

**The Transported Probability Density Function Approach
For Predicting Turbulent Combusting Flows**

by
M. KAKHI BEng, ACGI

Thesis submitted for the degree of
Doctor of Philosophy
in the faculty of Engineering
University of London
and for the
Diploma of Membership of Imperial College

Department of Chemical Engineering,
Imperial College of Science, Technology and Medicine
London

September 26, 1994



Abstract

The work described in this thesis focused on the use of the transported scalar probability density function (pdf) formalism for predicting turbulent reacting flows. The configuration of the turbulent jet diffusion flame was investigated, and the predictions were compared against experimental measurements of the velocity and scalar fields in such flows.

The calculation procedure employed a phenomenological turbulence model (k - ε and full Reynolds stress) to approximate the evolution of the velocity field. The governing equations for the velocity field were solved using a well-established finite-difference solver for boundary layer flows. The scalar field was evaluated through a Monte Carlo simulation of the joint scalar pdf equation. The pdf equation uses the coalescence-dispersion mixing model and the linear mean square estimation (LMSE) closure to represent the effects of molecular mixing on the pdf. Furthermore, global and systematically reduced reaction schemes were employed to describe the behaviour of the reactive scalar field. In order to examine the behaviour of the mixing models, studies in homogeneous, isotropic turbulence were carried out. The reduced schemes were investigated in a confined laminar jet diffusion flame geometry.

The comparison of the predictions and measurements of the turbulent jet diffusion flame using the global reduced scheme indicated overpredictions in the levels of carbon monoxide and hydrogen; this finding was consistent with the predictions of the laminar flame when assuming equal diffusivity of the scalars. However, many of the gross flame characteristics (e.g. temperature, fuel, water and CO₂ concentrations) were accurately predicted; in certain instances only the peak levels were accurately reproduced. It was found that in general the two mixing models yielded similar results, except in the prediction of jet flames where appreciable finite-rate effects (e.g. local extinction with subsequent downstream relighting) were experimentally observed. In these latter flows, the coalescence-dispersion mixing model failed to capture the effects of the local extinction whilst the LMSE closure predicted extinction with no re-lighting further downstream. It was also observed that the results were sensitive to the model 'constants' appearing in the equation for the viscous dissipation rate and the molecular mixing closures for the pdf equation. Difficulties were encountered when using the systematically reduced scheme; global extinction was consistently predicted by this mechanism for jet flows where such effects were not observed.

Acknowledgements

I whole-heartedly acknowledge the efforts of Dr. W.P. Jones, my Ph.D supervisor; his encouragement and constructive criticism throughout the course of the research served as a constant source of inspiration. I believe that his (very) patient disposition is possibly his finest attribute, and I have benefited considerably from the many discussions that we have had.

I would also like to take the opportunity to acknowledge the members of the Combustion Group, in particular Dr. A.R. Jones who had the misfortune of sitting through my numerous (and repetitive) presentations. At the same time I present my apologies to the members for my rather feeble record of attendance at Combustion Group meetings.

The financial support of Rolls-Royce plc was gratefully appreciated during the period of the research.

In retrospect the Ph.D was anything but an easy ride and it was not always a barrel of laughs. However, whilst writing the acknowledgements section of my thesis I try to remember the good times, and in this regard I am delighted to say that I have had more than my fair share. I would like to express my gratitude to Mr. M. Wille ('Matti'); besides being a first-class friend and colleague, Matti had on many occasions the decency to take on tasks within our group which the rest of us were either too bone-idle (or simply too incompetent) to deal with. His role as a $\text{\LaTeX}$ guru proved to be invaluable when I started to prepare this document. I would also like to express my gratitude to past members of this group with whom I shared (and still share) very enjoyable moments. In particular Drs. A.J. Marquis, P. Musonge and D.H. Sheen, and Messrs. M. Distelhoff, S. Ledin, A. Tober and M. Uhlmann. It is very gratifying to see that the new additions to our group, Messrs. N. Branley and Y. Prasetyo, have adopted the methods and traditions of our working environment so well, i.e. one of working and playing hard (sometimes simultaneously).

Our group suffered an enormous emotional blow when we received news of the death of one of our most appreciated (and celebrated) colleagues, Dr. J. Alvarez. It is not easy for me to write any more about this; suffice it to say that we all miss him dearly.

It seems only fair to put in a small word concerning the Particle Technology group. They are an interesting(?) bunch of people, especially Mr. S. Manimaaran ('Manny') - hang in there Manny, I'm sure you'll get what you really want *in the end*. I do not know how we can ever forgive the Particle Technology group for turfing us out of room 425, and then subsequently from the fourth floor of the Chemical engineering department - you bullies!

Last (but definitely not least) I present my most sincere thanks to my family: Rana, Nakissa (and Ali) and Morteza. In the case of my parents, for fear of trivializing their contribution any further through a set of clichés, I will simply point out that I will never forget your efforts - I dedicate my thesis to you both.

Contents

<i>List of Figures</i>	11
<i>List of Tables</i>	21
<i>Nomenclature</i>	23
1 Introduction	27
1.1 Preamble	27
1.2 Objectives of the Research	29
2 The Governing Equations of Turbulent Combusting Flows	31
2.1 Introduction	31
2.2 The Instantaneous Equations	31
2.2.1 Mass Continuity	31
2.2.2 Momentum Conservation Equation	32
2.2.3 Energy Conservation Equation	32
2.2.4 Species Conservation Equation	34
2.3 The Averaging Process	35
2.4 The Energy Cascade Viewpoint	37
2.5 Turbulence Models	38
2.5.1 Eddy-Viscosity Models	39
2.5.2 Second Moment Closures	42
2.5.2.1 Transport Terms	43
2.5.2.2 Viscous Destruction of $\widetilde{u_i'' u_j''}$	43
2.5.2.3 Fluctuating Pressure Terms	44
2.5.2.4 Scalar Variance Dissipation Rate	46
2.6 Mean Reaction Rates	47
3 Combustion Models for Non-Premixed Configurations	49
3.1 Introduction	49
3.2 The Conserved Scalar Formalism	50
3.3 Thermochemical Models	51
3.3.1 The Flame Sheet Approximation	51
3.3.2 The Chemical Equilibrium Approach	51
3.3.3 The Laminar Flamelet Approach	52
3.4 The Presumed-Shape Pdf Method	53

3.5	Applications of the Flamelet Technique with Flame Stretch	55
4	The Transported Pdf Approach	57
4.1	Introduction	57
4.2	Derivation of the Scalar Pdf Equation	57
4.3	Terms Appearing in the Pdf Equation	59
4.4	General Solution Features of Pdf Transport Equations	60
4.5	Basic Modelling Requirements of the Molecular Mixing Term	61
4.6	The Linear Mean Square Estimation (LMSE) Closure	62
4.7	Coalescence-Dispersion Mixing Models	63
4.7.1	Curl's Mixing Model	63
4.7.2	Modified Curl	65
4.7.3	Age-Biased Coalescence-Dispersion Model	67
4.7.4	Reaction Zone Conditioning	68
4.8	Langevin Models	69
4.9	Binomial Models	70
4.10	Mapping Closures	72
4.11	Closing Remarks Concerning Mixing Models	73
4.12	Models for the Turbulent Transport of the Pdf	74
4.13	The Monte Carlo Method for Solving the Scalar Pdf Equation	74
5	Details of the Calculation Procedure	79
5.1	Introduction	79
5.2	The Patankar-Spalding Method	79
5.2.1	The Basic Geometry of the Jet Configuration	79
5.2.2	The General form of the Equations	79
5.2.3	The Choice of Coordinate System	80
5.2.4	The Finite-Difference Approximations and Numerical Solution Method	82
5.3	The Treatment of the Scalar Field	83
5.3.1	The Details of the Thermochemistry	83
5.3.1.1	The Systematically Reduced Scheme of Chelliah, Seshadri and Law	84
5.3.1.2	The Global Alkane Hydrocarbon Mechanism of Jones and Lindstedt	85
5.3.1.3	Tabulation of the Thermochemistry	87
6	Results and Discussion	93
6.1	Introduction	93
6.2	Predictions of a Confined Laminar Jet Diffusion Flame	94
6.3	The Calculations of Statistically Homogeneous, Isotropic Turbulence	99
6.3.1	Non-Reacting Test Cases	99
6.3.2	Reacting Test Cases	100
6.3.2.1	Results using the Global Hydrocarbon Mechanism	101
6.3.2.2	Results using the Systematically Reduced Mechanism	102
6.4	Predictions of Propane-Fuelled Turbulent Jet Diffusion Flames	104

