per unit length in the x direction of fuel and oxidant respectively. P^* , U^* and N^* are averages computed across the flow as in Eq.(5.10). Subtracting Q times Eq.(6.22) from Eq.(6.21) and dividing by $\rho_{\eta}P^*$ yields

$$U^* \frac{\partial Q}{\partial x} = \langle W | \eta \rangle + N^* \frac{\partial^2 Q}{\partial \eta^2}$$
(6.23)

which is essentially the same as Eq.(5.13). Integrating Eq.(6.22) by parts yields

$$\rho_{\eta}P^{*}N^{*} = \frac{\partial}{\partial x} \int_{\eta}^{1} (\eta^{\circ} - \eta)\rho_{\eta}P^{*}U^{*}d\eta^{\circ} + (1 - \eta)\dot{m}'_{fu}(x)$$
(6.24)

If a full CFD prediction for the flow and mixing field is available, U^* N^* and can be obtained from Eqs(5.10)and (6.24), respectively. Equation.(4.5) should probably be used for obtaining the conditional direction velocity in the axial since there will be regions of low velocity the wakes of the jet inflows. In parametric studies in more approximate estimates for U^* and N^* will probably suffice.

In the above it has been assumed that the walls are nonreactive. There will corresponding equation for conditional be a the average enthalpy with a radiation source term. will standardized Additional terms the effects of conductive/convective heat transfer appear for at the wall and for variations in temperature with x of the "diluents".

7. CMC IN HOMOGENEOUS FLOWS

As noted in Section 4.5 the CMC equations are considerably simplified in cases where the flow and mixing can be taken as being homogeneous. Such flows are approximated experimentally by the flow behind а turbulence generating grid. Scalar mixing in such flows has been studied extensively for temperature in air by Warhaft and others (see Tong and Warhaft¹³⁶, for literature). Reacting experiments have been entry into this carried out in a tubular reactor by Toor and others and these have been modelled by Southwell¹²⁷. This work is briefly reviewed in Section 7.1. Homogeneous turbulent flows are particularly amenable to direct numerical simulation applicable (DNS), the pseudo-spectral method being where density and review results transport properties are constant. We obtained for simple kinetics in Section 7.2 and for multi-step one-step chemistry in Section 7.3.

7.1. Turbulent Mixing Reactors

Southwell¹²⁷ has modelled the experimental data of Vassilatos and Toor¹⁴⁰ and Li Toor⁸⁰ using CMC. and In the Vassilatos and Toor reactor consisted of hundred experiments the one parallel jets issuing hypodermic tubing closely set together. The two reactants from issue alternately from jets. The turbulence generated the is approximately homogeneous in the cross-stream direction and the rate of decay is such that it is essentially homogeneous in the flow direction. Dilute sodium hydroxide was one reactant and the speed of the reaction was varied by dissolved carbon dioxide (quite fast) acetic acid (very slow), using and hydrochloric acid (very fast) as the other reactant. Mixing data for this reactor is available from Shenoy and Toor¹¹⁸ and the variance of the mixture fraction fluctuations was fit by piecewise power law functions of used the distance downstream. These functions were to deduce the dissipation. unconditional scalar In the CMC modelling the conditional average dissipation was assumed to be equal to unconditional scalar the value. The predictions showed agreement with experimental good the unconditional average of fractional conversion results. the to product underpredicted in slightly overpredicted being slightly the slow case, and in the quite fast case. The very fast case indistinguishable from was

full equilibrium. More accurate modelling of the conditional scalar dissipation may improve predictions for the quite fast case.

In the experiments of Li and Toor⁸⁰ the reactor was similar to the above, but only had 14 jets. Mixing data for this reactor were obtained from Li⁷⁹ and Singh¹²⁰ and the mixture fraction variance was found to be quite strongly dependent on Reynolds number: once again piecewise power dissipation law fits were made to this data and the unconditional of was obtained by differentiation. mixture fraction the CMC In modelling conditional dissipation the scalar was once again taken the as unconditional value. The reactants were 1-naphthol (A)and diazotised sulfanilic acid (*B*) to produce two dyestuffs R and S through the series-parallel reactions

$$A + B \longrightarrow R \tag{7.1}$$

$$R + B \longrightarrow S \tag{7.2}$$

The first of these reactions is quite fast and the second reaction is much slower. Measurements were made only at the reactor exit. Figure 35 shows results obtained with the CMC modelling using the average value of the rate constants that have been published for the first reaction. Also shown predictions with the mechanistic Four-Environment Slow model are the of Tarbel1³⁴. the and most accurate of the unconditional Dutta moment closures methods used by chemical engineers. It is seen that the CMC predictions are good. They probably can be improved with improvements to the model for the conditional average scalar dissipation.

7.2. DNS with One-Step Chemistry

Direct numerical simulations with nonpremixed reactants with a single $al^{90,91}$ irreversible chemical step have been studied by Mell et Swaminathan and Mahalingam¹³³, and Mel1⁸⁹. Lee Pope⁷⁵ and consider a reversible one-step reaction with corresponding features high to a activation energy in the reaction Their main is in rate. interest predictions of extinction, and here CMC does poorly as can be expected closure is considered the conditional since only a first moment for In general all the other comparisons with DNS show average reaction rate.

CMC to make very good predictions for unconditional averages of reactant concentrations provided that the conditional average of the scalar dissipation for the mixture fraction is modelled adequately.

In principle, DNS allow us to examine in every detail the closure assumptions made in CMC modelling. The limitation is that the simulations are only available for fairly low turbulence Reynolds numbers and in most cases to low and moderate Damkohler number. Kosaly⁶⁵ has stated that CMC is exact at the limit of zero turbulence Reynolds number (no convection). We have not been able to find a satisfactory proof for this. If it is true, questions of validity at high Reynolds number must be addressed by other means. For reasons that will become apparent, we look first at the issues of correctly modelling the conditional average scalar dissipation. We then examine what should be the primary question - the validity of the fundamental closure hypotheses for the basic CMC equation, as have been summarised in Section 3.3. We then address issues concerned with the limitations of first order closure of the conditional average chemical reaction rate term.

7.2.1. Correctly modelling conditional scalar dissipation

Mell $et al^{91}$ carried out DNS in homogeneous turbulence with an initial turbulence Reynolds number based on the Taylor microscale $Re_{\lambda} = 35$ and for a simple equimolar reaction without any temperature dependence of DNS calculations were made for two values of the mean reaction rate. mixture fraction, <ξ>, which was also the stoichiometric value, ξ., namely, $<\xi > = \xi_s = 0.25$ and 0.5. For each of these cases, three different initial Damkohler numbers, Da_0 , were computed, $Da_0 = 0.5$, 2 and 8. In the CMC modelling two different models were used for the conditional average scalar dissipation:

Case (a):
$$\langle N | \eta \rangle_t = \langle N \rangle_t$$
 (7.3)

Case (b):
$$\langle N | \eta \rangle_t = \langle N | \eta = 0.5 \rangle_t F_1(\eta)$$
 (7.4)

where the function $F_1(\eta)$ is obtained from counterflow laminar flame modelling as

$$F_1(\eta) = \exp\left\{-2\left(\operatorname{erf}^{-1}(2\eta - I)\right)^2\right\}$$
(7.5)

The values of $\langle N \rangle_t$ and $\langle N | \eta = 0.5 \rangle_t$ on the RHS of Eq. (7.3) and (7.4), respectively, were obtained from the DNS. The modelling of Eq. (7.4), (7.5) is also obtained as the result of Kraichnan's²⁶ amplitude mapping closure (AMC) and solutions for the initial stage of mixing¹⁰⁵.

The unconditional average of the product of the reactant species mass fractions, $\langle Y_A Y_B \rangle$, gives a sensitive overall measure of the goodness of the CMC modelling, since this is proportional to the mean reaction rate and will have consequent effects on the unconditional averages of the species mass fractions. Table 2 from Mell *et al*⁹¹ shows the errors arising in the CMC predictions for this statistic obtained from

$$\langle Y_{A}Y_{B}\rangle_{CMC} = \int_{0}^{I} Q_{A}(\eta) Q_{B}(\eta) P(\eta) d\eta$$
 (7.6)

with $P(\eta)$ taken from the DNS. The error is reported relative to the DNS value. Time is normalised by the initial eddy turnover time, l_0/u'_0 , where l_0 is the initial integral length scale of the velocity field and u'_0 is the initial value of the rms velocity component fluctuations. It is seen that the errors arising from the use of the unconditional dissipation, Case (a), are large, particularly near the beginning of the experiment and when the Damkohler number, Da₀, is high. Results are much better for the counterflow model, Case (b), particularly where $\langle \xi \rangle = \xi_s = 0.5$ and Da_0 is not high.

These errors in the CMC predictions can almost entirely be attributed to errors in the modelling of $\langle N | \eta \rangle$. As shown in Section 4.2.3 errors in $\langle N | \eta \rangle$, $\Delta N(\eta)$, result in a false chemical source term, $\langle E_N \rangle$, which in the present context may be evaluated as

$$\langle E_{\rm N} \rangle = {\rm Da}_0^{-1} \int_0^1 \Delta N(\eta) P(\eta) \frac{\partial^2 Q}{\partial \eta^2} d\eta$$
 (7.7)

This false reaction rate term is essentially identical to the error in The second derivative $\partial^2 Q / \partial \eta^2$ peaks strongly at $\eta = \xi_s$ and errors $\langle Y_{\rm A}Y_{\rm B}\rangle$. at this location in η space are of most importance although this is modified by the values of $P(\eta)$ in this region. The strength of the peak at a given time strongly increases with Damkohler number, since the fluid is then more highly reacted and closer to the equilibrium chemistry limit second derivative becomes infinite. Figures 36-39 where the show the pdfs, $\langle N | \eta \rangle / \langle N \rangle$, $\langle N \rangle$ and the fraction of the reaction rate balanced by the transient and diffusive terms. Figure 38 also shows the modelling for $\langle N|\eta \rangle$ used in Case (b). Note that the modelling for Case (a) is inherent in the normalisation used: $\langle N | \eta \rangle \langle N \rangle$. It is seen that the modelling of Case (a) involves very large errors in $\langle N | \eta \rangle$, particularly at early times. From this information and Eq. (7.7), it is possible to make an approximate check on the errors involved in using the modelling of Case (a) for $\langle N|\eta \rangle$. A more accurate check is obtained by referring to the Case (b) results where the modelling for Case (b) is accurate. It is seen that the correct modelling of $\langle N | \eta \rangle$ accounts for essentially all of the error, except for $Da_0 = 8$, where errors of up to 5% remain unaccounted for by this means. Mell et al^{91} propose that the modelling of $\langle N|\eta \rangle$ by the counterflow or AMC model could be improved for the $\langle \xi \rangle = \eta_s = 0.25$ case by matching at $\eta =$ 0.25 rather than at $\eta = 0.5$ as is done in Eq. (7.4).

Swaminathan and Mahalingam¹³³ have also examined the effects of conditional modelling for the average scalar dissipation CMC on They compared results using the AMC model of Eq. (7.4), predictions. (7.5) with those using the model developed by Girimaji⁴¹ based on the use of a beta function pdf for the mixture fraction. This model yields

$$\langle N|\eta \rangle_{t} = -2 \langle N \rangle_{t} \langle \xi \rangle \left(I - \langle \xi \rangle \right) \frac{I(\eta)}{\sigma^{4} P(\eta)}$$
(7.8)

where σ^2 is the mixture fraction variance and $I(\eta)$ is given by

$$I(\eta) = \iint_{0}^{\eta} \left\{ <\xi > \left(\ln(\eta^{\circ}) - <\ln(\xi) > \right) + (1 - <\xi >) \left(\ln(1 - \eta^{\circ}) - <\ln(1 - \xi) > \right) \right\} P(\eta^{\circ}) d\eta^{\circ}$$

$$(7.9)$$

In theory, Girimaji's model should give better performance than the AMC model at later times since the AMC model strictly requires there to be some unmixed fluid present in the mixture. No such restriction applies to Girimaji's model. The simulations carried out by Swaminathan and Mahalingam¹³³ start with an inhomogeneous layer and it is only late in the calculation that the mixing field becomes homogeneous. At these later times Girimaji's model does seem to give better results for the dissipation. conditional average scalar Results for the CMC modelling appear better with the AMC model but this is clouded by the lack of homogeneity in the scalar fields at early times.

7.2.2. Validity of the primary closure hypothesis

We are concerned here with the fundamental closure hypotheses of CMC assumed in Eqs (3.13) and (3.28). As already pointed out, these can be shown to be essentially the same, and essentially the same as Eq. (3.31). Taking into account the fact that it is $\partial J_Y / \partial \eta$ that appears in the CMC Eq.(3.4), the closure hypothesis is correctly stated as

$$2 \frac{\partial \langle D\nabla Y'' \cdot \nabla \xi | \eta \rangle P(\eta) \rho_{\eta}}{\partial \eta} - \frac{\partial^2 \langle NY'' | \eta \rangle P(\eta) \rho_{\eta}}{\partial \eta^2} = 0$$
(7.10)

Mell⁸⁹ examines the "error" terms $\langle D\nabla Y'' \cdot \nabla \xi | \eta \rangle$ and $\langle NY'' | \eta \rangle$ in the data base of Mell *et* al^{91} and finds them to be significant fractions of the corresponding retained terms $\langle N | \eta \rangle \partial Q / \partial \eta$ and $\langle N | \eta \rangle Q$, particularly at the The signs of these "error" terms are however highest Damkohler number. the same and they may almost completely cancel when the true test of Eq.(7.10) is applied. Further examination of this issue using the DNS data bases is warranted.

7.2.3. Closure of the chemical source term

For a one-step irreversible reaction errors in first order conditional moment closure of the conditional reaction rate are given by

$$k < Y_{\rm A} Y_{\rm B} |\eta > - k Q_{\rm A} Q_{\rm B} = k < Y''_{\rm A} Y''_{\rm B} |\eta >$$
 (7.11)

where the effect of temperature fluctuations on the rate constant *k* are negligible. Mell⁸⁹ using the data bases of Mell *et al*⁹¹ found that the error was less than 1% of $k < Y_A Y_B |\eta >$ at Da₀ = 0.5, but rises to as much as 7% at Da₀ = 8. It should be noted that there could be a strong Reynolds number dependence on such results since the fluctuations in *N* about $<N|\eta>$ increase with Reynolds number. It is not yet clear whether the error will continue to increase with Da₀ as reaction becomes confined to a thin sheet and all terms in Eq.(7.11) become zero outside this sheet. This question should be answerable by appropriate asymptotic analysis. In Section 8 we consider the formulation and modeling of balance equations for covariances such as $<Y''_AY''_B|\eta>$ and closure of the chemical source term at the second order.

Inferring general conclusions from such results is foolhardy. If the strongly temperature dependent, we constant is expect to find the rate errors from first order conditional moment closure to be greatest at low Da_0 , since now there will be increases in Y''_A and Y''_B due to local extinctions or much reduced reaction rates where the temperature is low. Pope⁷⁵ studied reversible one-step Lee and chemistry with high a activation energy for the rate and constant, find that first conditional moment closure is good only at high significant Da_0 . At low Da_0 extinction occurs and first order closure becomes innaccurate. They did investigate the possibility of obtaining better results with a higher not order conditional moment closure.

7.3. Multi-step kinetics

Montgomery *et al*⁹⁹ consider the H₂-O₂ reaction from a fundamental kinetics point of view but end up calculating the reaction using a one-step reduced mechanism. They find good agreement with CMC predictions for unconditional averages of major species and temperature even when assuming $\langle N|\eta \rangle_t = \langle N \rangle_t$. Proper modelling for $\langle N|\eta \rangle_t$ is needed to get accurate predictions of reaction rate and conditional average statistics.

Mahalingam¹³³ Swaminathan and consider a 3-step series parallel reaction with two intermediate species. They find that the intermediate species are more sensitive to correct modelling of the scalar dissipation that a second order closure would improve predictions for the and

intermediate species with the highest Damkohler number.

Swaminathan and Bilger¹²⁹ have studied a 2-step reduced mechanism for hydrocarbon combustion. They find¹²⁵ that CMC predictions of both the major and minor species are excellent, although second order closure would improve predictions of the conditional average reaction rates.

8. CONDITIONAL VARIANCE EQUATION AND SECOND ORDER CLOSURE

In this section we consider the conditional expectations of the second moments of reactive scalars which are given by

$$K \equiv (Y'')^{2}; \quad G \equiv \langle K | \eta \rangle$$

$$K_{ij} \equiv Y''_{i}Y''_{j}; \quad G_{ij} \equiv \langle K_{ij} | \eta \rangle$$
(8.1)

The double prime denotes fluctuations with respect to the conditional mean $Y'' \equiv Y - Q,$ $Q \equiv \langle Y | \eta \rangle$. (conditional fluctuations) The matrix G_{ii} is the conditional covariance matrix. It is obvious that the matrices in Eq.(8.1) $G_{\rm ii} = G_{\rm ii}$. Normally, are symmetric: $K_{ij} = K_{ji}$ the conditional fluctuations of the reactive components are significantly smaller the than unconditional fluctuations and the conditional fluctuations can often be neglected when the average value of the source terms in kinetic equations is calculated (see Section 3.5). However, if conditional fluctuations of reactive components are not small, the conditional expectations of the second moments can be used to find more accurate average values of the source terms. Eq. (3.38) can be used for this purpose provided that the higher order terms are negligible. This will generally be the case if the corrections themselves are significantly less than first order the term. Since correlation coefficients lie between -1 and 1. this will be achieved for

$$[]^{1/2} << Q_{i}$$

$$[]^{1/2} / Q_{T} << 3Q_{T} / T_{a} \quad \text{if} \quad \beta \leq 1$$

$$<< 3/(\beta-2) \quad \text{if} \quad \beta > 2 \text{ and } T_{a} < Q_{T}$$
(8.3)

where β is the temperature index and T_a is the activation temperature.

Alternatively, we may use Eq.(2.10) & (2.15) and write

$$\langle W_{i}(\mathbf{Y}) | \eta \rangle = \int_{\infty} W_{i}(\mathbf{Z}) P(\mathbf{Z} | \eta) dZ_{1} dZ_{2} ... dZ_{n}$$
(8.4)

where $Y_{1,...,Y_{n-1}}$ are governed by Eq.(3.2); $Y_{n} \equiv h$ is governed by

Eq.(3.33) and $P(\mathbf{Z}|\eta)$ is the conditional pdf (introduced in Sec.2.1.3) which can be assumed to have a presumed form such as the Gaussian form

$$P(\mathbf{Z}|\boldsymbol{\eta}) = \left((2\pi)^{n} \det(G_{ij}) \right)^{-1/2} \exp\left[-\frac{1}{2} g_{ij}(Z_{i} - Q_{i})(Z_{j} - Q_{j}) \right]$$
(8.5)

inverse matrix of G_{ij} that is g_{ii} is where the $g_{ik}G_{kj}=\delta_{ij}$. A joint Gaussian form may be necessary to avoid unrealizable clipped portions of the Z domain. For extinction problems a bimodal form may be better since the fluid is either alight or extinguished with almost nothing in between.

CMC models for G have been derived by $Bilger^{11}$ using the decomposition technique and by Klimenko^{55,56} using the pdf approach. Li and Bilger⁷⁶ formulated a CMC model for G_{ij} using the decomposition and decomposition techniques technique. Derivations using the pdf are considered in Sections 8.1 and 8.2 respectively. The model for G_{ij} is formulated first and then the equation for G is derived by putting i=j. General discussion of the CMC equations for the second moments can be found in Section 8.3. Some comments on the application of second order closure are presented in Section 8.4 including simplifications that make the technique more tractable for practical systems. Without loss of generality we can put i=1, j=2 from here on in this section.