6.4.1	The Flame of Godoy	104
6.4.2	The Flame of Heitor, Moreira and Pires	110
6.5	Predictions of Methane-Fuelled Turbulent Jet Diffusion Flames	111
6.5.1	The Flame of Hassan, Lockwood and Moneib	111
6.5.2	The Flame of Masri, Bilger and Dibble	115
7	Closure	125
7.1	Summary	125
7.2	Conclusions	126
7.2.1	Laminar Flame Calculations	126
7.2.2	Homogeneous, Isotropic Turbulence	126
7.2.3	Turbulent Jet Diffusion Flames	127
7.3	Recommendations for Future Work	129
	<i>References</i>	<i>131</i>
A	Derivation of the Single-Point, Single-Time Scalar Pdf Equation	139
B	The Basic Equations that are Solved	143
B.1	Mass Continuity	143
B.2	Momentum Equation	143
B.2.1	The k - ϵ Model	144
B.2.2	The Reynolds Stress Closure	144
B.3	Scalar Pdf Transport Equation	145
C	The Transformation of the Equations to the (x, ω) Coordinate System	147
D	The Finite-Difference Procedure	149
D.1	Preliminary Remarks	149
D.2	Finite-Difference Approximations for the Internal Nodes	150
D.2.1	Axial Convection: I	151
D.2.2	Cross-Stream Convection: II	151
D.2.3	Cross-Stream Diffusion: III	152
D.2.4	Source Term: IV	153
D.2.5	The Finite-Difference Equation	155
D.3	The Treatment of the Boundaries	155
D.4	The High Lateral Flux Modification	157
D.5	The Calculation of the Cross Stream Distance	158
D.6	The Treatment of Confined Flows: Calculating $\frac{dp}{dx}$	159
E	Implementation of the Scalar Pdf Equation	161
E.1	Transport	161
E.2	Molecular Mixing	165
E.2.1	Modified Curl	165
E.2.2	Linear Mean Square Estimation (LMSE) Closure	166
E.3	Simulation of Homogeneous, Isotropic Turbulence	167

E.4	Evaluation of the Statistical Properties from the Discrete Ensemble	168
F	The Determination of the Scalar Bounds for Tabulating the Chemistry	171
F.1	Introduction	171
F.2	Composition Bounds for the Global Hydrocarbon Scheme	171
F.2.1	Element Conservation	171
F.2.2	Bounds for the Independent Scalars	173
F.2.2.1	Bounds for the Mixture Fraction	173
F.2.2.2	Bounds for $n_{C_xH_y}$ given ξ	173
F.2.2.3	Bounds for n_{CO} given ξ and $n_{C_xH_y}$	174
F.2.2.4	Bounds for n_{H_2} given ξ , $n_{C_xH_y}$ and n_{CO}	175
F.2.3	Determination of the Dependent Scalar Concentrations	177
F.2.4	Modifications for Flames with Nitrogen-Diluted Fuel Streams	177
F.3	Composition Bounds for the Systematically Reduced Methane/Air Scheme	178
F.3.1	Conservation of Atomic Species	178
F.3.2	Bounds for the Independent Scalars	178
G	The Stoichiometric Mixture Fraction of a Hydrocarbon	181
H	Details of the Thermochemistry	183
H.1	Introduction	183
H.2	Determination of the Mixture Temperature	183
H.3	Determination of the Equilibrium Constant	184
H.4	Example: Determination of a Reverse Rate Using the Global Scheme	187
	Figures Relating to Calculations	191

List of Figures

3.1	a.) Schematic of a laminar counterflow diffusion flame geometry. The width of the flame is exaggerated for clarity. b.) Locally the flame is treated as planar. . . .	53
4.1	Schematic of the mixing process based on coalescence-dispersion closures showing how a particle mixes and thereby crosses the reaction zone without reacting. T_{lim} is the temperature below which the reaction rates are negligibly low. . . .	68
4.2	Schematic of representative particles. Random sampling without replacement from x_{i-1} , x_i and x_{i+1} leads to the new ensemble at x_i . The shading of the particles only identifies where the particles originate from. In this notation, x refers to the coordinate direction, and the subscript i identifies the mesh position in that direction ($i = 1, \dots, N + 1$). . . .	76
5.1	Schematic of a jet flow. Ψ denotes the stream function and since only the half-jet is calculated (due to the axisymmetry of the flow), the I and E surfaces represent the required solution domain. . . .	80
5.2	Schematic of the two possible coordinate systems which can be used to track the evolution of the boundary layer. . . .	81
6.1	Schematic of the confined, axisymmetric, laminar CH_4 -air jet diffusion flame investigated by Mitchell et al. [69]. $D = 1.27\text{cm}$ and $D_o = 5.08\text{cm}$. Air and fuel are admitted at 187.7 cc/s and 5.7 cc/s respectively. . . .	94
6.2	Schematic of the piloted methane-air turbulent jet diffusion flame of Masri et al. [66]. The streams labelled 1, 2 and 3 contain the following: 1. CH_4 at 298K , $Re \approx 20000$, $u_{in} = 41\text{ms}^{-1}$ (flame L) and $u_{in} = 48\text{ms}^{-1}$ (flame B); 2. 'Hot' (fully) burnt products from a stoichiometric mixture of C_2H_2 , H_2 and air, 2600K , $Re \approx 604$, $u_{in} = 24\text{ms}^{-1}$; 3. Coflowing air at 298K , $Re \approx 10^6$, $u_{in} = 15\text{ms}^{-1}$. .	115
6.3	This figure illustrates the effect of introducing the enthalpy as an extra scalar in the manner proposed by Sion and Chen [86]. The 'original' temperature allows for a peak (stoichiometric) value of 2400K , whilst the addition of the 'elevated' enthalpy raises this to 2600K . Extra compositional states are also introduced which need to be accounted for when constructing the bounds for the interpolation table.	123

- D.1 Schematic of the typical computational molecule and control volume used to derive the difference approximations. The effect of the I and E boundaries are not considered at this stage. The notation ϕ_i^n denotes the value of the variable ϕ at node i ($1 \leq i \leq N+1$) and $x = x_n$. $\omega_{i+\frac{1}{2}} \equiv \frac{1}{2}(\omega_i + \omega_{i+1})$; this implies that the control volume surface lies midway between grid nodes irrespective of whether or not the mesh spacing is uniform. 149
- D.2 Schematic of the control volumes used to derive the difference approximations at the I and E boundaries. Note, $\omega_3 - \omega_2 = 2(\omega_2 - \omega_1)$; similarly $\omega_N - \omega_{N-1} = 2(\omega_{N+1} - \omega_N)$. The boundary can be a wall, an axis of symmetry or a free stream. 156
- D.3 Schematic of the control volumes along a given radial plane, indicating their relative placement and the associated notation. Because the finite-differencing is *conservative*, the fluxes leaving the top face of the first control volume is identically equal to the fluxes entering the bottom face of the second; i.e. $F_{2+\frac{1}{2}} = F_{3-\frac{1}{2}}$ and $G_{2+\frac{1}{2}} = G_{3-\frac{1}{2}}$ 158
- E.1 Schematic of the mixing operation using modified Curl. The randomly selected particles are represented in terms of the properties $\phi_1(t)$ and $\phi_2(t)$. They mix along a line joining them in composition space and jump to the new properties $\phi_1(t + \delta t)$ and $\phi_2(t + \delta t)$. The extent of mixing is constrained by either no change in their original values, to full mixing at their common mean value. 166
- 6.4 Initial conditions for ξ , CH_4 , CO , axial velocity, CO_2 and temperature specified at the first measuring station ($x = 12\text{mm}$) of the flame of Mitchell et al. [69]. . . . 192
- 6.5 Initial conditions for H_2 , H_2O , O_2 and N_2 specified at the first measuring station ($x = 12\text{mm}$) of the flame of Mitchell et al. [69]. 193
- 6.6 Initial conditions for H_2 , H_2O , O_2 , N_2 , CH_3 and H specified at the first measuring station ($x = 12\text{mm}$) of the flame of Mitchell et al. [69]. The effect of specifying radical species in the initial condition is investigated. 194
- 6.7 Radial profiles of N_2 , CH_3 and H from the calculation of the laminar flame of Mitchell et al. [69] using the reduced scheme of Chelliah et al. [10]; the interpolation table is *not* used. The effect of the two initial conditions specified in figure 6.6 is investigated. 195
- 6.8 Radial profiles of ξ , CH_4 and CO from the calculation of the laminar flame of Mitchell et al. [69] using the reduced schemes of Jones and Lindstedt [46] and Chelliah et al. [10]; the interpolation table is *not* used. 196
- 6.9 Radial profiles of H_2 , H_2O and O_2 from the calculation of the laminar flame of Mitchell et al. [69] using the reduced schemes of Jones and Lindstedt [46] and Chelliah et al. [10]; the interpolation table is *not* used. 197
- 6.10 Radial profiles of N_2 , CH_3 and H from the calculation of the laminar flame of Mitchell et al. [69] using the reduced schemes of Jones and Lindstedt [46] and Chelliah et al. [10]; the interpolation table is *not* used. 198
- 6.11 Radial profiles of axial velocity, CO_2 and temperature from the calculation of the laminar flame of Mitchell et al. [69] using the reduced schemes of Jones and Lindstedt [46] and Chelliah et al. [10]; the interpolation table is *not* used. . . . 199

6.12	Radial profiles of H_2 , H_2O and O_2 from the calculation of the laminar flame of Mitchell et al. [69] using the reduced scheme of Chelliah et al. [10]; the results from LSODE and the 'look-up' (interpolation) table are compared.	200
6.13	Radial profiles of N_2 , CH_3 and H from the calculation of the laminar flame of Mitchell et al. [69] using the reduced scheme of Chelliah et al. [10]; the results from LSODE and the 'look-up' (interpolation) table are compared.	201
6.14	Radial profiles of axial velocity, CO_2 and temperature from the calculation of the laminar flame of Mitchell et al. [69] using the reduced scheme of Jones and Lindstedt [46]; the results from LSODE and the 'look-up' (interpolation) table are compared.	202
6.15	Radial profiles of axial velocity, CO_2 and temperature from the calculation of the laminar flame of Mitchell et al. [69] using the reduced scheme of Chelliah et al. [10]; the results from LSODE and the 'look-up' (interpolation) table are compared.	203
6.16	Predictions of the passive scalar statistics in homogeneous, isotropic and inert turbulence. Modified Curl is employed with a double δ -function initial condition and with two characteristic turbulent time scales ($\equiv \frac{k}{\epsilon}$).	204
6.17	Predictions of the passive scalar statistics in homogeneous, isotropic and inert turbulence. LMSE is employed with a double δ -function initial condition and with two characteristic turbulent time scales ($\equiv \frac{k}{\epsilon}$).	205
6.18	Passive scalar statistics in homogeneous, isotropic and inert turbulence. Modified Curl is employed with a double δ -function initial condition and with two characteristic turbulent time scales ($\equiv \frac{k}{\epsilon}$). The evolution of the individual moments are shown.	206
6.19	Passive scalar statistics in homogeneous, isotropic and inert turbulence. LMSE is employed with a double δ -function initial condition and with two characteristic turbulent time scales ($\equiv \frac{k}{\epsilon}$). The evolution of the individual moments are shown.	207
6.20	The same conditions as in figure 6.18 are employed. The behaviour of the individual moments in the final stages of mixing are expanded on to show why 'wiggles' appear in the plots of the normalised moments.	208
6.21	The same conditions as in figure 6.20 are employed, but with LMSE.	208
6.22	Evolution of the pdf for a passive scalar in homogeneous, isotropic and inert turbulence. Modified Curl is employed with a double δ -function initial condition and with $\tau(\equiv \frac{k}{\epsilon}) = \tau_o + (C_{\epsilon_2} - 1) t$, where $\tau_o = 0.04s$	209
6.23	Evolution of the pdf for a passive scalar in homogeneous, isotropic and inert turbulence. LMSE is employed with a double δ -function initial condition and with $\tau(\equiv \frac{k}{\epsilon}) = \tau_o + (C_{\epsilon_2} - 1) t$, where $\tau_o = 0.04s$	210
6.24	Predictions of the density-weighted mean reactive scalar properties in homogeneous, isotropic turbulence. Modified Curl and LMSE are compared starting with a double δ -function condition and $\tau(\equiv \frac{k}{\epsilon}) = \tau_o + (C_{\epsilon_2} - 1) t$, where $\tau_o = 0.04s$. The reduced scheme of Jones and Lindstedt [46] is used <i>without</i> the interpolation table. $\tilde{\xi} = \xi_{st} = 0.05$ (for CH_4).	211
6.25	Predictions of the density-weighted mean reactive scalar properties in homogeneous, isotropic turbulence. The evolution of the initial δ -functions is monitored for the case of LMSE to explain the behaviour of this mixing model in figure 6.24).	212

- 6.26 Predictions of the density-weighted mean reactive scalar properties in homogeneous, isotropic turbulence. Modified Curl and LMSE are compared starting with a double δ -function condition and $\tau(\equiv \frac{k}{\varepsilon}) = \tau_o + (C_{\varepsilon_2} - 1) t$, where $\tau_o = 0.04s$. The reduced scheme of Jones and Lindstedt [46] is used *with* the interpolation table. $\tilde{\xi} = \xi_{st} = 0.05$ (for CH₄). 213
- 6.27 Evolution of $\tilde{P}(\tilde{\xi}; t)$ in homogeneous, isotropic turbulence. Modified Curl is employed with a double δ -function initial condition and with $\tau(\equiv \frac{k}{\varepsilon}) = \tau_o + (C_{\varepsilon_2} - 1) t$, where $\tau_o = 0.04s$. The reduced scheme of Jones and Lindstedt [46] is used *without* the interpolation table. $\tilde{\xi} = \xi_{st} = 0.05$ (for CH₄). 214
- 6.28 Evolution of $\tilde{P}(\tilde{T}; t)$ in homogeneous, isotropic turbulence. Modified Curl is employed with a double δ -function initial condition and with $\tau(\equiv \frac{k}{\varepsilon}) = \tau_o + (C_{\varepsilon_2} - 1) t$, where $\tau_o = 0.04s$. The reduced scheme of Jones and Lindstedt [46] is used *without* the interpolation table. $\tilde{\xi} = \xi_{st} = 0.05$ (for CH₄). 215
- 6.29 Predictions of the density-weighted mean reactive scalar properties in homogeneous, isotropic turbulence. Modified Curl and LMSE are compared starting with a continuous distribution taken from figure 6.27 at $t/\tau_o = 5$, where $\tau_o = 0.04s$. The reduced scheme of Jones and Lindstedt [46] is used *without* the interpolation table. $\tilde{\xi} = \xi_{st} = 0.05$ (for CH₄). 216
- 6.30 Predictions of the density-weighted mean reactive scalar properties in homogeneous, isotropic turbulence. Modified Curl is employed with a triple δ -function initial condition and $\tau(\equiv \frac{k}{\varepsilon}) = \tau_o + (C_{\varepsilon_2} - 1) t$, where $\tau_o = 0.04s$. The reduced scheme of Chelliah et al. [10] is used *without* the interpolation table. $\tilde{\xi} = \xi_{st} = 0.05$ (for CH₄). 217
- 6.31 Predictions of the density-weighted mean reactive scalar properties in homogeneous, isotropic turbulence. LMSE is employed with a triple δ -function initial condition and $\tau(\equiv \frac{k}{\varepsilon}) = \tau_o + (C_{\varepsilon_2} - 1) t$, where $\tau_o = 0.04s$. The reduced scheme of Chelliah et al. [10] is used *without* the interpolation table. $\tilde{\xi} = \xi_{st} = 0.05$ (for CH₄). 218
- 6.32 Predictions of the density-weighted mean reactive scalar properties in homogeneous, isotropic turbulence. Modified Curl and LMSE are compared starting with a continuous distribution taken from figure 6.27 at $t/\tau_o = 5$, where $\tau_o = 0.04s$. The reduced scheme of Chelliah et al. [10] is used *without* the interpolation table. $\tilde{\xi} = \xi_{st} = 0.05$ (for CH₄). 219
- 6.33 Predictions of the density-weighted mean reactive scalar properties in homogeneous, isotropic turbulence. Same conditions are used as in figure 6.30 except for the air temperature which is now 298K instead of 800K. LSODE is employed. 220
- 6.34 Predictions of the density-weighted mean reactive scalar properties in homogeneous, isotropic turbulence. Same conditions are used as in figure 6.30 except for the air temperature which is now 298K instead of 800K. The interpolation table is employed. 221
- 6.35 Predictions of $\tilde{\xi}$ in a non-reacting, constant density air-jet. The jet pipe diameter and mean velocity are 6mm and 19.9m s⁻¹ respectively. A comparison of the finite-difference and Monte Carlo simulation is shown, using the k - ε turbulence model. The plots on the left employ modified Curl (for the Monte Carlo simulation) and LMSE on the right. 222