8.1. The Pdf Method

First we derive the equation for $G_{12}^+ \equiv \langle Y_1 Y_2 | \eta \rangle = G_{12} + Q_1 Q_2$. The relationship of G_{12}^+ , the joint pdf $P(Z,\eta)$ the pdf $P(\eta)$ is given by

$$G_{12}^{+} P(\eta) \equiv \langle Y_1 Y_2 | \xi = \eta \rangle P(\eta) = \int_{\infty}^{\infty} Z_1 Z_2 P(Z_1, Z_2, \eta) \, dZ_1 dZ_2$$
(8.6)

The joint pdf equation (2.48) is used in the derivation. We assume that $\eta = Z_3$, $W_3 = 0$ and n = 3 in Eq.(2.48) and consider this equation as the equation for $P(Z_1, Z_2, \eta)$. Equation (2.48) is multiplied by Z_1Z_2 and integrated over all Z_1 and Z_2 . Note that Z_1 and Z_2 are independent variables and may be taken inside derivatives with respect to t, \mathbf{x} and η . The terms which

involve $\partial/\partial Z_1$ and $\partial/\partial Z_2$ are integrated by parts. Note also that $P(\mathbf{Z}, \eta) \rightarrow 0$ as $|\mathbf{Z}| \rightarrow \infty$. The result of the integration is

$$\frac{\partial G_{12}^{+}P(\eta)\rho_{\eta}}{\partial t} + \operatorname{div}\left(\langle \mathbf{v}K_{12}^{+}|\eta\rangle P(\eta)\rho_{\eta}\right) = \langle W_{1}Y_{2} + W_{2}Y_{1}|\eta\rangle P(\eta)\rho_{\eta}$$
$$- 2 \langle D(\nabla Y_{1}\cdot\nabla Y_{2})|\eta\rangle P(\eta)\rho_{\eta} + \frac{\partial J_{12}}{\partial \eta}$$
(8.7)

$$J_{12} \equiv -\frac{\partial \langle NK_{12}^{\dagger} | \eta \rangle P(\eta) \rho_{\eta}}{\partial \eta} + 2 \langle D(\nabla K_{12}^{\dagger} \cdot \nabla \xi) | \eta \rangle P(\eta) \rho_{\eta}$$
(8.8)

where $\rho_{\eta} \equiv \langle \rho | \eta \rangle$, $K_{12}^{\dagger} \equiv Y_1 Y_2$, $G_{12}^{\dagger} \equiv \langle K_{12}^{\dagger} | \eta \rangle$ and $N \equiv D(\nabla \xi)^2$ is the dissipation of the conserved scalar. The closure assumptions used by Klimenko⁵⁵

$$< NK_{12}^{+} |\eta > = < N |\eta > < K_{12}^{+} |\eta >$$
 (8.9)

$$\langle D(\nabla K_{12}^{+} \cdot \nabla \xi) | \eta \rangle = \langle N | \eta \rangle \frac{\partial \langle K_{12}^{+} | \eta \rangle}{\partial \eta}$$
(8.10)

are similar to the assumptions in Eqs.(3.47) and (3.48). As we note in Section 8.3 these assumptions may be too restrictive. With these assumptions Eq.(8.8) takes the form

$$J_{12} = - \frac{\partial \langle N|\eta \rangle P(\eta)\rho_{\eta}}{\partial \eta} G_{12}^{+} + \langle N|\eta \rangle P(\eta)\rho_{\eta} \frac{\partial G_{12}^{+}}{\partial \eta}$$
(8.11)

With the use of the pdf equation (3.15) and closure (8.11) Eq.(8.7) can now be written in the form

$$\frac{\partial G_{12}^{+}}{\partial t} + \langle \mathbf{v} | \boldsymbol{\eta} \rangle \cdot \nabla G_{12}^{+} + \frac{\operatorname{div} \left(\langle \mathbf{v}''(Y_{1} Y_{2})'' | \boldsymbol{\eta} \rangle P(\boldsymbol{\eta}) \rho_{\boldsymbol{\eta}} \right)}{P(\boldsymbol{\eta}) \rho_{\boldsymbol{\eta}}} - \langle N | \boldsymbol{\eta} \rangle \frac{\partial^{2} G_{12}^{+}}{\partial \boldsymbol{\eta}^{2}} = \langle W_{1} Y_{2} + W_{2} Y_{1} | \boldsymbol{\eta} \rangle - 2 \langle D(\nabla Y_{1} \cdot \nabla Y_{2}) | \boldsymbol{\eta} \rangle P(\boldsymbol{\eta}) \rho_{\boldsymbol{\eta}}$$
(8.12)

The last term in this equation may be written

$$< D(\nabla Y_1 \cdot \nabla Y_2) |\eta\rangle = < D(\nabla Y_1'' \cdot \nabla Y_2'') |\eta\rangle + \frac{\partial Q_1}{\partial \eta} \frac{\partial Q_2}{\partial \eta} N$$
(8.13)
Here, we have used Eq.(3.19), have neglected the terms which involve ∇Q since these terms are small for large Reynolds numbers (see Sec.2.3.3) and have taken into account that

$$< D(\nabla Y'' \cdot \nabla \xi) | \eta > = < N | \eta > \frac{\partial < Y'' | \eta >}{\partial \eta} = 0$$

according to Eq.(3.48). The identity

$$(Y_1Y_2)'' = K_{12}^+ - G_{12}^+ = \left((Q_1 + Y_1'') (Q_2 + Y_2'') \right) - Q_1Q_2 - G_{12} =$$
$$= Q_1Y_2'' + Q_2Y_1'' + K_{12} - G_{12}$$

is used to modify the third term in Eq.(8.12)

$$div \left(\langle \mathbf{v}''(Y_1Y_2)'' | \eta \rangle P(\eta)\rho_{\eta} \right) - Q_2 div \left(\langle \mathbf{v}''Y_1'' | \eta \rangle P(\eta)\rho_{\eta} \right) - Q_1 div \left(\langle \mathbf{v}''Y_2'' | \eta \rangle P(\eta)\rho_{\eta} \right) = div \left(\langle \mathbf{v}''K_{12}'' | \eta \rangle P(\eta)\rho_{\eta} \right) + \langle \mathbf{v}''Y_1'' | \eta \rangle P(\eta)\rho_{\eta}\nabla Q_2 + \langle \mathbf{v}''Y_2'' | \eta \rangle P(\eta)\rho_{\eta}\nabla Q_1$$

$$(8.14)$$

The next step in the derivation is obtaining the equation for the Q_1 Q_2 . product Equation (3.16) written for is multiplied $Q_1 Q_2$. by written for Q_2 is multiplied by Q_1 . Equation (3.16) Adding the results yields

$$\frac{\partial Q_1 Q_2}{\partial t} + \langle \mathbf{v} | \boldsymbol{\eta} \rangle \cdot \nabla (Q_1 Q_2) + \frac{1}{P(\boldsymbol{\eta}) \rho_{\boldsymbol{\eta}}} \left\{ Q_2 \operatorname{div} \left\{ \langle \mathbf{v}'' Y_1'' | \boldsymbol{\eta} \rangle P(\boldsymbol{\eta}) \rho_{\boldsymbol{\eta}} \right\} + Q_1 \operatorname{div} \left\{ \langle \mathbf{v}'' Y_2'' | \boldsymbol{\eta} \rangle P(\boldsymbol{\eta}) \rho_{\boldsymbol{\eta}} \right\} \right\}$$
$$= \langle N | \boldsymbol{\eta} \rangle \frac{\partial^2 Q_1 Q_2}{\partial \boldsymbol{\eta}^2} - 2 \langle N | \boldsymbol{\eta} \rangle \frac{\partial Q_1}{\partial \boldsymbol{\eta}} \frac{\partial Q_2}{\partial \boldsymbol{\eta}} + Q_2 \langle W_1 | \boldsymbol{\eta} \rangle + Q_1 \langle W_2 | \boldsymbol{\eta} \rangle \quad (8.15)$$

Subtracting Eq.(8.15) from Eq.(8.12) and using Eqs.(8.13) and (8.14) we obtain

$$\frac{\partial G_{12}}{\partial t} + \langle \mathbf{v} | \boldsymbol{\eta} \rangle \cdot \nabla G_{12} + \frac{\operatorname{div} \left(\langle \mathbf{v}'' Y_1'' Y_2'' | \boldsymbol{\eta} \rangle P(\boldsymbol{\eta}) \rho_{\boldsymbol{\eta}} \right)}{P(\boldsymbol{\eta}) \rho_{\boldsymbol{\eta}}} - \langle N | \boldsymbol{\eta} \rangle - \frac{\partial^2 G_{12}}{\partial \boldsymbol{\eta}^2} = \\ = \langle W_1'' Y_2'' + W_2'' Y_1'' | \boldsymbol{\eta} \rangle - 2 \langle D(\nabla Y_1'' \cdot \nabla Y_2'') | \boldsymbol{\eta} \rangle - \\ \langle \mathbf{v}'' Y_2'' | \boldsymbol{\eta} \rangle \nabla Q_1 - \langle \mathbf{v}'' Y_1'' | \boldsymbol{\eta} \rangle \nabla Q_2$$
(8.16)

Another form of this equation can be obtained with the use of Eq.(3.15):

$$\frac{\partial G_{12} P(\eta) \rho_{\eta}}{\partial t} + \operatorname{div} \left(\langle \mathbf{v} K_{12} | \eta \rangle P(\eta) \rho_{\eta} \right) = \\ - \frac{\partial^{2} \langle N | \eta \rangle P(\eta) \rho_{\eta}}{\partial \eta^{2}} G_{12} + \langle N | \eta \rangle P(\eta) \rho_{\eta} \frac{\partial^{2} Q_{12}}{\partial \eta^{2}} + \\ + P(\eta) \rho_{\eta} \left(\langle W_{1}^{"'} Y_{2}^{"'} + W_{2}^{"'} Y_{1}^{"'} | \eta \rangle - 2 \langle D(\nabla Y_{1}^{"'} \cdot \nabla Y_{2}^{"'}) | \eta \rangle + \langle \mathbf{v}^{"'} Y_{2}^{"'} | \eta \rangle \nabla Q_{1} - \langle \mathbf{v}^{"'} Y_{1}^{"'} | \eta \rangle \nabla Q_{2} \right)$$

The equation for the conditional variance can be easily obtained by putting $Y_1=Y_2=Y$ in Eq.(8.16)

$$\frac{\partial G}{\partial t} + \langle \mathbf{v} | \eta \rangle \nabla G + \frac{\operatorname{div} \left(\langle \mathbf{v}''(Y'')^2 | \eta \rangle P(\eta) \rho_{\eta} \right)}{P(\eta) \rho_{\eta}} - \langle N | \eta \rangle \frac{\partial^2 G}{\partial \eta^2} =$$
$$= 2 \langle W''Y'' | \eta \rangle - 2 \langle D(\nabla Y'')^2 | \eta \rangle + 2 \langle \mathbf{v}''Y'' | \eta \rangle \nabla Q \qquad (8.18)$$

8.2. Decomposition Method

The first step in the derivation of the equation for G_{12} using the decomposition method is the equation for $K_{12} \equiv Y_1''Y_2''$

$$\rho \frac{\partial K_{12}}{\partial t} + \rho \mathbf{v} \cdot \nabla K_{12} - \operatorname{div} \left(D \rho \nabla K_{12} \right) + 2 D \rho \left(\nabla Y_1'' \cdot \nabla Y_2'' \right) =$$
$$= \rho (W_1 Y_2'' + W_2 Y_1'') + E_{D12} + E_{D21} - E_{Q12} - E_{Q21}$$
(8.19)

where

$$E_{\text{Dij}} \equiv Y_{i}^{"}\rho D\nabla\xi \cdot \nabla -\frac{\partial Q_{i}}{\partial\eta} + Y_{i}^{"}\text{div}(\rho D\nabla Q_{j})$$
(8.20)

$$E_{\text{Qij}} = Y_{i}'' \left(\rho \frac{\partial Q_{j}}{\partial t} + \rho \mathbf{v} \cdot \nabla Q_{j} - \rho N \frac{\partial^{2} Q_{j}}{\partial \eta^{2}} \right)$$
(8.21)

This equation is derived from Eq.(3.21) which is valid for both Y''_1 and Y''_2 . The correlation is decomposed in a way which is similar to Eq.(3.17)

$$K_{12}(\mathbf{x},t) = G_{12}(\xi(\mathbf{x},t),\mathbf{x},t) + K_{12}''(\mathbf{x},t)$$
(8.22)

where K_{ij} and G_{ij} is given by (8.1). It is easy to see that $\langle K_{12}''(\mathbf{x},t) | \eta \rangle = 0$. The differentiation of Eq.(8.22) yields

$$\frac{\partial K_{ij}}{\partial t} = \frac{\partial G_{i}}{\partial t} + \frac{\partial G_{ij}}{\partial \eta} \frac{\partial \xi}{\partial t} + \frac{\partial K''_{ij}}{\partial t}$$
(8.23)

$$\nabla K_{ij} = \nabla G_{ij} + \frac{\partial G_{ij}}{\partial \eta} \nabla \xi + \nabla K''_{ij}$$
(8.24)

Equation (8.19) takes the form

$$\rho \frac{\partial G_{12}}{\partial t} + \rho \mathbf{v} \cdot \nabla G_{12} - \rho N \frac{\partial^2 G_{12}}{\partial \eta^2} + 2D\rho (\nabla Y_1'' \cdot \nabla Y_2'') = \rho (W_1 Y_2'' + W_2 Y_1'') + E_{D12} + E_{D21} - E_{Q12} - E_{Q21} + E_G - E_K$$
(8.25)

where

$$E_{\rm G} \equiv \operatorname{div}(\rho D \nabla G_{\rm ij}) + \rho D \nabla \xi \cdot \nabla - \frac{\partial G_{\rm ij}}{\partial \eta}$$
(8.26)

$$E_{\rm K} \equiv \rho \frac{\partial K_{12}''}{\partial t} + \rho \left(\mathbf{v} \cdot \nabla K_{12}'' \right) - {\rm div} \left(D \rho \nabla K_{12}'' \right)$$
(8.27)

Taking the expectation of Eq.(8.25), conditional on $\xi(\mathbf{x},t)=\eta$, yields

$$\rho_{\eta} \frac{\partial G_{12}}{\partial t} + \rho_{\eta} < \mathbf{v} | \eta > \nabla G_{12} - \rho_{\eta} < N | \eta > \frac{\partial^2 G_{12}}{\partial \eta^2} = \rho_{\eta} < W_1'' Y_2''' + W_2'' Y_1'' | \eta > - 2\rho_{\eta} < D(\nabla Y_1'' \cdot \nabla Y_2'') | \eta > - e_{\mathrm{K}} - e_{\mathrm{Q12}} - e_{\mathrm{Q21}}$$
(8.28)

where $e_{\rm K} \equiv \langle E_{\rm K} | \eta \rangle$ and

$$e_{\text{Qij}} \equiv \langle E_{\text{Qij}} | \eta \rangle = \rho_{\eta} \left(\nabla Q_{j} \langle Y_{i}'' | \eta \rangle - \frac{\partial^{2} Q_{j}}{\partial \eta^{2}} \langle Y_{i}'' | \eta \rangle \right)$$
(8.29)

According to the analysis of Sec.2.3.3, the terms $\langle E_{\text{Dij}} | \eta \rangle$ and $\langle E_{\text{G}} | \eta \rangle$ are small for large Reynolds numbers. These terms are neglected in Eq.(8.28). Let us analyze the unconditional mean of the term E_{K} .

$$\langle E_{\mathrm{K}} \rangle = \int e_{\mathrm{K}} P(\eta) d\eta = \langle \frac{\partial K_{12}'' \rho}{\partial t} + \operatorname{div} \left(\rho \mathbf{v} K_{12}'' \right) - \operatorname{div} \left(D \rho \nabla K_{12}'' \right) \rangle =$$

$$\frac{\partial \langle K_{12}'' \rho \rangle}{\partial t} + \operatorname{div} \left(\langle \rho \mathbf{v} K_{12}'' \rangle \right) - \operatorname{div} \left(\langle D \rho \nabla K_{12}'' \rangle \right) = \operatorname{div} \left(\langle \rho \mathbf{v}'' K_{12}'' \rangle \right) =$$

$$= \operatorname{div} \left(\int \rho_{\eta} \langle \mathbf{v}'' K_{12}'' | \eta \rangle P(\eta) d\eta \right) = \int \operatorname{div} \left(\rho_{\eta} \langle \mathbf{v}'' K_{12}'' | \eta \rangle P(\eta) \right) d\eta$$

$$(8.30)$$

The derivation of this equation is similar to the derivation of Eq.(3.27). The basic closure used here is similar to Eq.(3.28)

$$e_{\mathrm{K}}P(\eta) = \operatorname{div}\left(\rho_{\eta} < \mathbf{v}'' K_{12}'' | \eta > P(\eta)\right)$$
(8.31)

With this closure, equation (8.28) takes the form

$$\frac{\partial G_{12}}{\partial t} + \langle \mathbf{v} | \eta \rangle \cdot \nabla G_{12} - \langle N | \eta \rangle \frac{\partial^2 G_{12}}{\partial \eta^2} + \frac{\operatorname{div} \left(\langle \mathbf{v}'' K_{12}'' | \eta \rangle P(\eta) \rho_{\eta} \right)}{P(\eta) \rho_{\eta}}$$

$$= \langle W_1'' Y_2'' + W_2'' Y_1'' | \eta \rangle - 2 \langle D(\nabla Y_1'' \cdot \nabla Y_2'') | \eta \rangle - \nabla Q_1 \cdot \langle Y_2'' \mathbf{v}'' | \eta \rangle - \nabla Q_2 \cdot \langle Y_1'' \mathbf{v}'' | \eta \rangle + \frac{\partial^2 Q_1}{\partial \eta^2} \langle Y_2'' N'' | \eta \rangle + \frac{\partial^2 Q_2}{\partial \eta^2} \langle Y_1'' N'' | \eta \rangle$$
(8.32)

The governing equation for the conditional variance can be obtained from Eq.(8.32) by assuming $Y_1=Y_2=Y$

$$\frac{\partial G}{\partial t} + \langle \mathbf{v} | \boldsymbol{\eta} \rangle \cdot \nabla G - \langle N | \boldsymbol{\eta} \rangle \frac{\partial^2 G}{\partial \boldsymbol{\eta}^2} + \frac{\operatorname{div} \left(\langle \mathbf{v}'' K'' | \boldsymbol{\eta} \rangle P(\boldsymbol{\eta}) \rho_{\boldsymbol{\eta}} \right)}{P(\boldsymbol{\eta}) \rho_{\boldsymbol{\eta}}}$$

$$= 2 \langle W''Y'' | \eta \rangle - 2 \langle D(\nabla Y'')^2 | \eta \rangle - 2 \nabla Q \langle Y''v'' | \eta \rangle + 2 \frac{\partial^2 Q}{\partial \eta^2} \langle Y''N'' | \eta \rangle$$
(8.33)

8.3. Discussion

If the Reynolds numbers is large and differential diffusion effects represent neglected, equations (8.32)and (8.33) the most complete are form of the second moment CMC equation. Let us compare the equations derived by the pdf and decomposition methods. Equations (8.18) and (8.33) are the same except that there is one extra term in Eq.(8.33). The last term of Eq.(8.33), which is denoted in this section $-T_4$, as does not appear in Eq.(8.18). A similar conclusion can be drawn form comparison of Eqs.(8.32) and (8.16): the last two terms of Eq.(8.32) does not appear in Eqs.(8.16). If we apply hypothesis (3.47) to these terms, we obtain that these terms are zero. For example, $\langle Y''N''|\eta\rangle = 0$. This indicates that (3.47), (3.48),hypotheses (8.9)and (8.10)used closure to obtain Eqs.(8.16) and (8.18) are more restrictive than closure hypotheses (3.10),(3.28) and (8.31). The more restrictive hypotheses, applied to Eqs.(8.32) and (8.33), transform these equations into Eqs.(8.16) and (8.18).

The more restrictive hypotheses in Eqs.(8.16) and (8.18), used in the earler work of Klimenko⁵⁵, correspond to the old Kolmogorov theory⁶³ which neglects the large-scale (> $l_{\rm K}$) fluctuations of the scalar dissipation. Klimenko⁵⁶ demonstrated that the refined Kolmogorov theory⁶⁴ requires that the additional source term

$$-T_4 \approx 2 \left(\frac{\partial^2 Q}{\partial \eta^2} \right)^2 \left(\langle N | \eta \rangle \right)^2 \tau_{\rm N}; \tag{8.34}$$

should be added to Eq.(8.18). Here, $\tau_{\rm N}$ is the integral Lagrangian time scale for the scalar dissipation N. It appears that the term T_4 is Eq.(8.33). Equations (8.32)automatically present in (8.33)and were obtained by Li and Bilger⁷⁶ (although the third terms on the left-hand side of Eqs.(8.32) and (8.33) were missed). Term (8.34) can be considered as a modeling assumption for the term T_4 in Eq.(8.33).