- 6.36 Predictions of $\overline{\xi'^2}$ in a non-reacting, constant density air-jet. The jet pipe diameter and mean velocity are 6mm and 19.9m s^{-1} respectively. A comparison of the finite-difference and Monte Carlo simulation is shown, using the $k-\epsilon$ turbulence model. The plots on the left employ modified Curl (for the Monte Carlo simulation) and LMSE on the right. 223
- 6.37 Radial profiles of $\tilde{\xi}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\epsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. 224
- 6.38 Radial profiles of $\tilde{X}_{N_2}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\epsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. 225
- 6.39 Radial profiles of $\tilde{u}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\epsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. 226
- 6.40 Radial profiles of $\tilde{X}_{C_3H_8}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\epsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. 227
- 6.41 Radial profiles of $\tilde{X}_{O_2}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\epsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. 228
- 6.42 Radial profiles of $\tilde{X}_{CO_2}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\epsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. 229
- 6.43 Radial profiles of $\tilde{X}_{CO}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\epsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. 230
- 6.44 Radial profiles of $\tilde{X}_{H_2}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\epsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. 231
- 6.45 Radial profiles of $\tilde{X}_{H_2O}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\epsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. 232
- 6.46 Radial profiles of $\frac{k}{\epsilon}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\epsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. 233
- 6.47 Radial profiles of the turbulence kinetic energy (k) for the flame of Godoy [31] at $Re = 24000$. The $k-\epsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. 234
- 6.48 Radial profiles of the viscous dissipation rate of the turbulence kinetic energy (ϵ) for the flame of Godoy [31] at $Re = 24000$. The $k-\epsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. 235
- 6.49 This figure shows the axial evolution of the integral of the mixture fraction convection as defined in equation (6.17), for the flame of Godoy [31] at $Re = 24000$. The comparison of the integral with 5×10^4 and 1×10^6 particles is shown. The $k-\epsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. The result is only shown for modified Curl. 236

- 6.50 Radial profiles of $\tilde{\xi}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\varepsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. Calculations are reported for modified Curl using 5×10^4 (solid line) and 1×10^6 particles (dashed line). 237
- 6.51 Radial profiles of $\tilde{X}_{CO}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\varepsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. Calculations are reported for modified Curl using 5×10^4 (solid line) and 1×10^6 particles (dashed line). 238
- 6.52 Radial profiles of $\tilde{u}$ for the flame of Godoy [31] at $Re = 24000$. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. A comparison is made of the $k-\varepsilon$ and Reynolds stress turbulence models on the left and right hand sides, respectively. 239
- 6.53 Radial profiles of $\tilde{\xi}$ for the flame of Godoy [31] at $Re = 24000$. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. A comparison is made of the $k-\varepsilon$ and Reynolds stress turbulence models. 240
- 6.54 Radial profiles of $\tilde{X}_{CO}$ for the flame of Godoy [31] at $Re = 24000$. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. A comparison is made of the $k-\varepsilon$ and Reynolds stress turbulence models. 241
- 6.55 Radial profiles of $\tilde{\xi}$ for the flame of Godoy [31] at $Re = 42700$. The $k-\varepsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. 242
- 6.56 Radial profiles of $\tilde{X}_{CO_2}$ for the flame of Godoy [31] at $Re = 42700$. The $k-\varepsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. 243
- 6.57 Radial profiles of $\tilde{X}_{CO}$ for the flame of Godoy [31] at $Re = 42700$. The $k-\varepsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. 244
- 6.58 Predictions of $\tilde{X}_{CO}$ and $\tilde{X}_{CO_2}$ for the flame of Heitor et al. [37], burner 2 at $Re = 37000$. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. The $k-\varepsilon$ (solid line) and Reynolds stress (dotted line) turbulence models are used. Crosses and circles refer to the measurements at diametrically opposing locations. 245
- 6.59 Predictions of $\tilde{X}_{O_2}$ and $\tilde{T}$ for the flame of Heitor et al. [37], burner 2 at $Re = 37000$. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. The $k-\varepsilon$ (solid line) and Reynolds stress (dotted line) turbulence models are used. Crosses and circles refer to the measurements at diametrically opposing locations. 246
- 6.60 Predictions of $(\widetilde{v''^2})^{\frac{1}{2}}$, $\widetilde{u''v''}$ and $\tilde{u}$ for the flame of Heitor et al. [37], burner 2 at $Re = 37000$. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. The $k-\varepsilon$ (solid line) and Reynolds stress (dotted line) turbulence models are used. Crosses and circles refer to the measurements at diametrically opposing locations. 247

- 6.61 Predictions of $(\overline{u'^2})^{\frac{1}{2}}$ and $(\overline{T'^2})^{\frac{1}{2}}$ for the flame of Heitor et al. [37], burner 2 at $Re = 37000$. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. The $k-\varepsilon$ (solid line) and Reynolds stress (dotted line) turbulence models are used. Crosses and circles refer to the measurements at diametrically opposing locations. 248
- 6.62 Predictions of $\tilde{X}_{CO}$ and $\tilde{T}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. 249
- 6.63 Predictions of $\tilde{X}_{O_2}$ and $\tilde{X}_{CO_2}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. 250
- 6.64 Predictions of $\tilde{X}_{CH_4}$ and $\tilde{X}_{H_2}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. 251
- 6.65 Axial evolution of $\tilde{T}$, $(\overline{T'^2})^{\frac{1}{2}}$, $\tilde{X}_{CO_2}$, $\tilde{X}_{CO}$, $\tilde{X}_{O_2}$ and $\tilde{X}_{H_2}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. 252
- 6.66 Axial evolution of $\tilde{X}_{O_2}$ for the flame of Hassan et al. [35] in the burner stabilised configuration with and without N_2 -dilution (SA and NA flames). The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. 253
- 6.67 Axial evolution of $\tilde{X}_{CO_2}$ for the flame of Hassan et al. [35] in the burner stabilised configuration with and without N_2 -dilution (SA and NA flames). The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. 254
- 6.68 Axial evolution of $\tilde{X}_{CO}$ for the flame of Hassan et al. [35] in the burner stabilised configuration with and without N_2 -dilution (SA and NA flames). The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. 255
- 6.69 Radial profiles of $\tilde{\xi}$, $\tilde{T}$ and $\tilde{q}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Chelliah et al. [10] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. In this calculation the mixture is 'ignited' at $x/D = 15$ 256
- 6.70 Radial profiles of $\tilde{X}_{O_2}$, $\tilde{X}_{CO_2}$, $\tilde{X}_{CO}$ and $\tilde{X}_{H_2O}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Chelliah et al. [10] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. In this calculation the mixture is 'ignited' at $x/D = 15$ 257
- 6.71 Radial profiles of $\tilde{X}_{CH_4}$, $\tilde{X}_{H_2}$, $\tilde{X}_{CH_3}$ and $\tilde{X}_H$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Chelliah et al. [10] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. In this calculation the mixture is 'ignited' at $x/D = 15$ 258

- 6.72 Radial profiles of $\tilde{\xi}$ and $\bar{T}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Chelliah et al. [10] is employed, with modified Curl as the mixing model. $k-\varepsilon$ is used for the velocity field. In this figure, the effect of igniting the mixture at $x/D = 10$ and $x/D = 15$ is compared. 259
- 6.73 Radial profiles of $\tilde{X}_{CH_4}$ and $\tilde{X}_{O_2}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Chelliah et al. [10] is employed, with modified Curl as the mixing model. $k-\varepsilon$ is used for the velocity field. In this figure, the effect of igniting the mixture at $x/D = 10$ and $x/D = 15$ is compared. 260
- 6.74 Radial profiles of $\tilde{X}_{CH_3}$ and $\tilde{X}_H$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Chelliah et al. [10] is employed, with modified Curl as the mixing model. $k-\varepsilon$ is used for the velocity field. In this figure, the effect of igniting the mixture at $x/D = 10$ and $x/D = 15$ is compared. 261
- 6.75 Radial profiles of $\tilde{\xi}$ and $\bar{T}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Chelliah et al. [10] is employed, with modified Curl as the mixing model. $k-\varepsilon$ is used for the velocity field. In this figure, the results of the interpolation table are compared with using LSODE for the test case where ignition is performed at $x/D = 10$ 262
- 6.76 Radial profiles of $\tilde{X}_{CH_3}$ and $\tilde{X}_H$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Chelliah et al. [10] is employed, with modified Curl as the mixing model. $k-\varepsilon$ is used for the velocity field. In this figure, the results of the interpolation table are compared with using LSODE for the test case where ignition is performed at $x/D = 10$ 263
- 6.77 Radial profiles of $\tilde{Y}_{CH_4}$ and $\tilde{\xi}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$ 264
- 6.78 Radial profiles of $\bar{q}$ and $\tilde{Y}_{N_2}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$ 265
- 6.79 Radial profiles of $\tilde{Y}_{O_2}$ and $\tilde{Y}_{CO_2}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$ 266
- 6.80 Radial profiles of $\tilde{Y}_{CO}$ and $\bar{T}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$ 267
- 6.81 Radial profiles of $\tilde{Y}_{H_2O}$ and $\tilde{Y}_{H_2}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$ 268
- 6.82 Radial profiles of $(\widetilde{Y_{CH_4}''^2})^{\frac{1}{2}}$ and $(\widetilde{\xi''^2})^{\frac{1}{2}}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$. 269
- 6.83 Radial profiles of $(\overline{\varrho'^2})^{\frac{1}{2}}$ and $(\widetilde{Y_{N_2}''^2})^{\frac{1}{2}}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$. 270