Let us compare Eq.(4.2) and Eq.(8.33). Equation (8.33)has some dissipation term $2 < D(\nabla Y'')^2 |\eta\rangle$, the additional terms: the generation term $-2 < \mathbf{v''} Y'' \mid \eta > \nabla Q$ the extra-generation and term due *N*-fluctuations to

 $2 < Y''N'' | \eta > \partial^2 Q / \partial \eta^2$. These terms need modeling. Li and Bilger⁷⁶ considered the one-step irreversible reaction in a steady-state turbulent scalar mixing layer and suggested

$$2 < D(\nabla Y'')^2 |\eta\rangle = C_1 G / \tau_{\rm Y}$$
(8.35)

$$2 < Y'' N'' |\eta > \frac{\partial^2 Q}{\partial \eta^2} = C_2 < N |\eta > G^{1/2} \frac{\partial^2 Q}{\partial \eta^2}$$
(8.36)

where $\tau_{Y}=k_{t}/\varepsilon_{t}$ is the integral time scale, C_{1} and C_{2} are constants. The generation term $-2 < \mathbf{v}'' Y'' | \eta > \nabla Q$ was neglected. We can expect that in turbulent shear flow this term can be neglected in most of the cases since the transverse gradients of Q are small in shear flows (see Section 5). The last term on the left-hand side of Eq.(8.33) can also be neglected.

Bilger⁷⁶ Li and compared the modeling results with direct experimental measurements of the conditional variance G made in the reacting turbulent scalar mixing layer presented in Section 5.8.1. Their modelled equation for the normalised first moment \hat{Q}_1 includes an extra term $-\hat{G}$ on the RHS to account for second order closure of the reaction rate. Here \hat{G} is the normalised version of G, using the same normalisation as for \hat{O} . The effect of this term was found to be entirely negligible. The modelled equation for \hat{G} was

$$\frac{\partial \hat{G}}{\partial \hat{x}_1} = -2\hat{G}\left(2\hat{Q}_1 - \eta + \eta_s\right) + AC_2\hat{x}_1\hat{G}^{1/2}\frac{\partial^2 Q}{\partial \eta^2} - \mu_k C_1\hat{x}_1\hat{G}^{1/2}\hat{G}$$
(8.37)

Here m is the power law index for the decay of the turbulence kinetic energy with distance x_1 ; with $\mu_k = 1.3$ taken from the experimental data. In this modelled equation the chemical term has been closed at the second moment level: the extra term involving the conditional third moment $\langle Y''^3 | \eta \rangle$ being found in the experimental data to be never more than 15% of the other chemical term. The boundary conditions used for \hat{G} are

$$\hat{G} = 0$$
 for $x_1 = 0$; and $\hat{G} = 0$ at $\eta = 0, 1$ for $x_1 > 0$.

With these boundary conditions Eq. (8.37) has a singularity at $x_1=0$. Li and Bilger⁷⁶ overcame this by reformulating Eq.(8.37) in terms of $\hat{G}^{1/2}$ which involves dividing the equation through by $\hat{G}^{1/2}$. The term on the RHS

involving $\partial^2 \hat{Q} / \partial \eta^2$ then becomes a source for $\hat{G}^{1/2}$ as soon as \hat{Q} becomes nonlinear under the influence of reaction. It is evident that the sign taken for C_2 in the modelling of Eq. (8.36) needs to reflect whether the species is a reactant or a product. Here species I is a reactant and C_2 is positive. It was set equal to 1.1 to give a good match to the data.

Figure 40 shows the general nature of the results predicted by Li and Bilger⁷⁶. It is seen that the conditional variance peaks at $\eta = \xi_s = 0.5$ this peak increases with distance and that downstream before finally The solutions reflect an approximate balance between the decaying. source term, Eq. (8.36) and the chemical term acting as a sink. The dissipation term, Eq. (8.35) and the convective term, the LHS of Eq.(8.37), are an order of magnitude lower.

Figure 41 shows comparison of the predictions with the data. Separate solutions were made for the value of η_s pertaining to each data set. The predictions for different models scalar figure shows two of the dissipation. Model A is for that presented here and in Section 5.8.1. Model B is more sophisticated and allows for $\langle N|\eta \rangle$ to vary with η . The models give almost identical solutions for the first moment, \hat{Q}_1 , but the more sophisticated model gives a somewhat higher peak value for \hat{G} . The value of $C_2 = 1.1$ was chosen to give the best fit at $x_1 = 15M$ for Model B. figure shows that there is no consistent dependence The of the for the conditional variance position experimental data on across the layer as was also found for the first conditional moment (see Section 5.1 16). This was assumed in the modelling. The data indicate and Figs. 1 and the conditional variance begins to fall that peak at large distances downstream, and this is not predicted by the modelling. The modelling also predicts narrower profiles in η space than is found in the experimental data.

As has been pointed out, Li and Bilger⁷⁶ did not include the third term on the LHS of Eq. (8.33). Estimates indicate that it is an order of magnitude smaller than the source term, Eq. (8.36), and the chemical sink term and of the same order as the dissipation term, Eq. (8.35) and the convective term. Its inclusion may not greatly change the magnitude of the solutions obtained, but may improve the profile shape in η space and the x_1 dependence. The use of the source term of Eq. (8.34) in place of that in Eq. (8.36) is also likely to significantly change the profile shape in η space and in x_1 . This has not been investigated as yet.

Swaminathan and Bilger¹³¹ examine DNS data bases for nonpremixed homogeneous turbulence with reaction in decaying a two-step reduced hydrocarbon combustion¹²⁹ in mechanism for terms of the conditional variance Eq. (8.36). For homogeneous flow this may be written

$$\frac{\partial G}{\partial t} - 2 \langle Y''W'' | \eta \rangle - \langle N | \eta \rangle \frac{\partial^2 G}{\partial \eta^2} - 2 \langle Y''N'' | \eta \rangle \frac{\partial^2 Q}{\partial \eta^2} + 2 \langle D\nabla Y'' \cdot \nabla Y'' | \eta \rangle - e_{\rm K} = 0$$

$$(8.38)$$

Figure 42 shows results for the fuel species in two different simulations It is seen that the conditional species fluctuation scalar R1 and R5. dissipation T_4 , is far from being negligible term, as assumed in Klimenko⁵⁵ and Eq. (8.9). Indeed it is the largest term and is balanced largely by the reaction rate correlation term, T_2 so that an estimate for G can be made by setting these two terms equal. It is seen that the primary closure assumption of Eq. (8.31) is much better. Note that for homogeneous flow this becomes $e_{\rm K} = 0$ and that $e_{\rm K}$ for the fuel species appears as $e_{q_{ee}}$ in Fig. 42. It can be noted that the terms in the balance for R1. This is in part for simulation R5 are much larger than the terms due to the higher turbulence Reynolds number producing higher fluctuations in N, but also due to the fact that the Damkohler numbers in simulation R5 are about half those in R1.

Swaminathan and Bilger¹³¹ also examine the modelling for terms T_4 and T_5 given in Eqs (8.36) and (8.35) respectively. The "constant" C_2 can be expressed

$$C_{2} \equiv \frac{R_{\rm NY} < N''^{2} |\eta|^{1/2}}{< N |\eta|^{1/2}}$$

where $R_{\rm NY}$ is the conditional correlation coefficient between N'' and Y''. It is found that R_{NY} is a function of η but apparently not a strong function the Damkohler Reynolds number. or number. For a first of the approximation, the assumption of it having a magnitude of about 0.8with the appropriate sign may be sufficient. The sign is positive for reactant species and negative for product species. The "constant" C_1 in Eq.(8.35) is also found to be dependent on η , but is also dependent on the Damkohler number.

Swaminathan and Bilger ¹³² have continued the analysis of this same data base in terms of the progress variables for each of the two reaction and have included analysis of the conditional covariance equation. A steps conditional dissipation progress new model for the of the variable fluctuations is proposed based stationary laminar flamelet on modeling. This model is an improvement over that of Eq. (8.35), but further work is still needed to improve its general applicability.

8.4. Second Order Closure

It is becoming evident that the use of a second order closure for the conditional reaction rate term will be of value in such problems the as prediction of autoignition in diesel engines and the formation of NO. It may also be capable of handling flows with local and even bulk ignition and extinction behaviour. Further investigation of the correct form of the conditional variance and conditional covariance equations and appropriate direct numerical modelling for the unclosed terms is merited. Data from simulations (DNS) and laboratory experiment should be useful for doing this.

In systems involving complex multi-step chemistry it may be possible to simplify the making of the second order corrections to the basic first Correlations moment closure. between species mass fractions and temperature could be related to correlations among the much fewer progress variables necessary for a reduced kinetic mechanism. Thus for the H_2 - O_2 system Kronenburg et al⁶⁸ use a full mechanism to compute the leading second order reaction rates but the order of the corrections to these variance of conditional all related to the the fluctuations in rates are total moles per unit mass which is a suitable progress variable for the the one-step reduced mechanism for this system. Only one extra equation for this variance - need then be solved to obtain closure which is made using a presumed form for the conditional pdf of this progress variable. Results for NO formation in a turbulent jet diffusion flame of hydrogen are shown in Fig. 43 for distances from the nozzle exit plane, x_{1} , equal to 0.125, 0.5 and 0.75 of the visible flame length, $L_{\rm v}$. Results at the downstream positions including at the visible flame length (not shown excellent. Discrepancies remain positions here) are at close to the due nozzle. These likely to be to the effects of differential are

diffusion (see Section 9.4) and problems of modelling the mixing at the outer edges of the jet where the flame sits at these locations.

Mastorakos et al^{87} have used DNS to study autoignition in homogeneous turbulence with initially nonpremixed cold fuel and hot air. They find turbulence has a significant effect the that the very on time for ignition. Locally high values of scalar dissipation result in lowering the due conduction losses. temperature to high Fluctuations about the quite large and must be conditional average temperature are included in the modelling of the reaction rate if predictions of the ignition time are to be at all accurate. Mastorakos and Bilger⁸⁶ have formulated a second order closure CMC model for this system and find excellent agreement of the predictions with the DNS as is shown in Fig. 44.

9. CONDITIONAL MOMENT METHODS: FURTHER TOPICS

The topics considered in Section 9 are prospective topics for futher development of the Conditional Moment Closure. These involve the differential diffusion, CMC with multiple conditions, CMC for spray compustion, CMC for premixed combustion and CMC & Lagrangian modelling. The CMC-like techniques can be applied to generate conditional equations with conditioning variables which are different from the conventional mixture fraction. New types of conditions generate new equations with new applications. We shall refer to these CMC-like methods as conditional methods. The extend of the discussions presented here is determined by the relevance to the original topic of this review - the turbulent combustion.

9.1. Differential Diffusion

In this section we consider the equation for the conditional mean $Q \equiv \langle Y | \xi = \eta \rangle \equiv \langle Y | \eta \rangle$ when the conserved scalar and the reactive scalar have different diffusion coefficients

$$\rho \frac{\partial \xi}{\partial t} + \rho \left(\mathbf{v} \cdot \nabla \xi \right) - \operatorname{div} \left(D \rho \nabla \xi \right) = 0$$
(9.1)

$$\rho \frac{\partial Y}{\partial t} + \rho \left(\mathbf{v} \cdot \nabla Y \right) - \operatorname{div} \left(D_{Y} \rho \nabla Y \right) = W \rho$$
(9.2)

That is $D \neq D_Y$ in these equations. The difference in the diffusion coefficients can affect the CMC equation for Q as well as induce some additional generation of the conditional variance G. Here we focus only on the conditional expectations of the first moments. In Sec. 9.1.1 the general unclosed CMC equation is derived. In Sec.9.1.2. we follow Kronenburg and Bilger⁶⁶ and consider the closures which provide good agreement with the DNS calculations.

9.1.1 Derivation of the unclosed equation

As in Sec.3.1.2, we derive the governing equation for $Y\psi_{\eta}$ where $\psi_{\eta} \equiv \delta(\xi(\mathbf{x},t)-\eta)$ and then average it. Equation (2.43) is used as the equation for ψ_{η} ($\eta = Z_1$, $\xi = Y_1$, $W_1 = 0$, n = 1). This equation is multiplied by Y and added to Eq.(9.2) multiplied by ψ_{η} to obtain

$$\frac{\partial \psi_{\eta} \rho Y}{\partial t} + \operatorname{div} \left(\mathbf{v} \psi_{\eta} \rho Y \right) + \frac{\partial \psi_{\eta} Y \operatorname{div}(D \rho \nabla \xi)}{\partial \eta} - \psi_{\eta} \operatorname{div}(D_{Y} \rho \nabla Y) = \psi_{\eta} W \rho \quad (9.3)$$

The derivation of the following equations is similar to the derivation of Eqs.(3.7) and (3.8)

$$\operatorname{div}\left(YD\rho\nabla\psi_{\eta}\right) = \frac{\partial^{2}}{\partial\eta^{2}}\left(\psi_{\eta} YD\rho(\nabla\xi)^{2}\right) - \frac{\partial}{\partial\eta}\left(\psi_{\eta} \operatorname{div}(YD\rho\nabla\xi)\right)$$
$$= \frac{\partial^{2}}{\partial\eta^{2}}\left(\psi_{\eta} YD\rho(\nabla\xi)^{2}\right) - \frac{\partial}{\partial\eta}\left(\psi_{\eta} Y\operatorname{div}(D\rho\nabla\xi)\right) - \frac{\partial}{\partial\eta}\left(\psi_{\eta} D\rho(\nabla\xi\cdot\nabla Y)\right) \quad (9.4)$$

$$\operatorname{div}\left(\psi_{\eta}D_{Y}\rho\nabla Y\right) = \psi_{\eta}\operatorname{div}\left(D_{Y}\rho\nabla Y\right) - \frac{\partial}{\partial\eta}\left(\psi_{\eta}D_{Y}\rho(\nabla\xi\cdot\nabla Y)\right)$$
(9.5)

The two last term on the left-hand side of Eq.(9.3) are modified using Eqs.(9.4) and (9.5)

$$\frac{\partial \psi_{\eta} \rho Y}{\partial t} + \operatorname{div} \left(\mathbf{v} \psi_{\eta} \rho Y \right) + \frac{\partial^{2}}{\partial \eta^{2}} \left\{ \psi_{\eta} Y D \rho (\nabla \xi)^{2} \right\} - \frac{\partial}{\partial \eta} \left\{ \psi_{\eta} (D + D_{Y}) \rho (\nabla \xi \cdot \nabla Y) \right\} - \operatorname{div} \left(D \rho \nabla (Y \psi_{\eta}) \right) - \operatorname{div} \left(\psi_{\eta} (D_{Y} - D) \rho \nabla Y \right) = \psi_{\eta} W \rho$$
(9.6)

Averaging of Eq.(9.6) yields

$$\frac{\partial QP(\eta)\rho_{\eta}}{\partial t} + \operatorname{div}\left(\langle \mathbf{v}Y|\eta \rangle P(\eta)\rho_{\eta}\right) = \langle W|\eta \rangle P(\eta)\rho_{\eta} + \frac{\partial J_{Y}}{\partial \eta} + e_{D}$$
(9.7)

where

$$J_{Y} \equiv \langle (D+D_{Y})(\nabla Y \cdot \nabla \xi) | \eta \rangle P(\eta) \rho_{\eta} - \frac{\partial \langle NY | \eta \rangle P(\eta) \rho_{\eta}}{\partial \eta}$$
(9.8)
$$e_{D} \equiv \operatorname{div} \left(\langle (D_{Y} - D)\rho \nabla Y | \eta \rangle P(\eta) \right) + \nabla^{2} \left(\langle D\rho Y | \eta \rangle P(\eta) \right) - \operatorname{div} \left(\langle Y \nabla (D\rho) | \eta \rangle P(\eta) \right)$$
(9.9)

and $N \equiv D(\nabla \xi)^2$. The differential diffusion effects are significant at moderate Reynolds numbers and the term e_D is retained in Eq.(9.7). We use the term "moderate" to emphasize that we do not consider the limit Re $\rightarrow\infty$ but, practically, the Reynolds number can be as large as in real combustors. The first term of the representation of e_D in Eq.(9.9) can be

expected to be most significant.

Considering the differential diffusion effects we still can apply the CMC approximation (3.10) to J_Y and we still should require that Q=Y=a, W=0, a=const is the solution of Eq.(9.7) but the solution $Y=\xi$, W=0 is not valid if $D\neq D_Y$ in Eqs.(9.1) and (9.2). Hence, as in Sec.3.1.3, we can find the coefficient A in Eq.(3.10) but the coefficient B is not determined. The approximation for J_Y takes the form

$$J_{\rm Y} = - \frac{\partial \langle N|\eta \rangle P(\eta)\rho_{\eta}}{\partial \eta} Q + B^{\circ} \langle N|\eta \rangle P(\eta)\rho_{\eta} \frac{\partial Q}{\partial \eta}$$
(9.10)

where $B^{\circ} \equiv B/(\langle N | \eta \rangle P(\eta) \rho_{\eta})$ is unknown.

The unclosed equation can also be derived by the decomposition method. Details of the derivation are similar to those given in Section 3.2 and may be found in Bilger¹². The resulting equation is

$$\rho_{\eta} \frac{\partial Q}{\partial t} + \rho_{\eta} < \mathbf{v} | \eta > \nabla Q - \rho_{\eta} \frac{D_{Y}}{D} < N | \eta > \frac{\partial^{2} Q}{\partial \eta^{2}} = \rho_{\eta} < W | \eta > + e_{Q} + e_{Y}$$
(9.11)

where

$$e_{Q} \equiv \langle \operatorname{div}(\rho D_{Y} \nabla Q) + \rho D_{Y} \nabla \xi \cdot \nabla \frac{\partial Q}{\partial \eta} | \eta \rangle + \left(\frac{D_{Y}}{D} - I \right) \langle \operatorname{div}(\rho D \nabla \xi) | \eta \rangle \frac{\partial Q}{\partial \eta}$$
(9.12)

and

$$e_{\rm Y} \equiv - \langle \rho \frac{\partial Y''}{\partial t} + \rho \mathbf{v} \cdot \nabla Y'' - \operatorname{div} \left(D_{\rm Y} \rho \nabla Y'' \right) |\eta\rangle$$
(9.13)

In the above the ratio $D_{\rm Y}/D$ has been assumed constant even though the diffusivities are varying. These equations can be compared with Eqs (3.22) - (3.24) for the equal diffusivity case. It is seen that $D_{\rm y}$ replaces D in some places and that a new term appears in the definition of $e_{\rm Q}$, involving the difference in the diffusivities

$$e_{Q3} \equiv \rho_{\eta} \left(\frac{D_{Y}}{D} - l \right) M_{\eta} \frac{\partial Q}{\partial \eta}$$
(9.14)

where

$$M_{\eta} \equiv \frac{1}{\rho_{\eta}} \langle div(\rho D \nabla \xi) | \eta \rangle$$
(9.15)

is the conditional diffusion and is related to the conditional dissipation N_{η} by

$$M_{\eta}P_{\eta}\rho_{\eta} = \frac{\partial N_{\eta}P_{\eta}\rho_{\eta}}{\partial \eta}$$
(9.16)

as can be seen from Eq. (2.49). It is seen that M_{η} remains finite at large Reynolds numbers and hence e_{Q3} will also remain finite. It is the source term that generates differential diffusion. At moderate and high Reynolds number the other terms in e_Q are negligible, as before, and Eq. (9.11) may be written

$$\rho_{\eta} \frac{\partial Q}{\partial t} + \rho_{\eta} \langle \mathbf{v} | \eta \rangle \cdot \nabla Q = \rho_{\eta} \langle W | \eta \rangle + \rho_{\eta} \frac{D_{Y}}{D} N_{\eta} \frac{\partial^{2} Q}{\partial \eta^{2}} + \rho_{\eta} \left(\frac{D_{Y}}{D} - I \right) M_{\eta} \frac{\partial Q}{\partial \eta} + e_{Y} \qquad (9.17)$$

It is apparent that, if differential diffusion effects are to tend to zero at high Reynolds number, the term $e_{\rm Y}$ must be such as to counteract the effects of $e_{\rm Q3}$ at high Reynolds number. Closure for $e_{\rm Q3}$ is first examined for nonreacting homogeneous flow.

9.1.2. Closure for nonreacting homogeneous flow

Kronenburg and Bilger⁶⁶ consider the mixing of two nonreactive scalars with mass fractions, normalized by their maximum values, Y_A and Y_B , and initial conditions such that they are essentially separated so that while $Y_A + Y_B = 1$ everywhere, $Y_A \approx 1$, $Y_B \approx 0$ or $Y_A \approx 0$, $Y_B \approx 1$ over most of the field. The mixture fraction, ξ , and differential diffusion, z, are defined

$$\xi \equiv (1 + Y_{\rm A} - Y_{\rm B})/2 \tag{9.18}$$

$$z \equiv (1 - Y_{\rm A} - Y_{\rm B})/2 \tag{9.19}$$

For species diffusivities D_A and D_B the balance equations for ξ and z in uniform property flow are

$$\frac{\partial \xi}{\partial t} + \left(\mathbf{v} \cdot \nabla \xi\right) - D_{\xi} \nabla^2 \xi = -d \nabla^2 z \tag{9.20}$$

$$\frac{\partial z}{\partial t} + \left(\mathbf{v} \cdot \nabla z\right) - D_{\xi} \nabla^2 z = -d \nabla^2 \xi$$
(9.21)

where $D_{\xi} \equiv (D_A + D_B)/2$ and $d \equiv (D_A - D_B)/2$. It is seen that the mixture fraction has a differential diffusion source term, but this is found to be of little significance.

The conditional average Q_z of z, $Q_z \equiv \langle z | \eta \rangle$ has a balance equation similar to Eq. (9.17), and for homogeneous turbulence and scalars this may be written

$$\frac{\partial Q_z}{\partial t} = \frac{D_Y}{D} \langle N | \eta \rangle \frac{\partial^2 Q_z}{\partial \eta^2} - d \langle \nabla^2 \xi | \eta \rangle + d \langle \nabla^2 z | \eta \rangle \frac{\partial Q_z}{\partial \eta} + e_Y$$
(9.22)

In this equation $-d < \nabla^2 \xi |\eta >$ is the source term for differential diffusion corresponding to e_{Q3} in Eq.(9.14). It will remain significant at high Reynolds numbers. A model for e_Y is required which will give the right behaviour for Q_z .

Kronenburg and Bilger⁶⁶ carried out direct numerical simulations of this problem using a pseudo-spectral code. A typical result for z as a scatterplot against ξ and Q_z against η is shown in Fig. 45 In this simulation $Sc_A = I$ and $Sc_B = 0.5$. It is seen that while the conditional variance of z is quite large, Q_z is of the same order of magnitude, being positive for low values of η and negative at high values. The shape of $Q_z(z)$ mirrors that of M_η which has an "N"-like shape for this flow. (It is noted that for homogeneous turbulence with a Gaussian mixture fraction pdf, N_η is independent of η so that $N_\eta = \langle N \rangle^7$; and from Eq.(9.16) it is seen that M_η will be a straight line going through zero at $\eta = \langle \xi \rangle$ and having a slope of $-\langle N \rangle/\langle (\xi'')^2 \rangle$.)

From the DNS data it was found that $e_{\rm Y}$ has the shape of $Q_{\rm z}$ and a rate proportional to the reciprocal of the Kolmogorov time scale, $\tau_{\rm K}$. The model recommended for $e_{\rm Y}$ is

$$e_{\rm Y} = -0.4 \left| \frac{D_{\rm A} - D_{\rm B}}{D_{\xi}} \right|^{1/2} \frac{Q_{\rm z}}{\tau_{\rm K}}$$
(9.23)

With this model for $e_{\rm Y}$ Eq.(9.22) gives good predictions of $Q_{\rm z}$ over the range of DNS investigated including variations in $\langle \xi \rangle$ and $Sc_{\rm A}$, $Sc_{\rm B}$. The model also predicts that $Q_{\rm z} \rightarrow 0$ as the Reynolds number becomes large. Nilsen & Kosaly¹⁰⁰ have carried out DNS studies at higher Reynolds numbers and confirm this modelling for $e_{\rm Y}$.

Yeung & Pope¹⁴⁷ studied differential diffusion in decaying isotropic turbulence using DNS. They report results for the correlation coefficient, ρ_{AB} , for the two species. This is found to decay towards zero. As shown by Kronenburg and Bilger⁶⁶, the correlation coefficient contains no information about the conditional average differential diffusion and

$$|\rho_{AB}| \approx 1 - 2 \Big(\langle (z'')^2 \rangle / \langle (\xi')^2 \rangle \Big)^2$$
 (9.24)

where $z'' \equiv z - Q_z$. The correlation coefficient is thus more a measure of the conditional variance. We note that if the conditional variance is very small, A and B could be nearly perfectly correlated but still show a large amount of differential diffusion with $Q_z \neq 0$. For closure of the chemical source terms it is apparent that the conditional average of the differential diffusion is the quantity of primary importance.

9.1.3. Closure for reacting flow

Nilsen & Kosaly¹⁰¹ have used DNS to study differential diffusion in homogeneous turbulence with a single step irreversible reaction with differing Schmidt numbers for the reactants. They present a model for the differential diffusion which neglects the contribution from e_Y discussed in the previous section, but they require the modelling of a conserved scalar with an intermediate Schmidt number. Very recent results ⁶⁷,⁷⁰ at Sydney University indicate that good predictions are obtained if the above model for e_Y is used for those species that have a non-unity Lewis number.

9.2. CMC with Multiple Conditions

Previously, we mainly considered combustion occurring in a turbulent flow resulted from mixing of two streams: stream of fuel and and stream of oxidizer. This two-stream mixing process is characterized by one scalar value - the mixture fraction. There are, however, some practical cases when one mixture fraction variable is not sufficient. These cases are referred to as cases with multi-stream mixing. For example, mixing of three separate streams of fuel, oxidizer and an inert gas should be characterized by two independent conserved scalars. Some features of three-stream mixing are considered by Juneja and Pope⁴⁸. More complicated mixing schemes may need more than two scalars. In terms of the conditional methods advanced in the present review, this requires consideration of conditional expectations with several different conditioning variables.

The CMC equations can be generalized for conditional expectations with multiple conditions. In this section, Q is defined as $Q \equiv \langle Y | \xi_1 = \eta_1, ..., \xi_n = \eta_n \rangle \equiv \langle Y | \underline{\eta} \rangle$ where $\underline{\eta} \equiv (\eta_1, ..., \eta_n)$. The conserved scalars ξ_i and the reactive scalar Y are governed by the equations

$$\rho \frac{\partial \xi_{i}}{\partial t} + \rho \left(\mathbf{v} \cdot \nabla \xi_{i} \right) - \operatorname{div} \left(D \rho \nabla \xi_{i} \right) = 0; \quad i=1,...,n$$

$$\rho \frac{\partial Y}{\partial t} + \rho \left(\mathbf{v} \cdot \nabla Y \right) - \operatorname{div} \left(D \rho \nabla Y \right) = W \rho$$
(9.26)

The derivations of the CMC equations with multiple conditions are similar to the derivations of the CMC with one-variable conditions considered in Sec.3.1 and Sec.3.2. We assume here that the conserved scalars do not coincide with each other. Practically, such a situation, when we need more than one conserved scalar, can appear if combustion occurs in a turbulent flow formed by three or more streams. The example is given by an oxidizer jet and a fuel jet injected into air. In this case, the mixing characteristics are determined by two conserved scalars. The main terms of the CMC equation which are related to double-conditioning ($\xi_1 = \eta_1$ and $\xi_2=\eta_2$) were obtained by Bilger¹¹, except that he considered one of the scalars to be reactive. In that case the reactive scalar was formulated as a progress variable and the application envisaged was for a conventional 2-stream mixing problem with local extinction or ignition events giving rise to large fluctuations about the conditional mean. Little further progress has been made on this problem and it will not be considered in detail here.

The relationship of the conditional expectation Q, the joint pdf $P(Z,\underline{\eta})$ (where Z is the sample space variable for Y) and the pdf $P(\underline{\eta})$ is given by Eq.(2.15)

$$Q P(\underline{\eta}) \equiv \langle Y | \underline{\xi} = \underline{\eta} \rangle P(\underline{\eta}) = \int_{-\infty}^{+\infty} Z P(Z, \underline{\eta}) dZ$$
(9.27)

We utilize the joint pdf equation (2.48) which is valid for the case of large Re numbers and consider this equation as the equation for $P(Z,\eta)$. We put in Eq.(2.48) $\eta_i = Z_i, W_i = 0$ (i=1,...,n); $Z = Z_{n+1}, W = W_{n+1}$. This equation is multiplied by Z and integrated over all Z. The terms which involve $\partial/\partial Z$ are integrated by parts. The result of the integration is

$$\frac{\partial}{\partial t} \left(QP(\underline{\eta}) \rho_{\eta} \right) + \operatorname{div} \left(\langle \mathbf{v}Y | \underline{\eta} \rangle P(\underline{\eta}) \rho_{\eta} \right) = \langle W | \underline{\eta} \rangle P(\underline{\eta}) \rho_{\eta} + \frac{\partial J_{i}}{\partial \eta_{i}}$$
(9.28)

$$J_{i} \equiv 2 \langle D(\nabla Y \cdot \nabla \xi_{i}) | \underline{\eta} \rangle P(\underline{\eta}) \rho_{\eta} - \frac{\partial}{\partial \eta_{j}} \left(\langle N_{ij}Y | \underline{\eta} \rangle P(\underline{\eta}) \rho_{\eta} \right)$$
(9.29)

where $\rho_{\eta} \equiv \langle \rho | \underline{\eta} \rangle$, $N_{ij} \equiv D(\nabla \xi_i \cdot \nabla \xi_j)$ is the dissipation tensor and i,j=1,...,nhere and further on. A sum is taken over repeated indices. Equation (3.4) has n+4 independent variables: t, x_1, x_2, x_3 and $\eta_1,...,\eta_n$.

The flux of a reactive scalar in conserved scalar space, J_i , is approximated by the diffusion approximation

$$J_{\rm i} = A_{\rm i}Q + B_{\rm ij} \frac{\partial Q}{\partial \eta_{\rm j}}$$
(9.30)

The arguments supporting this approximation is similar to the arguments for Eq.(3.10) given in Sec.3. We do not assume any particular form of the coefficients A_i and B_{ij} but they are fully determined by a the following constraint: if W=0 and a=const, $b_k=\text{const}$ then $Y=a+b_k\xi_k$ is a solution of Eq.(9.26) for any arbitrary velocity field. According to Eq.(2.17) this solution corresponds to $Q=a+b_k\eta_k$. The substitution of $Y=a+b_k\xi_k$ into Eq.(9.29) yields

$$J_{i} = -\left(a + b_{k}\eta_{k}\right)\frac{\partial}{\partial\eta_{j}}\left(\langle N_{ij}|\underline{\eta}\rangle P(\underline{\eta})\rho_{\eta}\right) + b_{j}\langle N_{ij}|\underline{\eta}\rangle P(\underline{\eta})\rho_{\eta}$$
(9.31)

while the substitution of $Q=a+b_k\eta_k$ into Eq.(9.30) yields

$$J_{i} = (a+b_{k}\eta_{k})A_{i} + b_{j}B_{ij}$$

$$(9.32)$$

Since Eqs.(9.31) and (9.32) are valid for any arbitrary constants a and b_k , the coefficients A_i and B_{ij} must be determined by the corresponding terms in Eq.(9.31). The closure for J_i takes the form

$$J_{i} = -Q \frac{\partial}{\partial \eta_{j}} \left(\langle N_{ij} | \underline{\eta} \rangle P(\underline{\eta}) \rho_{\eta} \right) + \langle N_{ij} | \underline{\eta} \rangle P(\underline{\eta}) \rho_{\eta} \frac{\partial Q}{\partial \eta_{j}}$$
(9.33)

Equation (9.28) takes the form

$$\frac{\partial}{\partial t} \left(QP(\underline{\eta})\rho_{\eta} \right) + \operatorname{div} \left(\langle \mathbf{v}Y | \underline{\eta} \rangle P(\underline{\eta})\rho_{\eta} \right) = \langle W | \underline{\eta} \rangle P(\underline{\eta})\rho_{\eta} - Q \frac{\partial^{2}}{\partial \eta_{i} \partial \eta_{j}} \left(\langle N_{ij} | \underline{\eta} \rangle P(\underline{\eta})\rho_{\eta} \right) + \langle N_{ij} | \underline{\eta} \rangle P(\underline{\eta})\rho_{\eta} \frac{\partial^{2}Q}{\partial \eta_{i} \partial \eta_{j}}$$
(9.34)

Equation (9.34) is the CMC equation with multiple conditions. The alternative form of this equation

$$\frac{\partial Q}{\partial t} + \langle \mathbf{v} | \underline{\eta} \rangle \nabla Q + \frac{\operatorname{div} \left(\langle \mathbf{v}'' Y'' | \underline{\eta} \rangle P(\underline{\eta}) \rho_{\eta} \right)}{P(\underline{\eta}) \rho_{\eta}} - \langle N_{ij} | \underline{\eta} \rangle \frac{\partial^2 Q}{\partial \eta_i \partial \eta_j} = \langle W | \underline{\eta} \rangle (9.35)$$

can be obtained by using the equation for the joint pdf $P(\underline{\eta})$. The double-prime denotes the fluctuations about the conditional mean $(\cdot)'' \equiv (\cdot) - \langle \cdot | \eta \rangle$.

9.2.2. The decomposition method

In this section we consider the alternative way to derive the equation for $Q(\underline{\eta}, \mathbf{x}, t) \equiv \langle Y(\mathbf{x}, t) | \underline{\eta} \rangle$. This derivation is based on the decomposition

$$Y(\mathbf{x},t) = Q(\underline{\xi}(\mathbf{x},t),\mathbf{x},t) + Y''(\mathbf{x},t)$$
(9.36)

The differentiation rules for Eq.(9.36) are given by

$$\frac{\partial Y}{\partial t} = \frac{\partial Q}{\partial t} + \frac{\partial Q}{\partial \eta_{i}} \frac{\partial \xi_{i}}{\partial t} + \frac{\partial Y''}{\partial t}$$
(9.37)

$$\nabla Y = \nabla Q + \frac{\partial Q}{\partial \eta_{i}} \nabla \xi_{i} + \nabla Y''$$
(9.38)

$$div(\rho D\nabla Y) = div(\rho D\nabla Q) + \frac{\partial Q}{\partial \eta_{i}} div(\rho D\nabla \xi_{i}) + \rho D(\nabla \xi_{i} \cdot \nabla \xi_{j}) \frac{\partial^{2} Q}{\partial \eta_{i} \partial \eta_{j}} + \rho D\nabla \xi_{i} \cdot \nabla \frac{\partial Q}{\partial \eta_{i}} + div(\rho D\nabla Y'')$$
(9.39)

We substitute Eqs.(9.37) -(9.39) into Eq.(9.26) and use Eq.(9.25). The result is conditionally averaged (with the condition $\underline{\xi}(\mathbf{x},t)=\underline{\eta}$)

$$\rho_{\eta} \frac{\partial Q}{\partial t} + \rho_{\eta} < \mathbf{v} | \underline{\eta} > \nabla Q - \rho_{\eta} < N_{ij} | \underline{\eta} > \frac{\partial^2 Q}{\partial \eta_i \partial \eta_j} = \rho_{\eta} < W | \underline{\eta} > + e_{Q} + e_{Y} \quad (9.40)$$

where

$$e_{\rm Q} \equiv \langle \operatorname{div}(\rho D \nabla Q) + \rho D \nabla \xi_{\rm i} \cdot \nabla \frac{\partial Q}{\partial \eta_{\rm i}} | \underline{\xi}(\mathbf{x}, t) = \underline{\eta} \rangle$$
(9.41)

$$e_{\mathbf{Y}} \equiv - \langle \rho \frac{\partial Y''}{\partial t} + \rho \left(\mathbf{v} \cdot \nabla Y'' \right) - \operatorname{div} \left(D \rho \nabla Y'' \right) \mid \underline{\xi}(\mathbf{x}, t) = \underline{\eta} \rangle$$
(9.42)

This equation is unclosed. In order to close this equations we consider the identity

$$-\int e_{Y}P(\underline{\eta})d\eta_{1}...d\eta_{n} = \langle \frac{\partial Y''\rho}{\partial t} + \operatorname{div}\left(\rho \mathbf{v}Y''\right) - \operatorname{div}\left(D\rho\nabla Y''\right) \rangle =$$

$$\frac{\partial \langle Y''\rho \rangle}{\partial t} + \operatorname{div}\left(\langle\rho \mathbf{v}Y''\rangle\right) - \operatorname{div}\left(\langle D\rho\nabla Y''\rangle\right) = \operatorname{div}\left(\langle\rho \mathbf{v}''Y''\rangle\right) =$$

$$= \int \operatorname{div}\left(\rho_{\eta}\langle\mathbf{v}''Y''|\underline{\eta}\rangle P(\underline{\eta})\right)d\eta_{1}...d\eta_{n} \qquad (9.43)$$

which derivation is similar to derivation of Eq.(3.27). The hypothesis which is utilized here is

$$e_{\mathbf{Y}}P(\underline{\mathbf{\eta}}) = -\operatorname{div}\left(\rho_{\mathbf{\eta}} < \mathbf{v}'' \mathbf{Y}'' | \underline{\mathbf{\eta}} > P(\underline{\mathbf{\eta}})\right)$$
(9.44)

is the generalization of Eq.(3.28) and does not need any specific comments. The term e_Q is small for large Reynolds numbers and can be neglected (see Sec.2.3.3). Equation (9.40) takes the form

$$\frac{\partial Q}{\partial t} + \langle \mathbf{v} | \underline{\eta} \rangle \cdot \nabla Q + \frac{\operatorname{div} \left(\langle \mathbf{v}'' Y'' | \underline{\eta} \rangle P(\underline{\eta}) \rho_{\eta} \right)}{P(\underline{\eta}) \rho_{\eta}} - \langle N_{ij} | \underline{\eta} \rangle \frac{\partial^2 Q}{\partial \eta_i \partial \eta_j} = \langle W | \underline{\eta} \rangle (9.45)$$

9.2.3. Discussion

It is easy to see that both methods of derivation of the CMC equation with multiple conditions yield identical equations (9.35) and (9.45). This supports the validity of these equations. Practically, solving the CMC equation with multiple conditions is more complicated than solving the standard CMC equation. The conditional dissipation tensor $\langle N_{ij} | \underline{\eta} \rangle$ needs further approximations which must be consistent with the equation for $P(\underline{\eta})$. The large number of independent variables represent the additional difficulty. Equations (9.35) and (9.45) must be solved in a domain which has a configuration that is more complicated than the interval $\eta_{min} \leq \eta \leq \eta_{max}$ for one-variable CMC. The paper by Juneja and Pope⁴⁸ analyzes general properties of the two-scalars pdf $P(\eta_1, \eta_2)$ and presents some interesting DNS results for $P(\eta_1, \eta_2)$.

9.3. CMC and Spray Combustion

In this section we consider a spray of droplets in a turbulent flow, the effects of the evaporation of the droplets and the mixing and combustion of the vapour. Our primary interest is in the formulation of the CMC method taking into account the significant variations of the concentrations of evaporated species in the inter-droplet space. In this consideration we follow Klimenko and Bilger⁶¹. Existing methods (see Faeth^{35,36}, Bachalo¹) treat the problem of the mixing of the spray as a whole arising from the penetration of the spray into the surrounding air and from the mixing processes associated with the large scale turbulence. The droplets themselves and the inter-droplet distances are small compared with the integral scale of the turbulence. Evaporation from the droplets

causes a lot of fine scale structure in the scalar field and this can be important in the combustion process. Thus, if the spray is dilute it can affect the structure and pollutant formation in the reaction zone of the flame; and if the spray is less dilute it will affect formation of soot and other pollutants such as PCAH (poly-cyclic aromatic hydrocarbons) and NO_x from fuel bound nitrogen.

9.3.1. Basics of droplet evaporation and combustion

Williams¹⁴⁴ treats the theory of quiescent burning droplets. This can be recast in terms of mixture fraction φ introduced by Bilger¹⁴. The conditions at the surface which separates liquid and gas phases for the sensible enthalpy *h*, for the mass fraction *Y* of the evaporating substance and for mixture fraction are given by

$$\dot{m}(Y_{\rm d}-Y_{\rm s}) = -\left(\rho D \frac{\partial Y}{\partial n_{\rm s}}\right)_{\rm s}$$
(9.46)

$$\dot{m}(h_{\rm d}-h_{\rm s}) = -\left(\rho D \frac{\partial h}{\partial n_{\rm s}}\right)_{\rm s} \tag{9.47}$$

$$\dot{m}(f_{\rm d}-f_{\rm s}) = -\left(\rho D \frac{\partial \varphi}{\partial n_{\rm s}}\right)_{\rm s} \tag{9.48}$$

where $m \equiv \rho v_n$ is the local mass flux, \mathbf{n}_s is the outer normal vector at the droplet surface, subscript "d" corresponds to conditions inside the droplets and subscript "s" denotes values taken at the "gas side" of the surface ($\varphi = f_d$ inside the droplets and $\varphi = f_s$ at the droplet surface). Mixture fraction φ is defined as the mass fraction of droplet material such that it is conserved under chemical reaction and applies in the gas phase between the droplets (in most of the cases φ is introduced so that $f_d=1$ and $f_0=0$; where f_0 is the value of φ in the inter-droplet space before evaporation starts). The new notation " ϕ " is used to distinguish the mixture fraction from the conserved scalar ξ which is not affected by the evaporation processes. The scalar transport equations determine evolution of scalars Y, h and φ in the gas phase but in the case of scalar φ this equation has zero source term. Differential diffusion is not considered here and the diffusion coefficients, D, are assumed to be the same for all components. Concentration $Y_s = Y_s(h_s)$ is the saturation concentration

corresponding to h_s . In many cases, not only the boundary conditions for *Y*, *h* and φ but also the scalar fields themselves have a certain degree of similarity.