- 6.84 Radial profiles of $(\widetilde{Y''^2_{O_2}})^{\frac{1}{2}}$ and $(\widetilde{Y''^2_{CO_2}})^{\frac{1}{2}}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$. 271
- 6.85 Radial profiles of $(\widetilde{Y''^2_{CO}})^{\frac{1}{2}}$ and $(\widetilde{T''^2})^{\frac{1}{2}}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$. 272
- 6.86 Radial profiles of $(\widetilde{Y''^2_{H_2O}})^{\frac{1}{2}}$ and $(\widetilde{Y''^2_{H_2}})^{\frac{1}{2}}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$. 273
- 6.87 Pdf profiles at $x/D = 20$ of $\tilde{P}(\hat{\xi}; x, \omega)$ for the (L) flame of Masri et al. [67]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. $k-\epsilon$ is used for the velocity field. 274
- 6.88 Pdf profiles at $x/D = 20$ of $\tilde{P}(\hat{\xi}; x, \omega)$ for the (L) flame of Masri et al. [67]. The reduced scheme of Jones and Lindstedt [46] is employed, with LMSE as the mixing model. $k-\epsilon$ is used for the velocity field. 275
- 6.89 Pdf profiles at $x/D = 20$ of $\tilde{P}(\hat{T}; x, \omega)$ for the (L) flame of Masri et al. [67]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. $k-\epsilon$ is used for the velocity field. 276
- 6.90 Pdf profiles at $x/D = 20$ of $\tilde{P}(\hat{Y}_{CO}; x, \omega)$ for the (L) flame of Masri et al. [67]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. $k-\epsilon$ is used for the velocity field. 277
- 6.91 Radial profiles of $\tilde{Y}_{CH_4}$ and $\tilde{\xi}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$ 278
- 6.92 Radial profiles of $\tilde{q}$ and $\tilde{Y}_{N_2}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$ 279
- 6.93 Radial profiles of $\tilde{Y}_{O_2}$ and $\tilde{Y}_{CO_2}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$ 280
- 6.94 Radial profiles of $\tilde{Y}_{CO}$ and $\tilde{T}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$ 281
- 6.95 Radial profiles of $\tilde{Y}_{H_2O}$ and $\tilde{Y}_{H_2}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$ 282
- 6.96 Radial profiles of $(\widetilde{Y''^2_{CH_4}})^{\frac{1}{2}}$ and $(\widetilde{\xi''^2})^{\frac{1}{2}}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$. 283
- 6.97 Radial profiles of $(\widetilde{\rho''^2})^{\frac{1}{2}}$ and $(\widetilde{Y''^2_{N_2}})^{\frac{1}{2}}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$. 284

- 6.98 Radial profiles of $\tilde{Y}_{\text{CH}_4}$ and $(\widetilde{Y_{\text{CH}_4}''^2})^{\frac{1}{2}}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model (solid line) and this is compared with a test case where chemical reaction is not performed (dashed line). $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$ 285
- 6.99 Radial profiles of $\bar{T}$ and $(\overline{T'^2})^{\frac{1}{2}}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model (solid line) and this is compared with a test case where chemical reaction is not performed (dashed line). $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$. 286
- 6.100 Radial profiles of $\tilde{Y}_{\text{CO}}$ and $\bar{T}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model; the dashed line incorporates enthalpy as an extra scalar, and this is compared with the solution (corresponding to the solid line) presented in figure 6.80. 287
- 6.101 Radial profiles of $\tilde{Y}_{\text{CO}}$ and $\bar{T}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. In these calculations, enthalpy is included as an extra scalar, and the result should be compared with figure 6.94. 288
- 6.102 Radial profiles of $\tilde{Y}_{\text{CH}_4}$ and $\tilde{\xi}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. A Reynolds stress model is used for the velocity field, with $C_{\varepsilon_2} = 1.8$. $r = \frac{D}{2} = 3.6\text{mm}$ 289
- 6.103 Radial profiles of $\tilde{Y}_{\text{CO}}$ and $\bar{T}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. A Reynolds stress model is used for the velocity field, with $C_{\varepsilon_2} = 1.8$. $r = \frac{D}{2} = 3.6\text{mm}$ 290
- 6.104 Radial profiles of $\tilde{Y}_{\text{CH}_4}$ and $\tilde{\xi}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with LMSE as the mixing model. A Reynolds stress model is used for the velocity field, with the solutions for $C_{\varepsilon_2} = 1.8$ (also shown in figure 6.102) and $C_{\varepsilon_2} = 1.9$ being compared. $r = \frac{D}{2} = 3.6\text{mm}$ 291
- 6.105 Radial profiles of $\tilde{Y}_{\text{CO}}$ and $\bar{T}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with LMSE as the mixing model. A Reynolds stress model is used for the velocity field, with the solutions for $C_{\varepsilon_2} = 1.8$ (also shown in figure 6.102) and $C_{\varepsilon_2} = 1.9$ being compared. $r = \frac{D}{2} = 3.6\text{mm}$ 292
- 6.106 Radial profiles of $\tilde{Y}_{\text{CO}}$ and $\bar{T}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with LMSE as the mixing model. The effect of C_D is investigated. $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$ 293
- 6.107 Radial profiles of $\tilde{Y}_{\text{CO}}$ and $\bar{T}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with LMSE as the mixing model. The effect of C_D is investigated. $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$ 294

List of Tables

2.1	Model constants appearing in the Reynolds stress and viscous dissipation rate equations.	46
5.1	Data required to construct the forward rate constants for the global reaction scheme	87
6.1	Experimental conditions for the propane-air jet diffusion flame of Godoy [31]. Burner diameter 6mm.	104
6.2	Stabilised natural gas diffusion flames, Hassan et al. [35]. $D = 7.74\text{mm}$. Burner velocity chosen to conform with the fuel mass flow rate, $\rho_{CH_4} = 0.656 \text{ Kg m}^{-3}$ and $\mu_{CH_4} = 1.121 \times 10^{-5} \text{ Kg m}^{-1}\text{s}^{-1}$ at 298K.	112
6.3	Stabilised natural gas/N ₂ -diluted diffusion flames, Hassan et al. [35]. $D = 7.74\text{mm}$. $Re \approx 16500$	112
D.1	The terms grouped into the diffusion coefficient and source terms in the finite-difference formulation are detailed. The equations are written down in appendix B. The value of the model constants are assigned in table 2.1 (page 46).	154