We denote by $\langle \cdot \rangle_g$ the value of (·) averaged over the gas phase for distances small compared to the integral length scale of the turbulence and large compared with the inter-droplet distance, $r_c = c^{-1/3}$ with c the number density of the droplets. At these scales we define the overall mixture fraction (including liquid and gas) of the droplet material as ξ , and the mass fraction of liquid droplets as α_d so that $\xi = \alpha_d f_d + \alpha_g f_g$ where $\alpha_{g}=1-\alpha_{d}, f_{g}\equiv\langle\phi\rho\rangle_{g}/\rho_{g}, \rho_{g}\equiv\langle\rho\rangle_{g}$. The overall density is given by $\rho_{+}=\alpha_{d}^{\circ}\rho_{d}+\alpha_{g}^{\circ}\rho_{g}$ where $\alpha_{d}^{\circ}\equiv4\pi r_{d}^{3}c/3$ is the specific volume occupied by the droplets, $\alpha_{g}^{\circ}=1-\alpha_{d}^{\circ}, \alpha_{d}=\alpha_{d}^{\circ}\rho_{d}/\rho_{+}$ and r_{d} is the droplet radius. Existing methods³⁵ can be used to find the first and second moments of $f_{\rm g}$ and $\alpha_{\rm d}$ in the spray field. They will determine the gross burning characteristics of the spray. The CMC equations with condition $\xi = \eta$ can be also used to find the overall evaporation characteristics. The overall mixture fraction ξ is a conserved scalar whose fluctuations are determined by turbulent macro-transport and ξ is not directly affected by the evaporation processes. Our prime interest here is, however, different: application of the CMC equations to the inter-droplet combustion processes.

The characteristics of the inter-droplet field φ are determined by the evaporation rate. The mass evaporation rate $J_{\rm m}$ of a single droplet is given by $J_{\rm m}=4\pi r_{\rm d}^2 \dot{m}$. In order to close the problem $J_{\rm m}$ should be expressed in terms of the characteristics of the inter-droplet field. The mass evaporation rate $J_{\rm m}$ of a single droplet is determined by the Nusselt number¹⁴

Nu =
$$\frac{2r_{\rm d}\dot{m}}{\rho_1 D_1 \ln(1+B)} = \frac{J_{\rm m}}{2\pi r_{\rm d}\rho_1 D_1 \ln(1+B)}; \quad B = \frac{f_{\rm s}-f_2}{f_{\rm d}-f_{\rm s}},$$
 (9.49)

where f_2 is a constant which is discussed in Sec.9.3.3. Index '1' is related to the values taken in the inter-droplet space, at sufficient distance from each individual droplet. If droplets are non-inertial and density and diffusivity are constants then Nu=2. For inertial droplets the Nu number can be estimated as Nu=2(1+0.3Re_d^{1/2}).

9.3.2. Formulation of the CMC model

Fluctuations of the reactive species can arise from two main causes: 1) turbulent macro-transport and 2) inter-droplet micro-processes. In general, averages with double conditioning $Q \equiv \langle Y | \xi = \eta, \varphi = f \rangle$ should be used here. The variations of ξ are related to the turbulent macro-transport and the variations of ϕ are related to inter-droplet micro-processes. We are most interested, however, in the description of the inter-droplet processes. In order to exclude the macro-fluctuations from our consideration we investigate the case of homogeneous turbulence with ξ =const. The conditional expectation Q is introduced here as $Q_i \equiv \langle Y_i | \phi = f \rangle \equiv \langle Y_i | f \rangle$ where index "i" corresponds to different species.

The CMC equations in homogeneous turbulence can be written here as usual (see Eq.(4.30))

$$\frac{\partial Q_{i}}{\partial t} = \langle N|f \rangle \frac{\partial^{2} Q_{i}}{\partial f^{2}} + \langle W_{i}|f \rangle$$
(9.50)

$$\frac{\partial Q_{\rm h}}{\partial t} = \langle N | f \rangle \frac{\partial^2 Q_{\rm h}}{\partial f^2} + \langle W_{\rm h} | f \rangle$$
(9.51)

where $Q_i \equiv \langle Y_i | f \rangle$, $Q_h \equiv \langle h | f \rangle$, $N \equiv D(\nabla \varphi)^2$, W_i and W_h are the chemical source terms which do not include the mass exchange due to evaporation. The boundary conditions for Eqs.(9.50) and (9.51) can be written as

$$f=f_{\rm s}: \quad \frac{(Y_{\rm i})_{\rm d}-Q_{\rm i}}{f_{\rm d}-f_{\rm s}} = \frac{\partial Q_{\rm i}}{\partial f}, \quad \frac{q}{f_{\rm d}-f_{\rm s}} = -\frac{\partial Q_{\rm h}}{\partial f}, \quad Q_{\rm s}=Y_{\rm s}(h_{\rm s})$$
(9.52)

$$f=f_0: \quad Q_i=(Y_i)_0, \quad Q_h=h_0$$
(9.53)

where q is the evaporation enthalpy $q \equiv h_s \cdot h_d$; $(Y_i)_d$ specifies conditions inside the droplets: $(Y_i)_d = Y_d = I$ for the evaporating substance and $(Y_i)_d = 0$ for others. The index "0" in Eq.(9.53) corresponds to the conditions in the inter-droplet space before evaporation and reactions start. Boundary conditions (9.52) are obtained from Eqs.(9.46)-(9.48). In general, f_s is unknown function of time so Eqs.(9.50) and (9.51) are to be solved in the time-dependent domain $f_s(t) \ge f \ge f_0$. Function $f_s(t)$ is determined by the

saturation condition $Q_s=Y_s(h_s)$ for evaporating substance.

As usual, equations (9.50) and (9.51) are to be solved in conjunction with the pdf equation

$$\frac{\partial P(f)\rho_{\rm f}}{\partial t} + \frac{\partial^2 \langle N|f \rangle P(f)\rho_{\rm f}}{\partial f^2} = 0$$
(9.54)

where $P(f)\rho_f = \alpha_d^{\circ}\delta(f-f_d)\rho_d + \alpha_g^{\circ}P_g(f)\rho_f$, $\rho_f \equiv \langle \rho | f \rangle$ and P(f) is the pdf of φ . The Delta-function in the pdf P(f) is related to the conditions inside droplets. Equation (9.54) is valid only in the gas phase $f_s(t) > f \ge f_0$ where the pdf is smooth $P(f) = \alpha_g^{\circ}P_g(f)$. Modeling of $\langle N | f \rangle$ is required to close Eqs. (9.50) and (9.51). If P(f) is given, $\langle N | f \rangle$ can be calculated from (9.54) with proper boundary conditions for $\langle N | f \rangle P(f)$ (see Sec.2.4). The pdf P(f) and the dissipation $\langle N | f \rangle$ have certain distinctive features which require special consideration. Modeling of P_f and $\langle N | f \rangle$ is considered in the next section.

9.3.3. The mixture fraction pdf and dissipation in sprays

In this sections we consider specific features of $N_f \equiv \langle D(\nabla \phi)^2 | \phi = f \rangle$ and $P_f = P(f)$ which are related to the spray micro-structure. The new notations $N_{\rm f}$ and $P_{\rm f}$ are used here for the conditional dissipation and the pdf of the mixture fraction φ . The mixture fraction φ was introduced in Sec.9.3 as the mass fraction of the droplet material such that it is conserved under chemical reaction. Here, we consider the micro-structure of the mixture fraction field which is related to discrete nature of the evaporation sources. Various types of spray micro-structure can be identified. These are considered in two main classes: those in which the droplets are essentially non-inertial with no motion relative to the fluid; and those in which the droplets are inertial and move relative to the surrounding fluid. Several regimes exist within each of these classes. Information on the pdfs and scalar dissipation is obtained using dimensional analysis, asymptotic matching methods and existing analytical solutions.

Figure 46 shows schematically two classes of the micro-structure of

the spray. In Fig. 46(a) the inertial time scale for the droplets τ_d (equations determining τ_d are discussed later) is small compared with the Kolmogorov time scale $\tau_{\rm K} = (\nu/\epsilon_t)^{1/2}$, where ϵ_t is averaged dissipation of energy and ν is kinematic viscosity coefficient. The droplets have lost their initial momentum from the spray nozzle and now are moving with essentially no relative motion to the gas phase. Evaporation from the droplets causes the scalar field to be spherically symmetric close to the droplets. In Fig. 46(b) the inertia of the droplets causes motion relative to the gas phase as the gas phase accelerates due to fine scale turbulent motion. This will occur¹ when $\tau_d > \tau_K$. The wakes of the droplets cause a lot of fine scale structure in the scalar field.

In the absence of the discrete sources of the evaporating gas - the droplets - the small scale structure of the turbulent scalar field is determined by two parameters: scalar dissipation and viscosity (diffusivity). In the theory of small-scale turbulence, the required characteristics of the scalar field are determined by the analysis of the dimensions involved (that is by applying the Π -theorem). In the presence of the evaporating droplets the structure of the turbulent scalar field is more complicated and may involve several different asymptotic zones. The basic principles of the analysis remain the same: finding the determining parameters for each zone and then using the Π -theorem to determine the shape of the functions $\langle N|f \rangle$ and P(f) that we are interested in. It should not be forgotten that $\langle N|f \rangle$ and P(f) are also linked by Eq.(9.54).

i) Non-inertial Droplets

Here, we consider the droplets which do not have any significant velocity relatively to the continuous phase (Fig. 46(a)). For a single droplet under stagnant conditions the solution f(r) is well-known¹⁴⁴. The function f(r) determines also the pdf P_f and the conditional scalar dissipation N_f (Section A in Tab. 3). At distances of a few times larger than r_d the Stefan flow can be neglected and the scalar f transport is determined by diffusion. Since r_c is normally much smaller than turbulence integral macroscale L, we apply here the logic of Kolmogorov⁶³ theory of small-scale turbulence. The diffusion asymptote (Section A+ in Tab. 3) has two determining parameters for $f_c f_2$: diffusion coefficient D_1 and the diffusion component of the flux $J_1 \equiv J_m (f_d - f_2)/\rho_1$ (where $J_m = 4\pi r_d^2 \dot{m}$ is the mass evaporation rate of the droplet; f_2 specifies effective conditions for f at a large distance from the droplet; $D=D_1$, $\rho=\rho_1$ for $r \gg r_d$; effects

related to $Sc \neq 1$ or $Pr \neq 1$ are not considered). The determining parameters are framed in the table. If droplet is surrounded by a turbulent flow, the dissipation of turbulent energy ε_t should also be included into the list of determining parameters for scales of order of or larger than Kolmogorov length scale $\ell_{\rm K}$ since the scalar field f is affected by turbulence and $\epsilon_{\rm t}$ is a determining parameter for the small scale turbulence. (We assume that the Stefan flows do not destroy small-scale structure of turbulence but the global expansion due to the Stefan flows must be, of course, taken into consideration. Normally, $\ell_{\rm K}$ is at least few times larger than $r_{\rm d}$.) Equations for P_f and N_f predicted by the Π -theorem are included in Sec.B, Tab. 3. The Kolmogorov Scale Zone is asymptotically matched with the Near Zone for $r_d \ll r \ll \ell_K$. This specifies constant $(2\pi)^2$ in Sec.B-, Tab. 3. Constant A_1 in Sec.B+ remains unknown.

At the distances ~ $r_c \equiv c^{-1/3}$ (where c is the number density of the droplets) the scalar field f is not determined by a single droplet. There are two cases case 1: $\ell_{\rm K} < r_{\rm c}$ and case 2: $\ell_{\rm K} > r_{\rm c}$ (strictly speaking $\ell_{\rm K} \ll r_{\rm c}$ and $\ell_{\rm K} \gg r_{\rm c}$) considered in Sec.C, Tab. 3. The determining parameters for equations in Sec.C are: the diffusion component of the collective evaporation rate $W_1 \equiv W_m (f_d - f_2) / \rho_1$ (where W_m is collective mass evaporation rate per unit volume); concentration c; dissipation of the turbulent energy ε_t (in the case 1) and diffusion coefficient D_1 (in the case 2). These parameters yield equations for value f° with the dimension of f. If droplets are different, then the values of J_1 are different for different groups of the droplets $cJ_1=W_1\beta$. The mean value of β over all droplets is unity. The pdfs considered in Sec.A,B are conditioned on J_1 . If all of the droplets are similar then $\beta \equiv l$ and $J_1 = W_1/c$. In general, it is plausible to estimate: $f' \equiv (\langle \varphi^2 \rangle_g - \langle \varphi \rangle_g^2)^{1/2} = \operatorname{const} f^\circ$, but we should note here that in some cases the asymptote of the pdf $P_{\rm f}$ has the tail with non-integrable second moment and f° determines the dispersion only for the core part of the pdf. The core parts of $P_{\rm f}$ and $N_{\rm f}$ (Sec.C, Tab. 3) are asymptotically matched with the equations of Sec.B (case 1) or with the equations of Sec.A (case 2). In the Inter-droplet Zone the density does not vary $\rho = \rho_1$ and we assume that $f_g \equiv \langle \varphi \rho \rangle_g = \langle \varphi \rangle_g$ ($\langle \varphi \rangle_g$ is determined by the core part of the pdf).

The shapes of the functions $P_{\rm K}^{\circ}$, $N_{\rm K}^{\circ}$, $P_{\rm c}^{\circ}$ and $N_{\rm c}^{\circ}$ are not determined by Π -theorem or by asymptotical matching, but we approximate $P_{\rm K}^{\circ}$ and $N_{\rm K}^{\circ}$ by assuming that B- equations are valid for $r \leq \ell_{\rm K}$, $f \geq f(\ell_{\rm K})$ and B+ equations are valid for $r \geq \ell_{\rm K}$, $f \leq f(\ell_{\rm K})$. This gives an approximate value of

 $A_1 \approx (4\pi)^{-2/7}$ (we assume here that $\phi_2 \approx 0$). The scalar field f in inter-droplet space is a superposition of the scalar fields generated at least by a few of the droplets located in the vicinity of the physical point under consideration. If sources J_1 of these droplets are approximately of similar intensity (or similar order of intensity) then, according to the central limit theorem, we expect the pdf P_f to be Gaussian. This is not valid for small distances $r \ll r_c$ when f is determined by one of the droplets. So we can assume that the pdf has a Gaussian core and an adjoined power function specified in Sec.C1- or Sec.C2-. The values of ϕ_1 and ϕ_2 are expected to be universal constants. Constant ϕ_1 is linked with the dispersion of the Gaussian core of the pdf.

ii) Inertial Droplets

If the characteristic relaxation time τ_d is greater than the Kolmogorov time scale τ_K , the droplets move relatively to the continuous phase¹. The heat and mass transport occur in the wake-like structures (Fig. 46(b)) which have determining parameters different from parameters considered in previous section.

For further estimations we consider the case when τ_d belongs to the inertial interval range (this is valid in most of the practical cases). The relative velocity of a droplet can be estimated¹⁴¹ as $u_d^2 = A_2 \varepsilon_t \tau_d$ (A₂=const). Let us assume first that τ_d is determined by the Stokes equation $\tau_d = \tau_{st} = 2r_d^2 \rho_d / (9\rho v)$. A droplet has a relative motion if $l < (\tau_d/\tau_K) \approx (r_d/\ell_K)^2 \rho_d/\rho$. The Reynolds number for the droplet motions is introduced as $\operatorname{Re}_{d} \equiv 2r_{d}u_{d}/v \approx (r_{d}/\ell_{K})^{2} (\rho_{d}/\rho)^{1/2}$. If $\operatorname{Re}_{d} \geq 1$ the Stokes equation overestimates τ_d and u_d . In this case τ_d should be corrected¹²⁶ $\tau_d = \tau_{st} (1+0.15 Re_d^{0.687})^{-1}$. After elementary transformations obtain we $Re_{st}=Re_d(1+0.15Re_d^{0.687})^{1/2}$ so the Reynolds number Re_d is a function of Re_{st} which is formally calculated from Re_{st} $\equiv 2r_d u_{st}/v$,

In the case of moving droplets it is more difficult to find analytical formulas for the Near Zone (Sec.A, Tab. 4), but the far asymptote of the Near Zone (Sec.A+, Tab. 4) is usually a laminar wake with the scalar *f* intensity $S_1 \equiv J_1/u_d$ which is then entrained and expanded by turbulence. Functions P_f and N_f are found by integrating over the wake sectors with given value of *f*. The estimations and matching procedures for other zones are similar to ones carried out in the previous section, except that f° can not be determined from dimension analysis and it is found by matching with zones B+ or A+. We note also that characteristic

droplet relaxation length $r_u \equiv \tau_d u_d \approx \tau_{st} u_{st}$ is normally much larger than r_c for droplets with $\tau_d / \tau_K \gg I$ since $r_u / r_c \approx (\tau_d / \tau_K) ((\rho_d / \rho)^{0.5} c_m)^{1/3} \sim \tau_d / \tau_K$ $(c_m \text{ is mass concentration of the dispersed phase and <math>((\rho_d / \rho)^{0.5} c_m)^{1/3} \sim I$ in most of the practical cases). Constant A_1 can be estimated as $A_1 \approx (4\pi)^{-1/3}/2$.

iii) Remarks for Practical Applications

Tables 3 and 4 specify various possible regimes and asymptotes. It is not easy to support all of these regimes in applications. In practice we can use the pdf with the Gaussian core and the power tail $P_{f} \sim (f-f_{1})^{a}$. The value of *a* can be taken to match the asymptote in the region which is most important for combustion processes. The scalar dissipation N_{f} can be found then from the pdf equation given by Eq.(9.54). Note that $N_{f}P_{f}\rho_{f}=cJ_{m}(f_{d}-f)$ is the quasi-steady-state $(\partial P_{f}/\partial t\approx 0)$ solution of the pdf equation. This solution is valid for the tail region but the time derivative is significant for the Gaussian core.

9.4. CMC for Turbulent Premixed Combustion

In turbulent premixed systems, most of the fluctuation in temperature and species mass fractions are associated with a reaction progress variable. For constant pressure adiabatic systems this is best defined in terms of the sensible enthalpy, rather than the temperature or the mass fraction of a major species. This is because its balance equation does not involve large contributions from variation of specific heat and it is least affected by differential diffusion effects. Bilger¹³ has developed the theory for application of CMC to premixed systems. It should be noted from the outset that CMC is not likely to make a contribution to the fundamental closure problem of turbulent premixed combustion -- prediction of the fields of mean and variance of the progress variable. The contribution that CMC can make in premixed systems is for prediction of the effects of turbulence on the detailed chemical structure of the flame including pollutant formation.

For species such as NO, most of the formation occurs in the post-flame gases where the progress variable is close to unity in an adiabatic system. In practice, systems are not adiabatic and the post flame gases are cooled by radiation. Bilger¹³ uses this to define a

progress variable, c, which can give resolution of the chemistry occurring in the post-flame gases:

$$c \equiv \left(h^{s} - h^{s}_{u} - 2h + 2h_{u}\right) / \Delta h^{s}$$

$$(9.55)$$

where h is the enthalpy as defined in Eq.(3.32) and h^{s} is its sensible component so that

$$h = h^{S} + \sum_{i} Y_{i}(h_{0})_{i}$$
(9.56)

with $(h_0)_i$ the enthalpy of formation of species *i*. Subscript *u* refers to the values in the unburnt premixture and Δh^s is the sensible enthalpy rise from the value in the unburnt premixture to the adiabatic equilibrium fully burnt value. For a system with radiation or other heat losses *h* becomes less than h_u and continues to decrease in the post-flame gases so that *c* increases above unity. All species and the temperature will then become single-valued functions of *c*. Bilger¹³ gives an example of how species and temperature might vary with *c* in such a system.