Nomenclature

a_r	Exponent of temperature in the evaluation of the rate constant, equation (2.18)
A_{ij}	Contribution to the fluctuating velocity-pressure gradient correlation term in the Reynolds stress equation
b_{ij}	Anisotropy tensor; $b_{ij} \equiv (\overline{u_i'' u_j''} / k) - (2/3)\delta_{ij}$
B_r	Pre-exponential factor of the r^{th} reaction step
$c_{p,\alpha}$	Specific heat at constant pressure of specie α
C_D	Ratio of the scalar to mechanical turbulence time scale, defined in equation (2.63)
C_α	Molar concentration of specie α (Kmol/m ³)
D	Diffusion coefficient
Da	Damköhler number, the ratio of an aerodynamic to reaction time scale e.g. $(\frac{k}{\epsilon}) \cdot (\frac{\nu}{S_l^2})^{-1}$
$\frac{d}{dt}$	Substantial derivative operator defined in terms of the unweighted mean velocity; $\frac{d}{dt} \equiv \frac{\partial}{\partial t} + \bar{u}_j \frac{\partial}{\partial x_j}$
$\frac{D}{Dt}$	Substantial derivative operator defined in terms of the density weighted mean velocity; $\frac{D}{Dt} \equiv \frac{\partial}{\partial t} + \tilde{u}_j \frac{\partial}{\partial x_j}$
$\frac{\mathcal{D}}{\mathcal{D}t}$	Substantial derivative operator defined in terms of the instantaneous velocity; $\frac{\mathcal{D}}{\mathcal{D}t} \equiv \frac{\partial}{\partial t} + u_j \frac{\partial}{\partial x_j}$
e	Internal energy per unit mass of mixture
$E\{\phi\}$	Mathematical expectation of the variable ϕ
E_r	Activation energy of the r^{th} reaction step
g	Gibbs free energy per unit mass of mixture; $g \equiv h - Ts$
g_i	Gravitational acceleration vector
h	Enthalpy per unit mass of mixture; $h \equiv e + \frac{p}{\rho}$
$J_{i,\alpha}$	Diffusional flux vector; $J_{i,\alpha} \equiv -\frac{\mu}{Sc} \frac{\partial Y_\alpha}{\partial x_i}$ for $\alpha \leq N$ and $J_{i,\alpha} \equiv -\frac{\mu}{Pr} \frac{\partial h}{\partial x_i}$ for $\alpha = N + 1$
k	Kinetic energy per unit mass; $k \equiv \frac{1}{2} \overline{u_i' u_i'}$ or $\frac{1}{2} \overline{u_i'' u_i''}$
k_{fr}, k_{br}	Forward and reverse rate constants of the r^{th} reaction step
Ka	Karlovitz number, the ratio of a reaction time scale to that of the fine scale dissipative motions e.g. $(\frac{\nu}{S_l^2}) \cdot (\frac{\nu}{\epsilon})^{-\frac{1}{2}} = \frac{\sqrt{Re}}{Da}$
$K_c(T)$	Equilibrium constant in terms of concentration, defined in equation (H.15)
$K_p(T)$	Equilibrium constant in terms of partial pressure, defined in

	equation (H.10)
$K_X(T, p)$	Equilibrium constant in terms of mole fraction, defined in equation (H.11)
L	Integral length scale of turbulence
$\dot{m}_\alpha$	Mass flow rate of specie α
Ma	Mach number of the flow; $Ma \equiv \frac{U}{\sqrt{\gamma RT}}$, where U is a representative velocity for the mixture
n_α	Specific mole number of specie α ; $n_\alpha \equiv \frac{Y_\alpha}{W_\alpha}$ (Kmol/Kg)
N	Number of species ($N+1$ includes the species field <i>and</i> the enthalpy (or other strictly conserved scalar, e.g. mixture fraction)
N	Penultimate number of nodes used in the finite-difference procedure
$\mathcal{N}$	Number of particles or realisations at a given position and time
$\mathcal{O}(\phi)$	Symbol to denote ' of the order of magnitude of ϕ '
p	Thermodynamic pressure
p_α	Partial pressure exerted by specie α ; $p_\alpha = X_\alpha p$
$\tilde{p}(\underline{\phi}, \underline{\psi})$	Fine-grained probability density function (pdf) of the event $\underline{\psi} = \underline{\phi}$; $\tilde{p}(\underline{\psi}, \underline{\phi}) \equiv \prod_{\alpha=1}^{N+1} \delta \{ \phi_\alpha(\mathbf{x}, t) - \psi_\alpha \}$
$P_{\phi_\alpha}(\psi_\alpha; \mathbf{x}, t)$	Pdf of the scalar random variable $\phi_\alpha(\mathbf{x}, t)$ with sample space ψ_α at position $\mathbf{x}$ and time t
$\tilde{P}_{\phi_\alpha}(\psi_\alpha; \mathbf{x}, t)$	Density-weighted pdf; $\tilde{P}_{\phi_\alpha}(\psi_\alpha; \mathbf{x}, t) \equiv \frac{\varrho(\underline{\psi})}{\bar{\varrho}(\mathbf{x}, t)} P_{\phi_\alpha}(\psi_\alpha; \mathbf{x}, t)$
Pr	Prandtl number for the mixture; $Pr \equiv \frac{c_p \mu}{K}$, K is the thermal conductivity
$\dot{r}_\alpha$	Net formation rate of specie α ; $\dot{r}_\alpha \equiv \frac{\dot{S}_\alpha}{\varrho W_\alpha}$ (Kmol Kg ⁻¹ s ⁻¹)
R	Universal gas constant; $R = 8314.3$ J Km ⁻¹ K ⁻¹
Re	Reynolds number of the large scale turbulence; $Re \equiv \frac{u' L}{\nu}$
s	Entropy per unit mass of mixture (J Kg ⁻¹ K ⁻¹)
Sc	Schmidt number for the mixture; $Sc \equiv \frac{\mu}{\varrho D}$
S_l	Laminar flame speed
$\dot{S}_\alpha$	Net formation rate of specie α per unit volume (Kg m ⁻³ s ⁻¹) $\dot{S}_\alpha(\underline{\phi}) \equiv \varrho(\underline{\phi}) \dot{S}_\alpha^*(\underline{\phi})$
t	Time
T	Temperature
u	Velocity
u'	Characteristic velocity of the energy-containing motions; $u' \sim \mathcal{O}(\sqrt{k})$
W_α	Molar mass of specie α (Kg/Kmol)
$\mathbf{x}$	Position vector
x, r	Coordinate directions in a cylindrical-polar frame of reference
X_α	Mole fraction of specie α ; $X_\alpha \equiv n_\alpha / (\sum_\beta n_\beta)$
Y_α	Mass fraction of specie α
z_α	Strictly conserved scalar e.g. element specific mole number or enthalpy per unit mass of mixture

Greek Symbols

γ	Ratio of specific heats; $\gamma \equiv \frac{c_p}{c_v}$
δ_{ij}	Kronecker delta; $\delta_{ij} = 1$ for $i = j$, zero otherwise
$\delta\{x\}, \delta(x)$	Delta function for the variable x
$\Delta h_{f,\alpha}$	Standard heat of formation of specie α
ε	Viscous dissipation rate of turbulence kinetic energy, defined in equation (2.35)
η	Kolmogorov length scale; $\eta = (\frac{\nu^3}{\varepsilon})^{\frac{1}{4}}$
ϑ_α	Chemical potential of specie α (J/Kmol), defined in equation (H.5)
κ	Von Karman constant; $\kappa \approx 0.41$
μ	Mixture molecular viscosity
μ_t	Turbulent eddy viscosity; $\mu_t \propto \bar{\rho} \frac{k^2}{\varepsilon}$
ν	Kinematic viscosity; $\nu \equiv \frac{\mu}{\bar{\rho}}$
$\nu'_{\alpha r}, \nu''_{\alpha r}$	Stoichiometric coefficients of specie α appearing in the r^{th} reaction step on the left and right hand side (respectively) of a chemical equation
ξ	Mixture fraction, defined in equation (3.3)
ρ	Density of the mixture
σ	Root-mean-square of the mixture fraction
σ_{ij}	Stress tensor, defined in equation (2.5)
σ_t	Turbulent Prandtl/Schmidt number, used in equation (2.41)
τ	Characteristic turbulent time scale; $\tau \equiv \frac{k}{\varepsilon}$
τ_{ij}	Viscous stress tensor, defined as: $\tau_{ij} = p\delta_{ij} + \sigma_{ij}$
v	Kolmogorov velocity scale; $v = (\nu\varepsilon)^{\frac{1}{4}}$
$\phi_\alpha(\mathbf{x}, t)$	Instantaneous scalar random variable of specie α for $\alpha = 1, \dots, N$. Enthalpy corresponds to $\alpha = N + 1$
χ_ϕ	Instantaneous scalar dissipation rate; $\chi_\phi = \frac{\nu}{Sc} \left(\frac{\partial \phi}{\partial x_i} \frac{\partial \phi}{\partial x_i} \right)$
ψ_α	Sample space of the random variable $\phi_\alpha(\mathbf{x}, t)$
Ψ	Stream function, defined in equation (5.3)
ω	Dimensionless stream function, defined in equation (5.4)
$\dot{\omega}_r$	Reaction rate of the r^{th} chemical step, $\text{Kmol m}^{-3} \text{s}^{-1}$
Ω_n	Normalised moment of the pdf, defined in equation (4.7)

Superscripts

A'	Fluctuating part of the unweighted decomposition of an instantaneous variable
A''	Fluctuating part of the density weighted decomposition of an instantaneous variable
$\bar{A}$	Unweighted average
$\tilde{A}$	Density weighted average or density weighting
A^n	Time level index in finite-difference equations
$\hat{A}$	Sample space of the random variable A

Subscripts

$A_{i,j,k}$	Coordinate indices
$A_{\alpha,\beta}$	Scalar field indices
A_{st}	Stoichiometric condition
A_{eq}	Equilibrium condition
$\underline{A}$	Scalar or vector set of values, $\{A_1, A_2, \dots\}$

Chapter 1

Introduction

1.1 Preamble

Turbulent combustion is a phenomenon which plays a central role in many engineering devices. Essentially it involves the basic process of combustion in which the mixing rates of fuel and oxidant are significantly influenced by the turbulence prevailing in the flow. The automotive, aircraft and power generation industries (to name a few) rely on the control of turbulent combustion so that their equipment (e.g. reciprocating engines, gas turbines and furnaces) can run as efficiently as possible. The design of such equipment is also affected by limitations, set out through legislation, on the levels of combustion-generated pollutant emissions; for example coal-fired furnaces may exhaust oxides of sulphur into the atmosphere leading to the problem of acid rain, whilst aircraft and automobile engines produce levels of CO, oxides of nitrogen and unburnt hydrocarbons (UHC) which may be (in the long run) harmful to the environment.

The experimental testing of engineering equipment is ultimately the most reliable way of assessing their suitability for practical/commercial applications. However, it is difficult to evaluate the effects of changes to a particular design purely from the experimental point of view; for many applications, extensive and repeated measurements *during the design and development stage* can be too expensive. As an alternative to experiments, predictive methods based on the solution of the conservation equations of mass, momentum and energy can be employed; these conservation equations can be expressed as a set of coupled non-linear partial differential equations (pde's). The complexity of the pde's rules out the possibility of obtaining analytical solutions, and numerical methods would demand computational resources orders of magnitude above the most powerful computers available today. The most popular predictive approach so far has been a statistical one whereby the conservation equations are ensemble-averaged. Whilst the numerical solution of the ensemble-averaged equations is feasible, the price that is paid is a loss of information concerning the fine details of the turbulent flow. The loss of information manifests itself as terms in the ensemble-averaged equations which cannot be evaluated (in any known way) as a function of the basic variables (e.g. pressure, velocity, temperature, species concentration) - this is the 'closure' problem. In order to represent the behaviour of these terms, mathematical modelling is required; in other words subject to certain assumptions, a functional form is postulated for these terms so that they can be evaluated. The resulting mathematical model is often referred to as a moment closure.

The requirements imposed on mathematical models for predicting the performance of com-

bustion systems nowadays are quite demanding. To predict with good accuracy the velocity and temperature fields is not always sufficient. It is also desirable to have calculation methods which are capable of reproducing finite-rate effects (e.g. extinction, ignition and emissions of pollutants) accurately; furthermore the methods need to be sufficiently general so as to reproduce measurements in a wide range of flows of engineering interest. Closure approximations are invariably required for the turbulent transport (diffusion) of momentum; this is achieved through the use of phenomenological turbulence models. Currently the most widely-used turbulence models in engineering calculations are based on either a two-equation model of turbulence (k - ϵ) or a Reynolds stress closure. However, the central difficulty in turbulent combustion is in the representation of the turbulence-chemistry interactions. More specifically, the averaged net formation rates which appear in the transport equations for the mean mass fraction of each chemical species present need to be evaluated. The *instantaneous* formation rates are strongly non-linear functions of the temperature and the species concentrations. Consequently the *averaged* formation rates cannot be evaluated purely in terms of the mean temperature and species concentrations; such a practice would lead to substantial errors, since in turbulent combustion the fluctuations in scalar properties are comparable (in magnitude) to the mean properties. A mathematically rigorous and computationally tractable way of calculating the mean formation rates is through the use of a joint probability density function (pdf) of the species mass fractions and the temperature.

In this thesis non-premixed combustion is considered. This implies that the fuel and air nominally enter in separate streams prior to mixing and reaction. The classical approach in non-premixed combustion has been to assume 'fast chemistry', in other words the time scales for the chemical reactions are presumed to be far shorter than the turbulent mixing time scales. With this assumption, and for the configuration of a two-stream mixing process (which is representative of many combustion devices), the thermochemical state can often be determined uniquely in terms of a single independent scalar variable (the mixture fraction). In these circumstances the specification of the joint pdf of the species concentrations and the temperature reduces to that of the pdf of the mixture fraction. By assuming that the latter pdf has a two-parameter form, it is possible to presume a suitable shape for the pdf of the mixture fraction in terms of its mean and variance; the moments of the pdf are in turn obtained from the solutions of their respective modelled transport equations. This approach (known as the conserved scalar formalism) works relatively well for situations where there are no appreciable finite-rate effects; this implies that the method is unable to accurately predict flows where the time scales of the chemistry and the turbulent mixing are comparable.

In order to reproduce finite-rate effects accurately more independent scalars need to be incorporated into the description of the pdf. With the specification of a multi-dimensional pdf the presumed shape method becomes impractical and the only viable technique appears to be the transported pdf approach. In this case, rather than presuming a shape for the scalar pdf, its exact evolution equation is modelled and then solved. The major appeal of this technique is that the source terms for chemical reaction can be treated without approximation, so that realistic chemistry can be explicitly incorporated into this methodology. Terms associated with molecular mixing and turbulent transport require modelling; in particular the turbulence-chemistry interactions still require some form approximation. As in other combustion formalisms, the degree of physics incorporated into the models describing the turbulence-chemistry interactions will determine the accuracy of the approach in reproducing effects such as extinction, and the levels of CO and UHC (for example in hydrocarbon flames). In practice a large number of scalars can appear in the

specification of the pdf, depending on the complexity of the reaction scheme that is being incorporated. These scalars act as independent variables, like position and time. As far as the solution of the resulting pdf equation is concerned, this implies that finite-difference procedures will be too expensive to employ; therefore a (computationally) more efficient stochastic (Monte Carlo) simulation of the pdf equation is used to overcome the limitations imposed by the finite-difference technique.

Recently work has appeared in the literature where direct numerical simulation (DNS) and large eddy simulation (LES) techniques have been applied to the problem of turbulent combustion. In the case of DNS, problems in idealized configurations and at low Reynolds numbers are considered, usually in homogeneous decaying turbulence (e.g. Vervisch et al. [101]). The use of such a method, with current computer capabilities, can only be directed at investigating the fundamental interactions occurring in flames for the aforementioned idealized configurations. By carefully scrutinizing the turbulence-chemistry interactions, it may be possible to infer improved closure principles for the (model-dependent) techniques used to analyse the high Reynolds number reacting flows of engineering interest. With regards to LES, although there are no Reynolds number restrictions (as experienced by DNS), modelling is required. Because chemical reactions occur in the fine scales of turbulent motion, the source terms for chemical production appear in an unclosed form. Therefore at first sight it may appear that there is little to be gained from such a technique in combusting flows. More research is required to substantiate whether LES in combustion is beneficial, particularly in complex flows where it is claimed that the potential advantage of LES over more conventional (phenomenological) approaches becomes apparent.

1.2 Objectives of the Research

The primary aim of this thesis is to apply the *scalar* transported (pdf) approach to predict the evolution of turbulent combusting flows. The configurations of a jet issuing into stagnant and coflowing surrounds are chosen as the basic geometries for the calculations using this formalism. In this respect, a relatively simple flow has been chosen so that efforts can be directed towards the evaluation of the combustion formalism, with the understanding that the turbulence models can reproduce the behaviour of the velocity field accurately.

A finite-difference procedure was employed to solve the equations governing the evolution of the velocity field. These included the momentum equation together with the modelled transport equations required to evaluate the Reynolds stresses. A $k-\varepsilon$ and full Reynolds stress closure were employed for this purpose. The scalar field was treated using a Monte Carlo simulation of the transported pdf equation. The number of independent scalars associated with the pdf was determined by the choice of the reaction scheme. Two different mechanisms were employed: a reduced scheme resulting from a systematic reduction of a detailed methane/air mechanism, and a global hydrocarbon scheme (which did not involve the appearance of radical species). The molecular mixing term which appears in an unclosed form in the pdf equation was approximated using a coalescence-dispersion model and a closure based on linear mean square estimation (LMSE).

The main motivation for this study has been the accurate prediction of CO levels in jet diffusion flames employing a hydrocarbon as the fuel (propane or methane). The ability of the single-point, single-time scalar pdf formalism in reproducing finite-rate effects is also assessed. Detailed

measurements from four turbulent jet diffusion flames were used in order to evaluate the proposed level of closure. The studies of Godoy [31] and Hassan, Lockwood and Moneib [35] relating to propane and methane flames (respectively) issuing into stagnant surrounds provided measurements of the CO levels, as well as the major species (CO_2 , O_2 , UHC). The data of Hassan et al. was particularly useful since it also included data relating to the mean and fluctuating temperature field, as well as an investigation of the effect of nitrogen-dilution of the fuel stream on the evolution of the flame. The measurements of the scalar field performed by Godoy was further augmented in the study of Heitor, Moreira and Pires [37] where information relating to the fluctuating velocity and temperature field was presented. The wealth of data relating to the mean, root-mean-square (rms) and pdf of the scalars, described in the work of Masri, Bilger and Dibble [66, 68], was used to study the ability of the pdf equation to represent effects of local extinction in flames.