The balance equation for the enthalpy is given by Eq.(3.33) and this and Eqs (3.2) and (9.56) can be used to derive the balance equation for c:

$$\rho \frac{\partial c}{\partial t} + \rho \mathbf{v} \cdot \nabla c - \nabla \cdot (\rho \alpha \nabla h^{S}) = \rho \frac{\sum_{i} W_{i} (h_{0})_{i} + W_{R}}{\Delta h^{S}} \equiv \rho S_{c} \qquad (9.57)$$

The rate of pressure rise term has been neglected. With ζ taken as the sample space variable for *c*, conditional averages *Q* are made for species mass fractions conditional on $c(\mathbf{x},t) = \zeta$ and result in a modelled balance equation

$$\frac{\partial Q}{\partial t} + \langle \mathbf{v} | \boldsymbol{\zeta} \rangle \cdot \nabla Q = \langle W | \boldsymbol{\zeta} \rangle + N_{\boldsymbol{\zeta}} \frac{\partial^2 Q}{\partial \boldsymbol{\zeta}^2} - \langle S_{\mathbf{c}} | \boldsymbol{\zeta} \rangle \frac{\partial Q}{\partial \boldsymbol{\zeta}} - \frac{\nabla \cdot (\langle \mathbf{v} '' \boldsymbol{Y}'' | \boldsymbol{\zeta} \rangle P(\boldsymbol{\zeta}))}{P(\boldsymbol{\zeta})}$$
(9.58)

where S_c is defined in Eq. (9.57) and the fluctuations \mathbf{v}'' and Y'' are now about their means conditional on $c = \zeta$. Also

$$N_{\zeta} \equiv \langle \alpha \nabla c \cdot \nabla c \, | \, \zeta \rangle \tag{9.59}$$

is the conditional scalar dissipation for the progress variable, *c*. A similar equation can be derived for the conditional enthalpy¹³. Eq.(9.58) is similar to the CMC equation for nonpremixed systems except that it contains the extra term involving $\langle S_c | \zeta \rangle$. First order closure for this term and for $\langle W | \zeta \rangle$ may be possible.

Mantel & Bilger⁸⁴ have studied the effects of turbulence on N_{ζ} using the DNS data base of Trouve and Poinsot¹³⁸. They find that at high turbulence levels N_{ζ} is significantly increased above its laminar values in the preheat zone of the flame. This DNS has a one-step irreversible reaction and there is no effect of turbulence on $\langle W_i | \zeta \rangle$ for unity Lewis number. O'Young & Bilger¹⁰⁷ have used advanced laser diagnostic methods to measure N_{ζ} in turbulent premixed propane-air flames. They find that at high turbulence levels N_{ζ} is considerably decreased below its laminar flame values. This seems to be associated with a decrease in OH concentrations¹⁰⁶. This suggests that multistep chemistry is needed to fully understand the structure of premixed flames. DNS with the 2-step reduced chemistry for methane of Swaminathan & Bilger¹²⁹ is being attempted in collaboration with Poinsot and his co-workers. This DNS should allow testing of the above model for CMC in turbulent premixed combustion.

9.5. Conditional methods and Lagrangian modelling

The techniques of deriving and closing equations for conditional expectations, which are presented in this review, can be applied to other conditional expectations with various conditions which are not analysed in previous sections. Klimenko⁵⁷ considered $Q_v \equiv \langle Y | \mathbf{v} \rangle$, the value of scalar *Y* conditioned on a fixed value of the velocity \mathbf{v} , and demonstrated that this value relates the Lagrangian phase-space density function $F_L(\mathbf{v}, \mathbf{x}; t)$ (or the Lagrangian pdf $P_L(\mathbf{v}, \mathbf{x}; t)$) and the Eulerian velocity pdf $P_v(\mathbf{v}; \mathbf{x}, t)$ by the equation

$$F_{\rm L} = Q_{\rm v} P_{\rm v} \tag{9.60}$$

The conditional technique which has some similarities with CMC but involves three conditioning variables - the velocity components - allows one to obtain the exact equation for $F_{\rm L}$

$$\frac{\partial F_{\rm L}}{\partial t} + \frac{\partial v_{\rm i} F_{\rm L}}{\partial x_{\rm i}} + \frac{\partial G_{\rm i} F_{\rm L}}{\partial v_{\rm i}} + \frac{\partial H_{\rm i}}{\partial v_{\rm i}} = W_{\rm v} P_{\rm v} + E$$
(9.61)

where

$$E \equiv E_{\rm D} - E_{\rm v}, \quad E_{\rm D} \equiv D \nabla \cdot (\langle \nabla c \, | \, \mathbf{v} \rangle P_{\rm v}), \quad E_{\rm v} \equiv \nu \frac{\partial \langle c \nabla v_{\rm i} \, | \, \mathbf{v} \rangle P_{\rm v}}{\partial v_{\rm i}}$$
$$H_{\rm i} \equiv \nu \frac{\partial \langle (\nabla v_{\rm i} \cdot \nabla v_{\rm j}) c \, | \, \mathbf{v} \rangle P_{\rm v}}{\partial v_{\rm j}} - (D + \nu) \langle \nabla v_{\rm i} \cdot \nabla c \, | \, \mathbf{v} \rangle P_{\rm v} + \langle c'' S''_{\rm i} \, | \, \mathbf{v} \rangle P_{\rm v}$$
(9.62)

i,*j* = 1,2,3, $G_i \equiv -\langle \partial p / \partial x_i | \mathbf{u} \rangle / \rho$ and $W_v \equiv \langle W | \mathbf{v} \rangle$ is the conditional expectation of the particle source. The convention of summation over repeated indices is applied. This equation is a direct consequence of the Navier-Stokes and scalar transport equations and it is shown to be consistent with the traditional Lagrangian models¹¹³ based on the Markov-process assumptions whose general representation is given by

$$\frac{\partial F_{\rm L}}{\partial t} + \frac{\partial v_{\rm i} F_{\rm L}}{\partial x_{\rm i}} + \frac{\partial A_{\rm i} F_{\rm L}}{\partial v_{\rm i}} - \frac{\partial^2 B_{\rm i} F_{\rm L}}{\partial v_{\rm i} \partial v_{\rm i}} = W_{\rm v} P_{\rm v}$$

$$(9.63)$$

where B_{ij} and A_i are the diffusion and drift coefficients. The findings of Ref. 57 involve several additional theoretical constraints which can be effectively used in Lagrangian modelling. These results are supported by the direct numerical simulations of Weinman and Klimenko¹⁴².

10. CONCLUSIONS AND PERSPECTIVES

It may be concluded that CMC methods have a sound theoretical basis and that CMC predictions of laboratory and DNS experiments give very good agreement. The novelty of this approach brings forward a rich variety of modeling problems that are proving stimulating theoretical and to the of workers in this field. The approach also gives much growing number new physical insight into phenomena of turbulence-chemistry the interactions.

In particular we note the following:

- The primary closure hypothesis is well defined mathematically and appears to be valid over a wide range of conditions. Its validity in flows with local extinctions and ignition and near the fast chemistry limit needs further investigation.
- Workable models for closing the other unclosed terms in the CMC equation are available, but further work is needed to improve them.
- additional • The dimensionality associated with the conditioning variable has been a deterrent to using the method in problems with 2 and 3 spatial dimensions. In problems with low spatial gradients it may be possible in the conditional averages to use a much CMC calculation coarser spatial grid in the than that needed in the flow solver.
- Computational costs are moderate even with full chemical mechanisms. Little work has been done on the numerical methods used and significant improvements may be possible.
- It seems likely that the method will be able to incorporate the effects of differential molecular diffusion.

Initially it appeared that the technique may be limited to problems extinction The where local and ignition phenomena are not significant. of closure recent success second order prediction of autoignition in nonpremixed turbulent flows gives hope that the technique may be more widely applicable than originally thought. There much further is development needed, however.

Acknowledgements-- Part of the first author's contribution to this review written while he held Australian Research Council (ARC) was an postdoctoral fellowship at the University of Sydney. The ARC also supports the research of the second author. The authors would also like to thank Drs Nedunche Swaminathan, Richard Brown and Egon Hassell who kindly took time to read the draft of this review and provided expert comment on it. Any ommissions however, responsibility errors or are, the of the authors.

REFERENCES

- 1. Bachalo, W.D., 25th Symposium (Int'l) on Combustion, p.333 (1994).
- 2. Barlow, R.S. and Carter, C.D., Combust. Flame, 97, 261 (1994).
- 3. Barlow, R.S. and Carter, C.D., Combust. Flame, 104, 288 (1996).
- 4 Barlow, R. S. and Frank, J. H., Web site: http://www.ca.sandia.gov/tdf/3rdWorkshop/M3APilot97/FlameD.html
- 5. Batchelor, G.K. *The theory of homogeneous turbulence*. Cambridge. Univ.Press (1953).
- 6. Bilger, R.W, Combustion Sci. and Technology, 11, 215-217, (1975).
- Bilger, R.W., "Turbulent Flows with Nonpremixed Reactants", in *Turbulent Reacting Flows*, P.A. Libby and F.A.Williams, Eds, Springer Verlag, pp. 65-113 (1980).
- 8. Bilger, R.W. 21st Symposium (Int'l) on Combustion, p.418 (1985).
- 9. Bilger, R. W., Annual Review Fluid Mech., 21, pp. 101-135. (1989)
- Bilger, R.W., Conditional Moment Methods for Turbulent Reacting Flow Using Crocco Variable Conditions, *Report TN F-99*, Dept. Mech Eng., University of Sydney, Feb 1991.
- Bilger, R.W., in Aerothermodynamics in Combustors, R.S.L.Lee, J.H.Whitelaw, T.S. Wung, Eds, Springer-Verlag, pp 1-18 (1992).
- 12 Bilger, R.W., Phys. Fluids A, 5, 436, (1993).
- 13 Bilger, R.W., "Conditional Moment Closure Modelling Advanced and Measurements", Laser in Turbulence and Molecular Processes in Takeno, *Combustion* Conference) T. (Sixth Toyota ed., Elsevier, Amsterdam 267-285 (1993).
- 14 Bilger, R.W., Unpublished lecture notes on combustion (1994).

- 15 Bilger, R.W., Antonia, R.A. and Srinivasan, K.R., *Phys.Fluids*, **19**, 1471 (1976).
- Bilger, R.W., Saetran, L.R. and Krishnamoorthy, L.V., J. Fluid Mech.
 233, 211 (1991).
- 17 Borghi, R., Advances in Geophysics, 18B, 349-365 (1974).
- 18 Borghi, R., *Recent Advances in Aerospace Sciences*, C.Casci, Ed., Plenum Press, New York, pp 117-138 (1985).
- 19 Borghi, R., Prog. Energy Combust. Sci., 14, 245 (1988).
- 20 Bracewell, R., *The Fourier Transform and its Applications*, McGraw-Hill, (1965).
- 21 Bray, K.N.C. and Libby, P.A. "Recent Developments in the BML Model of Premixed Turbulent Combustion", in *Turbulent Reacting Flows*, Libby, P.A. and Williams, F.A., (Eds), Academic Press, pp 115-151 (1994).
- K.N.C. 22 and Peters, N., "Laminar Flamelets Turbulent Bray, in Flames", in Turbulent Reacting Libby, P.A. Williams, Flows, and F.A., (Eds), Academic Press, pp 64-113, (1994).
- 23 Brown, R.J. and Bilger, R.W., J. Fluid Mech, 312, 373-407, (1996).
- 24 R.J. R.W., "Measurement Brown. and Bilger, and Modelling of Non-bouyant turbulent reacting plumes", Twelfth Australasian Fluid Mechanics Conference, The University of Sydney, pp. 621-624, (1995).
- 25 Brown, R.J. and Bilger, R.W., *Atmospheric Environment*, **32**, 611 628 (1998).
- 26 Chen, H., Chen, S. and Kraichnan, R.H., *Phys. Rev. Lett.*, **63**, 2657 (1989).
- 27 Chen, J.-Y. and Kollmann, W. "Comparison of Prediction and
Measurement in Nonpremixed Turbulent Flames", in *Turbulent Reacting Flows*, Libby, P.A. and Williams, F.A., (Eds), Academic Press, pp. 211-308 (1994).

- 28 Chen, J.-Y., Combust. Sci. Technol., 122, 63 94 (1997).
- 29 Correa, S.M., Combust. Flame, 93, 41-60 (1993).
- 30 Correa, S. M. and Braaten, M.E., Combust. Flame, 94, 469, (1993).
- 31 Corrsin, S., Advances in Geophysics, 18A, (25-60) (1974).
- 32 "Recent Developments in Pdf Methods", Turbulent Dopazo, С., in P.A. Williams, F.A., Reacting Flows, Libby, and (Eds), Academic Press, pp. 375-474 (1994).
- 33 Drake, M.C., 21st Symposium (Int'l) on Combustion, pp.1579-1589 (1986)
- 34 Dutta, A. and Tarbell, J.M., AIChE J., 35, 2013 (1989).
- 35 Faeth, G.M., Progr. Energy Combust. Sci., 13, 249 (1987).
- 36 Faeth, G.M., 26th Syposium (Int'l) on Combustion, pp. 1593-1612 (1996).
- 37 Favre, A., "Statistical Equations of Turbulent Gases in Problems of Hydrodynamics and Continuum Mechanics", S.I.A.M., Philadelphia, pp. 231-266 (1969).
- 38 Flury, M. and Schlatter, M. "Laser Doppler velocimetry measurements in turbulent nonpremixed hydrogen/helium flames", at internet address: http://www.ca.sandia.gov/tdf/DataArch/H2HeData.html
- 39 Fox, R.O., *Revue de Institut Francais du Petrole*, **51** (2) 215-243 (1996).
- 40 Gas Research Institute, GRI Mech 2.11, Web site http:/euler.berkeley.edu/gri-mech/

- 41 Girimaji, S.S., Phys. Fluids A, 4, 2529-2537 (1992).
- 42 Gough, A.J., Mobini, K., Chen, Y.-C. and Bilger, R.W., "Measurements a Confined Bluff-Body Burner Modelled and Predictions in as an Imperfectly Stirred Reactor", 27th Symposium (int'l) on Combust., The Combustion Institute, Pittsburgh, PA, 3181 (1998).
- 43 Hindmarsh, A.C., "ODEPACK, a Systemized Collection of ODE Solvers" in Scientific Computing, R. Stapleman et al (Eds), North Holland p.55 (1983).
- 44 Hinze, O.J., *Turbulence. An introduction to its mechanism and theory.* N.Y.: Mc Graw Hill (1959).
- 45 Jayesh and Warhaft, Z., Phys. Fluids A, 4, 2292 (1992).
- 46 Janicka, J. and Kollmann, W., 17th Symposium (Int'l) on Combustion, The Combustion Institute, p. 421, (1979).
- Jones, W.P., "Turbulence Modelling and Numerical Solution Methods for Variable Density and Combusting Flows", in *Turbulent Reacting Flows*, Libby, P.A. and Williams, F.A., (Eds), Academic Press, pp. 309-374 (1994).
- 48 Juneja, A. and Pope, S.B., *Phys Fluids* 8, 2161-2184 (1996).
- 49 Kailsanath, P., Sreenivasan, K.R. and Saylor, J. R. *Phys. Fluids A*, 5, 3207 (1993).
- 50 Kee, R.J., Rupley, F.M. and Miller, J.A. "Chemkin-II: A FORTRAN Chemical Kinetics Package for the Analysis of Gas Phase Chemical Reactions" Sandia Report SAND 89-8009, Sandia National Laboratories (1989).
- 51 Kee, R.J., Grcar, J.F., Smooke, M.D. and Miller, J.A., "A FORTRAN Program for modeling Steady One-Dimensional Premixed Flames" SANDIA Report SAND 85-8240, Sandia National Laboratories, Livermore, (1985).

- 52 Kent, J.H. and Bilger, R.W., "The prediction of turbulent diffusion flame fields and nitric oxide formation. - in *16th Symp.(Int'l) on Combustion*, The Combustion Inst. Pittsburgh, 1643-1656 (1977).
- 53 Kerstein, A.R., J. Fluid Mech., 240, 289-313 (1992).
- 54 Klimenko, A. Y., Fluid Dynamics, 25, 327-324 (1990).
- 55 Klimenko, A. Y., "Conditional Moment Closure and Diffusion in Conserved Scalar space", ECOLEN preprint No 1 / CIAM preprint No 7 (1992).
- 56 Klimenko, A. Y., Fluid Dynamics, 28, 630-637, (1993).
- 57 Klimenko, A.Y., Phys. Fluids, 7, 446 (1995).
- 58. "Conditional methods Klimenko, A.Y., and Lagrangian particle diffusion", 12th Australian Fluid pp.751-754 *Mechanics* Conference (1995).
- 57 Klimenko, A.Y., *Phys. Fluids*, **10**, No 4 (1998).
- 59. Klimenko, A. Y. and Bilger, R.W., "Relationship between conserved pdf's scalar dissipation turbulent flows", Charles scalar and in Kolling Research Laboratory *TNF-100*, Dept. of Mech. Eng., The University of Sydney (1993).
- 60 Klimenko, A. Y. and Bilger, R.W., "Some analytical and asymptotic results for pdfs and conditional expectations in turbulent shear flows", 10th Symposium on Turbulent Shear Flows, Pennsylvania State University, USA, v.3, Session 31, 25-29, (1995).
- 61 Klimenko, A. Y. and Bilger, R.W., "Mixing micro-structure in sprays and application to Conditional Moment Closure methods", *Australian Symposium on Combustion*, Session B2, paper 3, (1995).
- 62 Klimenko, A.Y., Bilger, R.W. and Roomina, M.R., *Combust. Sci. Tech.*, **107**, 403 (1995).

- 63 Kolmogorov, A.N., C.R.Acad.Sci. U.S.S.R., 30, 301-305, No 4 (1941).
- 64 Kolmogorov, A. N., J.Fluid Mech. 13. 82-85 (1962).
- 65 Kosaly, G., Personal Communication (1996).
- 66 Kronenburg, A. and Bilger, R.W. Physics Fluids, 9, 1435-1447 (1997).
- A. Bilger, R.W., "Modelling differential diffusion 67. Kronenburg, and in nonpremixed reacting turbulent flow". Combust. Sci. Technol., (submitted).
- 68 Kronenburg, A., Bilger R.W. and Kent, J.H., "Second Order Conditional Moment Closure for Turbulent Jet Diffusion Flames", 27th Symp. (Int'l) on Combustion, 1097 (1998).
- 69 R.W. J.H. of Kronenburg, A., Bilger, and Kent. "Computation diffusion conditional average scalar dissipation in turbulent jet flames", Flow, Turbulence and Combustion, submitted (1998).
- Kroneburg, R.W. Kent, J.H., "Modelling 70. A., Bilger, and soot turbulent diffusion formation in methane-air jet flames", Combust. Flame (to appear).
- 71 Kuznetsov, V.R., Fluid Dynamic, 17, 815-820, No 6 (1982).
- Kuznetsov, V.R. and Sabelnikov, V.A. *Turbulence and combustion*. Moscow, Nauka, (in Russian, 1986) . translation: *Turbulence and combustion*. Hemisphere (1989).
- 73 Launder, B.E., Int. J. Heat Fluid Flow, 10, 282-299 (1989).
- 74 LaRue, J.C. and Libby, P.A., Phys. Fluids, 17, 1956 (1974).
- 75 Lee, Y. Y. and Pope, S.B., Combust. Flame, 101, 501-528 (1995).
- 76 Li, J.D. and Bilger, R.W., Physics Fluids A, 5, 3255-3264 (1993).
- 77 Li, J.D. and Bilger, R.W., "Conditional Moment Closure of Reactive

Plumes in Homogeneous Turbulence", *Twelfth Australasian Fluid Mechanics Conference*, The University of Sydney, pp. 625-628, (1995).

- 78 Li, J.D. and Bilger, R.W., J. Fluid Mech 318, 338 (1996).
- 79 Li, K.T., "Chemical Reaction with complex reactions in unpremixed tubular reactors", PhD Thesis, Carnegie-Mellon University, (1985).
- 80 Li, K.T. and Toor, H.L., AIChE J., 32, 1312 (1986).
- 81 Libby, P.A. and Williams, F.A., (Eds) Turbulent Reacting Flows, Academic Press (1994).
- 82 Liew, S.K., Bray, K.N.C. and Moss, J.B., *Combust. Flame*, **56**, 199-213, (1984).
- 83 Lighthill, M. J., Introduction to Fourier Analysis and Generalised Functions, Cambridge University Press (1958).
- 84 Mantel, T. and Bilger, R.W. Combust. Sci. and Technol., **110**, 393 (1995).
- 85 Masri, A.R., Dibble, R.W. and Barlow, R.S., *Combust. Flame,*. **89**, 167, (1992).

 Mastorakos, E. and Bilger, R.W., "Second-order Conditional Moment Closure for for the Autoignition of Turbulent Flows", *Phys Fluids*, 10, 1246, (1998).

- 87 Mastorakos, E., Pires da Cruz, A., Baritaud, T.A. and Poinsot, T.J., Combust. Sci. Technol., 125, 243 (1997).
- 88 . Mauss, 23rd F., Keller, D. and Peters, N. Symp. (Int'l)on Combustion, The Combustion Institute, Pittsburgh, PA, 1990, pp 693-698.
- 89 Е., "Α validity investigation conditional Mell. W. of the moment closure model for turbulent combustion", unpublished manuscript, (1995).

- 90 Mell, W.E., Nilsen, V., Kosa'ly, G. and Riley, J.J., *Combust. Sci. Technol.*, **91**, 179, (1992).
- Mell, W.E., Nilsen, V., Kosa'ly, G. and Riley, J.J., *Phys. Fluids*, 6, 1331-1356 (1994).
- 92 Mi, J., Antonia, R.A. and Anselmet, F., *Phys. Fluids*, **7**, 1665 (1995).
- 93 Miller, J.A. and Bowman, C.T., *Progr. Energy Comb. Sci.*, **15**, 287-338 (1989).
- 94 Mobini, K., PhD Thesis, The University of Sydney, (1997).
- 95 Mobini, K., Bilger. R.W. and Smith, N.S.A.,"Investigation of pdf imperfectly stirred modeling", shape effects on reactor Australian Symposium on Combustion, Australia and New Zealand Section of The Combustion Institute, Adelaide, (1995).
- 96 Mobini, K., Bilger, R.W. and Smith, N.S.A.,"Imperfectly stirred modeling turbine reactor of a gas combustor" First Asia-Pacific Conference on Combustion, Japanese Section of The Combustion Institute, Osaka, 174-177 (1997).
- 97 Moin, P. and Kim, J. Scientific American, 276 (1), 62-68 (1997).
- 98 Monin, A.S. and Yaglom, A.M. . Statistical Fluid Mechanics. The MIT Press, Vol.1 (1973), Vol.2 (1975).
- 99 Montgomery, C.J., Kosa'ly, G. and Riley J.J. Combustion and Flame, 95, 247 (1993).
- 100 Nilsen, V. and Kosa'ly, G., "Differentially diffusing scalars in turbulence", *Phys. Fluids*, **9**, 3386 (1997).
- 101 Nilsen, V. and Kosa'ly, G., AIAA Paper # 97-0256, Reno, (1997).
- 102 Oboukhov, A.M., "Structure of the temperature field in turbulent

flow", Izv. Akad. Nauk U.S.S.R., Ser. Geography and Geophysics. 13, 58-69, No 1 (1949).

- 103. Oboukhov, A.M., Description of turbulence in terms of Lagrangian variable. Advances in Geophysics, 6, 113-116 (1959).
- 104. Oboukhov, A.M., J.Fluid Mech. 13. No 1 77-81 (1962).
- 105. O'Brien, E. E. and Jiang, T.-L., Phys. Fluids A, 3, 3121 (1991).
- 106. O'Young, F., "Simultaneous Laser Imaging of Turbulent Premixed Flames", Ph.D. Thesis, The University of Sydney (1996).
- 107. O'Young, F. and Bilger, R.W., Combust. Flame, 109, 682 (1997).
- 108. Papoulis, A. Probability, random variables, and stochastic processes. McGraw-Hill (1984).
- 109. Peters, N. Progr. Energy and Combust. Sci., 10, 319-340 (1984).
- 110. Peters, N., 21st Symp. (Int'l) on Combust., 1231 (1986).
- 111 Pitsch, H., Wan, Y.P. and Peters, N. SAE Paper 952357, 1995.
- 112. Plumb, R.A. and Ko, M.K.W., J. Geophys. Res., 97, 10145-10156, (1992).
- 113. Pope, S.B., Progress in Energy Combustion Sci., 11, 119-192 (1985).
- 114. Pope, S.B., 23rd Symposium (Int'l) on Combustion, 591-612 (1990).
- 115. Pope, S.B., *Turbulent Flows*, Cambridge University Press, 1999.
- 116. Roomina, M.R. Ph D Thesis, The University of Sydney (1998).
- 117. Roomina, M.R. and Bilger, R.W. "Conditional Moment Closure (CMC) Predictions for a Methanol Jet Flame", 1997 Australian Symposium on Combustion and Fifth Australian Flame Days, pp 53-57 (1997).

- 118. Senoy, U.V. and Toor, H.L., AIChE J., 35, 1692 (1989)..
- 119. Sinai, Ya.G and Yakhot, V. Phys. Rev. Lett, 63, 1962 (1989).
- 120. Singh, M., "Chemical reactions in one and two dimensional turbulent flow systems", PhD Thesis, Carnegie-Mellon University (1973).
- 121. Smith, N.S.A."Development of the Conditional Moment Closure Method for Turbulent Combustion", PhD Thesis, The University of Sydney, Australia (1994).
- 122. Smith, N.S.A. and Bilger, R.W., "Conditional moment closure for turbulent nonpremixed combustion modeled as an imperfectly stirred reactor", *Combustion and Flame*, (submitted) (1994).
- 123. Smith, N.S.A., Bilger R.W. and Chen, J.-Y. Twenty-Fourth Symposium (Int'l) on Combustion, 263 (1992).
- 124. Smith, N.S.A., Bilger, R.W., Carter, C.D., Barlow, R.S. and Chen, J-Y., *Combust. Sci and Technol*, **105**, 357-375 (1995).
- 125. Smith, N.S.A., Barlow, R.S., Chen, J.-Y. and Bilger, R.W., "Nitric Oxide formation in dilute hydrogen jet flames: isolation of the effects of radiation andturbulence-chemistry submodels", *Combust. Flame*, **117**, 4 (1999).
- 126. Smoot, L.D. and Smith, P.J., *Coal combustion and gasification*, Plenum Press (1985).
- 127. Southwell, J.P., "Reactions in Ocean Plumes", M.E.(Res) Thesis, The University of Sydney, (1993).
- 128. Starner, S.H., Bilger, R.W., Long, M.B., Frank, J.H. and Marran, D.H., *Combust. Sci. Technol.*, **129**, 141 (1997).
- 129. Swaminathan, N. and Bilger, R.W., *Combust. Sci. Technol*, **127**, 167 196 (1997).

- Swaminathan, N. and Bilger, R.W. "Assessment of Combustion Submodels for Turbulent Nonpremixed Hydrocarbon Flames", *Combust. Flame*, 116, 519 and 675 (1999).
- 131. Swaminathan, N and Bilger, R.W., "Conditional Variance Equation and its Analysis", 27th Symp. (Int'l) on Combustion, 1191-1198 (1998).
- 132. Swaminathan, N. and Bilger, R. W., "Study of the conditional covariance equations second order CMC closure", variance and for Phys. Fluids (to appear).
- 133. Swaminathan, N. and Mahalingam, S., *Combust. Sci. and Technol.*, **112**, 301 (1996).
- 134. Taylor, G.I., Proc. Lond. Math. Soc., 20, 196-212 (1921).
- 135. Tennekes, H. and Lumley, J.L., *A First Course in Turbulence*, MIT Press (1972).
- 136. Tong, C. and Warhaft, Z., J. Fluid Mech. 292, 1-38 (1995).
- 137. Toor,H.L., AIChEJ, 37, 1737-1738 (1991).
- 138. Trouve', A. and Poinsot, T., J. Fluid Mech., 278, 1 (1994).
- 139. Turns, S.R., An Introduction to Combustion, McGraw-Hill, (1996).
- 140. Vassilatos, G. and Toor, H.L., AIChE J., 11, 666, (1965)..
- 141. Vatazhin, A.B. and Klimenko, A.Y, *Fluid Dyn. Research*, **29**, 178, 1994 and *J. Aerosol Sci*, **25**, SUP/1, 373, (1994).
- 142. Weinman, K.A. and Klimenko A.Y. "Estimation of the Kolmogorov for function constant (Co)the Lagrangian structure using direct numerical simulation for continous scalar" Phys. Fluids, а (submitted).
- 143. Wilcox, D.C., AIAA J. 26, 1299 (1988).

- 144. Williams, F.A., Combustion Theory, Benjamin Cummings (1985).
- 145. Williams, F. Reduced Kinetic **Mechanisms** A. in and Asymptotic Approximations for Methane - Air Flames, Smooke, M. D. (Ed.), Springer-Verlag (Lecture Notes in Physics, vol. 384), pp.68-85 (1991).
- 146. Yakhot, V., Phys. Rev. Lett., 63, 1965 (1989).
- 147. Yeung, P.K. and Pope, S.B., Phys Fluids A 5, 2461 (1993).
- 148. Zhang, S. and Rutland, C.J., Combust. Flame, 102, 447 (1995).

FIGURE CAPTIONS

- Fig. 1. Scatter plots of all data and conditional averages conditional on mixture fraction for the temperature and OH mass fraction in a piloted diffusion flame of methanol at $x/D_j=20$, $U_j=60$ m/s and $r/D_j=:$ O, 1.7; \Box , 1.4; Δ , 1.1. The full curves are for a laminar counterflow diffusion flame with a strain rate parameter a = 5 s⁻¹. Data of Masri *et al*⁸⁵.
- Fig. 2. Clipped Gaussian pdfs with various values of mean mixture fraction (first column) and root mean square fluctuation of mixture fraction (second column) The delta functions at $\eta = 0, 1$ have strengths α,β of 0.048, 0.048; 0.21, 0.0; and 0.07, 0.0, respectively.
- Fig. 3. Beta function pdfs with the same mean and root mean square fluctuation of mixture fraction as in Fig. 2 Note the different behaviour near $\eta=0,1$ and the absence of delta functions.
- Fig. 4. Probability of the event $Z_1 < Y < Z_2$ shown (a) using the cumulative probability function and (b) using the pdf.
- Fig. 5. Probability of the event $Z < Y < Z + \Delta Z$.
- Fig. 6. Surface plot of a Gaussian joint pdf for two variables.
- Fig. 7. Contour plot of the Gaussian joint pdf of Fig. 6.
- Fig. 8. (a) Contour plot of the joint pdf P(Z₁,Z₂) -------; locus of <Y₁|Z₂> ------; locus of <Y₂|Z₁> ---------.
 (b) Conditional pdfs for two values: Z₁= a and b. Note that the peak of P(Z₂|b) is higher than the peak of P(Z₂|a) even though the peak of P(Z₂,b) is lower than the peak of P(Z₂,a). This is because P(Z₁=a)>P(Z₁=b).
- Fig. 9. Behaviour of conditional scalar dissipation with smooth pdfs.(a) Unbounded Gaussian pdf; (b) Smooth pdf with fixed bounds;(c) Smooth pdf with moving bounds.

Fig. 10. Qualitative behaviour of the correlation function K_{ξ} .

- Fig. 11. Pdfs $\stackrel{\circ}{P}$ $\stackrel{\circ}{(\eta)}$ in the wake of a heated cylinder versus $\stackrel{\circ}{\eta} = \Delta T / \Delta T_c$ for various transverse locations. Adapted from Klimenko *et al*⁶².
- Fig. 12. Normalised pdf integral across the flow, $P(\eta)$, Adapted from Klimenko *et al*⁶².
- Fig. 13. Normalised conditional scalar dissipation integral across the flow, $\hat{N}(\hat{\eta})$. Adapted from Klimenko *et al*⁶².
- Fig. 14. Integrated conditional scalar dissipation, N^* , measured in turbulent jet diffusion flames of air-diluted methane. Adapted from Starner *et al*¹²⁸.
- Fig. 15. Schematic diagram of the reactive scalar mixing layer. Adapted from Bilger *et al*¹⁶.
- Fig. 16. Conditional averages of reactant concentration conditional on mixture fraction as measured in a reacting scalar mixing layer with $N_{\rm D}$ = 0.42 and x_1/M = 21. Different symbols are used for different points across the flow. Adapted from Bilger¹².
- Fig. 17. Predictions for the conditional average reactant concentration in a reacting scalar mixing layer as a function of η and normalized downstream distance $\hat{x}_1 = N_D x_1 / M$ using Eq.(5.42) with A = 0.03, $\eta_s = 0.5$. Adapted from Bilger¹².
- Fig. 18. Conditional averages of reactant concentration at $\eta = 0.5$ in a reacting scalar mixing layer. Curves are for predictions using Eq. (5.42) with various values of A. Data points from experiments of Bilger *et al*¹⁶ : Δ , $N_{\rm D}=0.42$; \Box , $N_{\rm D}=2.6$. Figure adapted from Bilger¹².
- Fig. 19. CMC predictions (full lines) of Smith *et al*¹²⁴ for nitric oxide compared with laser induced fluorescene measurements (dots) of Barlow and Carter².
- Fig. 20. CMC predictions (full lines) of Smith $et a l^{124}$ for nitric oxide

compared with conditionally averages of measurements (connected circles) of Barlow and Carter².

- Fig. 21. CMC predictions (Smith *et al*¹²⁴) and laser measurements (Barlow and Carter²) of conditional average temperature in a hydrogen jet diffusion flame: $x_1 = \ell_f/2$ - predictions, -o-o-o measurements; $x_1 = \ell_f/8$ ---- predictions, -o-o- measurements.
- Fig. 22. Comparison of conditionally averaged NO mole fractions at $x_1 = \ell_f$ calculated from CMC models with 26 step and 2-step reduced with and without radiation losses. Bold/plain mechanisms. lines denote full/reduced mechanism calculations whilst solid/broken lines denote radiative/ adiabatic calculations¹²⁴.
- Fig. 23. Schematic diagram for experimental set-up for round turbulent reacting plume²³.
- Fig. 24. Experimental results and CMC predictions in a reacting turbulent round plume of NO into background O_3 : ----- conditional mean of data, - CMC predictions, reaction dominated limit²⁵.
- Fig. 25. Schematic of an axisymmetric combustor⁹⁴ Dimensions are in mm.
- Fig. 26. Mean streamlines calculated for the combustor of Fig. 25⁹⁴.
- Fig. 27. Contours of Favre-averaged mixture fraction, $f \equiv \langle \rho \xi \rangle / \langle \rho \rangle$, calculated for the combustor of Fig. 25⁹⁴.
- Fig. 28. Contours of Favre averaged variance of mixture fraction, $g \equiv \langle \rho \xi''^2 \rangle / \langle \rho \rangle$, for the combustor of Fig. 25⁹⁴.
- Fig. 29. Core and outlet averaged pdfs for an ISR model of the recirculation zone end of the combustor of Fig. 25⁹⁴.
- Fig. 30. Core-averaged conditional scalar dissipation for the ISR mode1⁹⁴ (full line) compared with laminar counterflow result with the same maximum (dashed line).
- Fig. 31. Predictions for the conditional average species mass fractions in

the recirculation zone of the combustor of Fig. 25 using the ISR $model^{94}$.

- Fig. 32. Predictions for the conditionally averaged temperature and NO mass fraction in the recirculation zone of the combustor of Fig. 25 using the ISR model⁹⁴.
- Fig. 33 Comparison between ISR predictions with the Miller-Bowman⁹³ (solid lines) as well as the GRI⁴⁰ (dotted lines) mechanisms and measured data (s y mbols)⁴². R=30.5 mm is the radius of the confining tube.
- Fig. 34. Schematic of a Dilution Flow Reactor
- Fig. 35. Comparison of predicted and experimental results: series parallel reaction of Li and Toor⁸⁰.
- Fig. 36. Pdfs of the mixture fraction for the spatially homogeneous $case^{91}$:(a) $\langle \xi \rangle = 0.25$; (b) $\langle \xi \rangle = 0.5$.
- Fig. 37. The value of $\langle N | \eta \rangle \langle N \rangle$ from the DNS data (symbols) and from the Case (b) model normalised by $\langle N \rangle$ from the DNS at times t = 0.9, 2.3, 3.7. Spatially homogeneous case.⁹¹ (a) $\langle \xi \rangle = 0.25$; (b) $\langle \xi \rangle = 0.5$.
- Fig. 38. Variation of the unconditional scalar dissipation with time⁹¹.
- Fig. 39. Time evolution of the terms on the left hand side (LHS) of the CMC equation, Eq. 4.30, expressed as a fraction of the reaction rate term. Spatially homogeneous case, $\langle \xi \rangle = \xi_s = 0.25$. (a) $Da_0 = 0.5$, (b) $Da_0 = 8$. Results of Mell *et al*⁹¹.
- Fig. 40. Predictions for normalized conditional variance in a reacting turbulent scalar mixing layer, Eq. (8.37), for $\xi_s = 0.5$. From Li and Bilger⁷⁶.
- Fig. 41. Predictions for conditional variance in a reacting turbulent scalar mixing layer compared with experiment⁷⁶. (a) $x_1/M = 8$, (b) $x_1/M = 12$.

- Fig. 42. Balance of terms in the conditional variance equation for the fuel species in nonpremixed reaction in decaying homogeneous turbulence¹³¹. T_i refers to the *i*th term as it appears in Eq. (8.38). (a) Simulation R1 with $Re_{\lambda} = 33.5$; (b) simulation R5 with $Re_{\lambda} = 50.5$.
- Predictions for conditional average mass fraction Fig. 43. of NO in a hydrogen jet diffusion flame using a partial second order conditional moment closu r e⁶⁸. Filled symbols are the conditional averaged experimental data of Barlow and Carter² with the bars indicating plus/minus two conditional standard deviations. Triangles are predictions with first order closure and squares for second order closure. (a) $x/L_v = 0.125$ and 0.5; (b) $x/L_v =$ 0.75.
- Fig. 44. Predictions and DNS data for conditional average and root mean square fluctuation of normalised temperature rise, θ , for autoignition in turbulent nonpremixed flow⁸⁶. t_1 is at $0.64\tau_h$ and t_2 is at $1.41\tau_h$ where τ_h is the shortest time to ignition of a uniform mixture. Ignition occurs at $2.05\tau_h$.
- Fig. 45. Scatterplot of differential diffusion, z, versus mixture fraction, ξ , and conditional average, Q_z , versus η for mixing in decaying homogeneous turbulence⁶⁶.
- Fig. 46. Qualitative structure of the interdroplet field⁶¹. (a) Non-inertial droplets; (b) inertial droplets.

TABLE CAPTIONS

- Table 1. Constants β_u , β_η , β_N , β_p for different types of flows.
- Table 2. Relative error in $\langle Y_F Y_O \rangle_t$ from CMC(a) and CMC(b) for all Da₀ values at each time considered. Mell et al ⁹¹.
- Table 3. Functions $P_f \equiv P(f)$ and $N_f \equiv \langle D(\nabla \varphi)^2 | \varphi = f \rangle$ for non-inertial droplets.
- Table 4. Functions $P_f \equiv P(f)$ and $N_f \equiv \langle D(\nabla \varphi)^2 | \varphi = f \rangle$ for inertial droplets.

TABLES

Flow	Dimension	$\beta_{\prime\prime}$	eta_η	β_N	β_p
Jet	2	1/2	1/2	5/2	1
	3	1	1	4	2
Wake	2	0	1/2	2	1/2
	3	0	2/3	7/3	2/3
Mixing	2	0	0	1	1
Layer					
Scalar mixing layer	2	0	0	1	-
Homogeneous turbulence	0	0	-	$2\beta + 1$	0

Table 1 Constants β_u , β_η , β_N , β_p for different types of flows

Table 2

Relative error in $\langle Y_F Y_O \rangle_t$ from CMC(a) and CMC(b) for all Da_0 values at each time considered [92]

		t = 0.24	0.93	1.62	2.30	2.99	3.68
$\xi_s = \langle \xi \rangle = 0.25$							
CMC(a)	$Da_0 = 0.5$	- 0.01	0.15	0.18	0.15	0.12	0.09
	$Da_0 = 2.0$	- 0.06	0.36	0.30	0.19	0.11	0.06
	$Da_0 = 8.0$	0.25	0.49	0.30	0.18	0.13	0.07
		- 0.02	0.0	0.02	0.025	0.034	0.04
CMC(b)		0.03	0.02	0.05	0.07	0.09	0.06
		0.02	0.04	0.11	0.10	0.15	0.13
$\xi_{\rm s} = \langle \xi \rangle = 0.5$							
CMC(a)		- 0.01	0.07	0.10	0.08	0.07	0.06
		0.02	0.25	0.24	0.18	0.13	0.10
		0.15	0.55	0.40	0.26	0.18	0.12
CMC(b)		- 0.02	- 0.01	0.01	0.01	0.01	0.01
		- 0.03	0.01	0.01	0.02	0.02	0.01
		- 0.03	0.01	0.01	0.02	0.02	0.01
		- 0.02	0.06	0.06	0.03	0.01	0.0

Table 3 Functions $P_f \equiv P(f)$ and $N_f \equiv \langle D(\nabla \varphi)^2 | \varphi = f \rangle$ for non-inertial droplets[†]

lear droplet r~rd	Kolmogorov Scale Zone	$r \sim r_k \equiv \left(\frac{D_1^3}{\varepsilon_t}\right)^{1/4}$	Interdroplet Zone	$r \sim r_c \equiv c^{-1/3}$
$m = f_{12} (f_{12} - f_{2}) \exp\left(-\frac{R}{2}\right)$	$\mathbf{B} \begin{bmatrix} \mathbf{J}_1, \mathbf{D}_1, \mathbf{\varepsilon}_t \end{bmatrix}$	17	c	
$\varphi = \frac{1}{4} \left(\frac{1}{4} \frac{1}{2} \right)^{3} $	$\langle \varphi r \rangle = f_1 + f_k F_k$	$\left(\frac{r}{r_{k}}\right); f_{k} \equiv \frac{J_{1}\varepsilon_{t}^{*}}{D_{1}^{7/4}}$	$P_{\rm f} = \frac{1}{\epsilon^{\rm o}} P_{\rm c}^{\rm o} \left(\phi_{\rm c} \right);$	$\phi_c \equiv \frac{f - f_g}{e^{\varphi}}$
$P_{f} = \frac{1}{(4\pi)^2 (D_1 \rho_1)} \times \frac{b_1}{1 n^{-4}} \ln^{-4} \left(\frac{f_d - f_d}{1 n^{-4}} \right)$	$P_{f} = \frac{cD_1^4 P_k^0(\phi_k)}{J_1 \varepsilon_k}$	$\phi_k \equiv \frac{f - f_1}{f_k}$		1
$I_d - I \longrightarrow (I_d - I)$	$J_1^2 \varepsilon_t N_1^2 \varepsilon_t$	-K	$N_{\rm f} = f^{\circ} W_1 N_{\rm C}^{\circ} \left(\phi_{\rm C} \right)$	
	$\frac{D_1^4}{D_1}$		C1. x/x #1	W_1, ε_t, c
Matc	$ \underset{\text{hing}}{\overset{\text{B}^{-}r/r_k \ll 1}{\longleftrightarrow}} f_2 = f_1 + f_k \phi_2 $	$\begin{bmatrix} \mathbf{J}_1 \\ \mathbf{J}_1 \\$	$\downarrow \qquad \qquad$	$f^{\circ} = \frac{W_1}{\varepsilon_t^{1/3} c^{2/9}}$
$\varphi = f_2 + \frac{J_1}{4\pi D_1 r}$	$F_k\left(\frac{r}{r_k}\right) = \frac{r_k}{(2\pi)r}$	$ \phi_2 = F_k \sim \left(\frac{x}{r_k}\right)^{-\frac{7}{3}} \leftarrow$	Matching	$f_1 = f_g + \phi_1 f^\circ$
andIt	$P_{k}^{\circ}(\phi_{k}) = \frac{(2\pi)^{-2}}{(\phi_{k} - \phi_{2})}$	$\frac{1}{p_{k}^{4}} P_{k}^{\circ}(\phi_{k}) = \frac{A_{1}}{\phi_{k}^{16/2}}$	$P_{c}^{*}(\phi_{c}) = \frac{\alpha}{(\phi_{c}-\phi_{c})}$	$(p_1)^{16/7}$
$N_f P_f = c J_1$	$N_{k}^{o}(\phi_{k}) = (2\pi)^{2} (\phi_{k} - \phi_{2})^{2}$	$N_{k}^{o}(\phi_{k}) = \frac{\phi_{k}^{16/}}{A_{1}}$	$N_{\rm c}^{\rm o}(\phi_{\rm c}) = \frac{(\phi_{\rm c} - \phi_{\rm c})}{A_{\rm c}}$	β^{1}
$N_{f} = (4\pi)^2 D_1^3 (f - f_2)^4$	J ²	12- x/x #1 T	†C2	$[W_1, D_1, c]$
$P_{\ell=} - C \left(\frac{J_1}{J_1} \right)^3 - \frac{J_2}{J_1}$	1 Matching	$ \xrightarrow{P_{c}^{\circ}(\phi_{c})} = (2\pi)^{-2} $	$(\phi_{0}-\phi_{1})^{-4}\beta^{3}$ $f^{\circ}=$	$=\frac{W_1}{2}$
$(4\pi)^2 (D_1) (f)$	$(f_2)^4$	$N_{c}^{o}(\phi_{c}) = (2\pi)^{2}$	$(\phi_c - \phi_1)^4 \beta^{-2} = f_2$	$= f_g + \phi_1 f^\circ$

[†] The symbols used in the table represent: determining parameters of each asymptotic zone are framed; φ is the mixture fraction and f is its The symbols used in the table represent: determining parameters of each asymptotic zone are framed; φ is the mixture fraction and *f* is its sample variable; $\rho_t \equiv \langle \rho | f \rangle$ —conditional expectation of the density; J_m —the mass flow rate of a single droplet; $J_1 = J_m(f_d - f_2)/\rho_1$ —the diffusion component of the evaporation rate; *c* is the number density of the droplets; $r_c = c^{-1/3}$ —distance between droplets; $W_m = cJ_m$ —the collective evaporation rate (identical droplets), $W_m = cJ_m/\beta$ ($\beta \neq 1$ characterizes different droplets); $W_1 = W_m (f_d - f_2)/\rho_1$ —the diffusion component of the collective evaporation rate (A_1 , ϕ_1 , ϕ_2 —the universal constants; P_k^0 , N_k^0 , P_c^0 , N_c^0 —the universal functions; r_k , r_k , r_k , r_k . Kolmogorov scales; D—diffusion coefficient; e_t —dissipation of energy; f_s —the mean value of φ in the gas phase; f^o is proportional to the dispersion of φ ; $f_2 = f_g + \varphi_1 f^0$ —effective boundary condition for φ in the Near Droplet Zone; D_1 and ρ_1 diffusion coefficient D and density ρ_i in the interdroplet space. The functions used for convenience $R^{-1} = \rho_1 D_1 \int_r^{\infty} (r^2 \rho_t D)^{-1} dr$; $R_w = J_m (4\pi D_1 \rho_1)/(R^4 D_1 \rho_1)$. [‡] The pdf is proportional to the concentration of the droplets $P_f - c$ and the framed parameters determine $P_f c$.

Table 4	
Functions $P_f \equiv P(f)$ and $N_f \equiv (D(\nabla \varphi)^2 \varphi = f)$	for inertial droplets

Near droplet _{Tud} ~r _d	Kolmogorov Scale Zone	$\tau \sim t_k \equiv \left(\frac{D_1}{\varepsilon_t}\right)^{1/2}$	Interdroplet Zone	$\left(\varepsilon_{t}\tau^{3}\right)^{1/2} \sim r_{c}$
$\mathbf{A} N_{f} P_{f} \rho_{f} = c J_{m} \left(f_{d} - f \right)$	$\mathbf{B} \left[\underline{S_1, D_1, \varepsilon_1} \right]^{\ddagger}$	C a ^{1/2}	c	
$ \begin{array}{c} \mathbf{A} \star \ \tau u_d / r_d \gg 1 \\ \hline D_1, S_1 \equiv J_1 / u_d \end{array} $	$+ \qquad \qquad$	$f_{k} \equiv \frac{S_{1}\varepsilon_{t}}{D_{1}^{3/2}}$ $f_{-f_{t}}$	$P_{f} = \frac{1}{f^{\circ}} P_{c}^{\circ} \left(\phi_{c} \right)$	$ \phi_c \equiv \frac{f - f_g}{f^\circ} $
$\varphi = f_2 + \frac{S_1}{4\pi D_1 \tau} \exp\left(-\frac{r^2}{4D_1 \tau}\right)$	$P_{f} = c u_{d} \frac{S_{1} r_{k} (\varphi_{k})}{S_{1} \varepsilon_{t}^{3/2}};$	$\phi_k \equiv \frac{1}{f_k}$	$N_{\rm f} = f^{\circ} W_1 N_{\rm c}^{\circ} \left(\phi_{\rm c} \right)$	-)
$f_{\max} = f_2 + \frac{S_1}{4\pi D_1 \tau}$	$N_{\rm f} = \frac{S_1 \mathcal{E}_{\rm t}}{D_1^{7/2}} N_{\rm k}^{\circ} (\phi_{\rm k})$	$N_{f}P_{f}=cJ_{1}$	C1- [SUE] [‡]	C1 $W_1, \varepsilon_t, c, u_d$
where t≣x _l /u _d ∉ Matchi	$ \begin{array}{c} \begin{array}{c} & \mathbf{z} \\ & \mathbf{z} \\ \end{array} \\ \xrightarrow{\mathbf{ng}} \\ & \mathbf{f}_2 = f_1 + f_k \phi_2 \\ \end{array} \\ \xrightarrow{\mathbf{ng}} \\ & \mathbf{f}_2 = f_1 + f_k \phi_2 \\ \end{array} $	$\frac{S_{1}, \varepsilon_{t}}{S_{1}, \varepsilon_{t}}^{\dagger}$		$f = \frac{m_1}{(\varepsilon_t c u_d)^{1/4}}$ $f_1 = f_0 + \phi_1 f^\circ$
	$F_{k}\left(\frac{\tau}{\tau_{k}}\right) = (4\pi)\tau^{-} + \phi_{2}$ $P_{k}^{\circ}\left(\phi_{k}\right) = \frac{1}{1-\tau^{-}}$	$P_{k}^{\circ}(\phi_{k}) = \frac{A_{1}}{\phi_{k}^{7/3}} \leftarrow$	$\stackrel{\text{Matching}}{\longrightarrow} P_c^{\circ}(\phi_c) = \frac{1}{(\phi_c)}$	$\frac{A_1\beta^{4/3}}{(-\phi_1)^{7/3}}$
$N_f P_f = CJ_1$	$8\pi (\phi_k - \phi_2)^3$ $N_k^{\circ} (\phi_k) = 8\pi (\phi_k - \phi_2)^3$	$N_{\rm k}^{\rm o}(\phi_{\rm k}) = \frac{\phi_{\rm k}^{7/3}}{A_1}$	$N_{\rm C}^{\rm o}(\phi_{\rm C}) = \frac{\langle \phi_{\rm C} \rangle}{\langle \phi_{\rm C} \rangle}$	$\frac{-\phi_1}{A_1\beta^{1/3}}$
$P_{f} = \frac{c u_{d}}{8\pi} \frac{S_{1}^{2}}{D_{1}} \frac{1}{(f - f_{2})^{3}}$		$c_{2-} (\varepsilon_{t}\tau^{3})^{1/2} \ll r$	$c S_1, D_1$	$\begin{array}{ccc} 2 & \begin{bmatrix} W_1, D_1, c, u_d \end{bmatrix} \\ \mathbf{c}^* & W_1 \end{array}$
$N_f = \frac{8\pi}{S_1} D_1 (f - f_2)^3$	Matching	$\overrightarrow{P_c^{\circ}(\phi_c)} = (8\pi)^{-1}$ $N_c^{\circ}(\phi_c) = (8\pi)$	$\frac{1}{(\phi_{c}-\phi_{1})^{-3}\beta^{2}} (\phi_{c}-\phi_{1})^{3}\beta^{-1}$	$f = \frac{1}{(D_1 c u_d)^{1/2}}$ $f_2 = f_g + \phi_1 f^\circ$

¹ See footnote of Table 3 for other notations. Variables $\tau \equiv x_1/u_d$, $S_1 \equiv J_1/u_d$ and $r^2 = x_2^2 + x_2^2$ are used in this table. u_d is the characteristic

slip velocity; x_1 is longitudinal and x_2 , x_3 are the transversal coordinates in a droplet wake. ¹ The pdf is proportional to the droplets concentration and the length of the wake $P_t/(cu_d)$, so that the framed parameters determine $P_t/(cu_d)$. The constants A_1 , ϕ_1 , ϕ_2 and the functions P_k^0 , N_k^0 , P_c^0 , N_c^0 are universal but they may be different from Table 3,

FIGURES



Fig. 1. Scatter plots of all data and conditional averages conditional on mixture fraction for the temperature and OH mass fraction in a piloted diffusion flame of methanol at $x/D_j = 20$, $U_j = 60$ m/s and $t/D_j = 1.7$ (\bigcirc), 1.4 (\square), 1.1 (\triangle). The full curves are for a laminar counterflow diffusion flame with a strain rate parameter $a = 5 \text{ s}^{-1}$. Data of Masri et al. [86].



Fig. 2. Clipped Gaussian pdfs with various values of mean mixture fraction (first column) and root mean square fluctuation of mixture fraction (second column). The delta functions at $\eta = 0.1$ have strengths γ_1 , γ_2 of 0.048; 0.048; 0.21, 0.0; and 0.07, 0.0, respectively.



Fig. 3. Beta function pdfs with the same mean and root mean square fluctuation of mixture fraction as in Fig. 2. Note the different behaviour near $\eta = 0, 1$ and the absence of delta functions.



Fig. 4. Probability of the event $Z_1 < Y < Z_2$ shown (a) using the cumulative probability function and (b) using the pdf.



Fig. 5. Probability of the event $Z < Y < Z + \Delta Z$.



Fig. 6. Surface plot of a Gaussian joint pdf for two variables.









Fig. 9. Behaviour of conditional scalar dissipation with smooth pdfs. (a) Unbounded Gaussian pdf; (b) smooth pdf with fixed bounds; (c) smooth pdf with moving bounds.



Fig. 10. Qualitative behavior of the correlation function K_{ξ} .



Fig. 11. Pdfs $\vec{P}(\vec{\eta})$ in the wake of a heated cylinder versus $\vec{\eta} \equiv \Delta T / \Delta T_c$ for various transverse locations. Adapted from Klimenko et al. [63].



Fig. 12. Normalised pdf integral across the flow, $\hat{P}(\hat{\eta})$, Adapted from Klimenko et al. [63].



Fig. 13. Normalised conditional scalar dissipation integral across the flow. $\hat{N}(\hat{\eta})$. Adapted from Klimenko et al. [63].



Fig. 14. Integrated conditional scalar dissipation, N^a, measured in turbulent jet diffusion flames of air-diluted methane. Adapted from Starner et al. [129].



Fig. 15. Schematic diagram of the reactive scalar mixing layer. Adapted from Bilger et al. [16].



Fig. 16. Conditional averages of reactant concentration conditional on mixture fraction as measured in a reacting scalar mixing layer with $N_D = 0.42$ and $x_1/M = 21$. Different symbols are used for different points across the flow. Adapted from Bilger [12].



Fig. 17. Predictions for the conditional average reactant concentration in a reacting scalar mixing layer as a function of η and normalized downstream distance $\hat{x}_1 = N_D x_1 / M$ using Eq. (219) with A = 0.03, $\eta_s = 0.5$. Adapted from Bilger [12].



Fig. 18. Conditional averages of reactant concentration at $\eta = 0.5$ in a reacting scalar mixing layer. Curves are for predictions using Eq. (219) with various values of A. Data points from experiments of Bilger et al. [16]: Δ , $N_D = 0.42$; \Box , $N_D = 2.6$. Figure adapted from Bilger [12].



Fig. 19. CMC predictions (full lines) of Smith et al. [125] for nitric oxide compared with laser induced fluorescence measurements (dots) of Barlow and Carter [2].



Fig. 20. CMC predictions (full lines) of Smith et al. [125] for nitric oxide compared with conditionally averages of measurements (connected circles) of Barlow and Carter [2].



Fig. 21. CMC predictions (Smith et al. [125]) and laser measurements (Barlow and Carter [2]) of conditional average temperature in a hydrogen jet diffusion flame: $x_1 = \ell_f/2 - -$ predictions; -O-O-O-measurements. $x_1 = \ell_f/8 - \cdots$ predictions; -O-• measurements.



Fig. 22. Comparison of conditionally averaged NO mole fractions at $x_1 = \ell_f$ calculated from CMC models with 26 step and 2-step reduced mechanisms, with and without radiation losses. Bold/plain lines denote full/reduced mechanism calculations whilst solid/broken lines denote radiative/adiabatic calculations [125].



Fig. 23. Schematic diagram for experimental set-up for round turbulent reacting plume [23].



Fig. 24. Experimental results and CMC predictions in a reacting turbulent round plume of NO into background O₃: - - - conditional mean of data; - - - CMC predictions; ... reaction dominated limit [25].



Fig. 25. Schematic of an axisymmetric combustor [95] dimensions are in millimeters.



Fig. 26. Mean streamlines calculated for the combustor of Fig. 25 [95].



Fig. 27. Contours of Favre-averaged mixture fraction. $f \equiv \langle \rho \xi \rangle / \langle \rho \rangle$, calculated for the combustor of Fig. 25 [95].



Fig. 28. Contours of Favre averaged variance of mixture fraction, $g = \langle \rho \xi'^2 \rangle / \langle \rho \rangle$, for the combustor of Fig. 25 [95].



Fig. 29. Core and outlet averaged pdfs for an ISR model of the recirculation zone end of the combustor of Fig. 25 [95].



Fig. 30. Core-averaged conditional scalar dissipation for the ISR model [95] (full line) compared with laminar counterflow result with the same maximum (dashed line).



Fig. 31. Predictions for the conditional average species mass fractions in the recirculation zone of the combustor of Fig. 25 using the ISR model [95].



Fig. 32. Predictions for the conditionally averaged temperature and NO mass fraction in the recirculation zone of the combustor of Fig. 25 using the ISR model [95].



Fig. 33. Comparison between ISR predictions with the Miller-Bowman [94] (solid lines) as well as the GRI [40] (dotted lines) mechanisms and measured data (symbols) [42]. R = 30.5 mm is the radius of the confining tube.



Fig. 34. Schematic of a dilution flow reactor.



Fig. 35. Comparison of predicted and experimental results: serie parallel reaction of Li and Toor [81].



Fig. 36. Pdfs of the mixture fraction for the spatially homogeneous case [92]: (a) $\langle \xi \rangle = 0.25$; (b) $\langle \xi \rangle = 0.5$.



Fig. 37. The value of $\langle N | \eta \rangle / \langle N \rangle$ from the DNS data (symbols) and from the Case (b) model normalised by $\langle N \rangle$ from the DNS at times t = 0.9, 2.3, 3.7. Spatially homogeneous case [92]. (a) $\langle \xi \rangle = 0.25$; (b) $\langle \xi \rangle = 0.5$.



Fig. 38. Variation of the unconditional scalar dissipation with time [92].



Fig. 39. Time evolution of the terms on the left-hand side of the CMC equation, Eq. (160), expressed as a fraction of the reaction rate term. Spatially homogenous case, $\langle \xi \rangle = \xi_s = 0.25$. (a) $Da_0 = 0.5$, (b) $Da_0 = 8$. Results of Mell et al. [92].


Fig. 40. Predictions for normalized conditional variance in a reacting turbulent scalar mixing layer, Eq. (298), for $\xi_s = 0.5$, From Li and Bilger [77].



Fig. 41. Predictions for conditional variance in a reacting turbulent scalar mixing layer compared with experiment [77]. (a) $x_1 l_{\mathcal{M}} = 8 x_1 l_{\mathcal{M}} = 12$.



Fig. 42. Balance of terms in the conditional variance equation for the fuel species in non-premixed reaction in decaying homogeneous turbulence [132]. T_i refers to the *i*th term as it appears in Eq. (199). (a) Simulation R1 with $Re_{\lambda} = 33.5$; (b) simulation R5 with $Re_{\lambda} = 50.5$.



Fig. 43. Predictions for conditional average mass fraction of NO in a hydrogen jet diffusion flame using a partial second order conditional moment closure [69]. Filled symbols are the conditional averaged experimental data of Barlow and Carter [2] with the bars indicating plus/minus two-conditional standard deviations. Triangles are predictions with first order closure and squares for second order closure. (a) $x/L_v = 0.125$ and 0.5; (b) $x/L_v = 0.75$.



Fig. 44. Predictions and DNS data for conditional average and root mean square fluctuation of normalised temperature rise, θ , for autoignition in turbulent non-premixed flow [87]. t_1 is at $0.64\tau_h$ and t_2 is at $1.41\tau_h$ where τ_h is the shortest time to ignition of a uniform mixture. Ignition occurs at $2.05\tau_h$.



Fig. 45. Scatterplot of differential diffusion, z, versus mixture fraction, ξ , and conditional average, Q_z , versus η for mixing in decaying homogeneous turbulence [67].



Fig. 46. Qualitative structure of the interdroplet field [62]. (a) Non-inertial droplets: (b) inertial droplets.