The shortcomings and achievements of the scalar transported pdf approach coupled to a moment closure are demonstrated, and subsequently suggestions are made for ways in which the theory can be improved. To this end, calculations are also reported for a confined laminar jet diffusion flame and a homogeneous, isotropic turbulence. In these simpler test cases, the capabilities of the chemistry and the mixing models can be evaluated more easily; at the same time the results appeared to be relevant when it came to describing some of the characteristics of the predictions of the turbulent jet flames.

Chapter 2

The Governing Equations of Turbulent Combusting Flows

2.1 Introduction

This chapter presents the basic equations which describe the evolution of turbulent combusting flows. The instantaneous equations are first outlined, followed by the discussion of the averaging procedure required to solve the conservation equations for flows of practical interest. Turbulence modelling is briefly reviewed and this is then followed by a discussion of the mean reaction rate term which appears when the species conservation equations are ensemble-averaged. This then motivates the discussion for combustion modelling which is described in the subsequent chapters.

2.2 The Instantaneous Equations

The governing equations describing the flow of a fluid can be derived from the principles of conservation of mass, momentum and energy; derivations can be found in works such as Schlichting [85]. Unless otherwise stated, cartesian tensor notation and S.I. units are used throughout.

2.2.1 Mass Continuity

$$\frac{\partial \varrho}{\partial t} + \frac{\partial \varrho u_j}{\partial x_j} = 0 \quad (2.1)$$

where $u_j(\mathbf{x}, t)$ is the instantaneous velocity vector, and $\varrho(\mathbf{x}, t)$ is the density. $u_j(\mathbf{x}, t)$ is representative of the velocity of the reactive mixture at a point, and is defined as a local mass-average instantaneous velocity:

$$u_j(\mathbf{x}, t) \equiv \frac{\sum_{\alpha=1}^N \varrho_{\alpha}(\mathbf{x}, t) \mathcal{U}_{j,\alpha}(\mathbf{x}, t)}{\sum_{\alpha=1}^N \varrho_{\alpha}(\mathbf{x}, t)} \quad (2.2)$$

where $\mathcal{U}_{j,\alpha}(\mathbf{x}, t)$ is the instantaneous velocity vector of specie α . From now on the dependence of the scalar and hydrodynamic variables on $(\mathbf{x}, t)$ is omitted for conciseness. In equation (2.1) the terms associated with molecular diffusion do not appear; this arises out of the physical requirement that there can be no *net* diffusion of mass in a multi-component system. The definition of u_i in

equation (2.2) and the diffusional flux vector of specie α , $J_{i,\alpha}$, ensure that this requirement is satisfied:

$$J_{i,\alpha} \equiv \rho_\alpha (\mathcal{U}_{i,\alpha} - u_i) \quad (2.3)$$

$(\mathcal{U}_{i,\alpha} - u_i)$ is sometimes referred to as the diffusional velocity vector associated with specie α . Substituting equation (2.2) into (2.3) and summing over α leads to the result $\sum_{\alpha=1}^N J_{i,\alpha} = 0$. Another requirement for the mixture, as a whole, is that there can be no net generation/destruction, although the individual species comprising the mixture can be produced/consumed through the effect of chemical reaction.

2.2.2 Momentum Conservation Equation

$$\rho \frac{\partial u_i}{\partial t} + \rho u_j \frac{\partial u_i}{\partial x_j} = \frac{\partial \sigma_{ij}}{\partial x_j} + \rho g_i \quad (2.4)$$

Equation (2.4) is essentially a generalisation of Newton's second law of motion for the flow of a fluid, written in a form appropriate to an infinitesimal and stationary control volume. It can be shown [1] that if the fluid is Newtonian (i.e. the stress components depend linearly on the rates of deformation) then,

$$\sigma_{ij} = \left(-p + \lambda \frac{\partial u_k}{\partial x_k} \right) \delta_{ij} + \mu \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) \quad (2.5)$$

μ is the mixture (molecular) viscosity and by invoking Stokes' hypothesis $\lambda = -\frac{2}{3}\mu$ [†]. Substituting for σ_{ij} in equation (2.4) using (2.5) and Stokes' hypothesis, yields the Navier - Stokes equations:

$$\rho \frac{Du_i}{Dt} = -\frac{\partial p}{\partial x_i} + \frac{\partial}{\partial x_j} \left[\mu \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} - \frac{2}{3} \delta_{ij} \frac{\partial u_k}{\partial x_k} \right) \right] + \rho g_i \quad (2.6)$$

where the operator $\frac{D}{Dt}$ is the substantial derivative defined as $\frac{\partial}{\partial t} + u_j \frac{\partial}{\partial x_j}$.

2.2.3 Energy Conservation Equation

The energy equation may expressed as,

$$\underbrace{\rho \frac{\partial}{\partial t} \left(h + \frac{1}{2} u_i u_i \right)}_{\text{I}} + \underbrace{u_k \frac{\partial}{\partial x_k} \left(h + \frac{1}{2} u_i u_i \right)}_{\text{II}} = \underbrace{\frac{\partial \dot{q}_i}{\partial x_i}}_{\text{III}} + \underbrace{\dot{Q}_{\text{rad}}}_{\text{IV}} + \underbrace{\frac{\partial}{\partial x_j} (u_i \tau_{ij})}_{\text{V}} + \underbrace{\frac{\partial p}{\partial t}}_{\text{VI}} \quad (2.7)$$

The internal energy per unit mass of mixture is related to the kinetic energy and enthalpy by,

$$e = h - \frac{p}{\rho} = \sum_{\alpha=1}^N Y_\alpha h_\alpha - \frac{p}{\rho} \quad (2.8)$$

The effect of chemical reaction appears in the energy equation through the definition of h_α :

$$h_\alpha \equiv \int_{T^\circ}^T c_{p,\alpha} d\hat{T} + \Delta h_{f,\alpha} \quad (2.9)$$

where T° is a reference temperature and $\Delta h_{f,\alpha}$ represents the standard heat of formation of specie α . The latter can be defined as the heat released when one kilomole of substance is formed from its

[†]The grouping $\lambda + \frac{2}{3}\mu$ is often referred to as the coefficient of bulk viscosity

elements in their respective *standard* states (usually 1atm and 298K). By convention, each element in its standard state is assigned an enthalpy of zero. The inclusion of $\Delta h_{f,\alpha}$ ($\alpha = 1, \dots, N$) in the evaluation of h accounts for the heat release (and absorption) effects due to the chemical reactions taking place.

The various effects which contribute to the diffusional heat flux (term III in equation (2.7)) need to be elaborated. Conduction of heat arising from temperature gradients is an obvious contributor and this is represented through Fourier's law. Since the instantaneous velocity of specie α ($U_{i,\alpha}$) can differ from the mass-average instantaneous velocity of the mixture (u_i), the total (diffusional) flux of enthalpy associated with this effect is: $\rho \sum_{\alpha=1}^N Y_{\alpha} (U_{i,\alpha} - u_i) h_{\alpha}$. Invoking Fick's law of diffusion,

$$\rho Y_{\alpha} (U_{i,\alpha} - u_i) \approx -\frac{\mu}{Sc} \frac{\partial Y_{\alpha}}{\partial x_i} \quad (2.10)$$

where Sc is the Schmidt number of the mixture. The presence of concentration gradients can produce a heat flux¹. It is conventional practice in the analysis of combustng flows to ignore such a transport process, partly because experience suggests that it does not provide a significant contribution in the energy balance. Consequently the heat flux vector becomes:

$$q_i = -\frac{c_p \mu}{Pr} \frac{\partial T}{\partial x_i} - \frac{\mu}{Sc} \sum_{\alpha=1}^N h_{\alpha} \frac{\partial Y_{\alpha}}{\partial x_i} \quad (2.11)$$

where Pr is the mixture Prandtl number. It is convenient to express q_i in terms of enthalpy rather than temperature. This can be achieved by firstly differentiating the relation $h = h(T, Y_{\alpha})$ with respect to x_i ; this yields,

$$\frac{\partial h}{\partial x_i} = c_p \frac{\partial T}{\partial x_i} + \sum_{\alpha=1}^N h_{\alpha} \frac{\partial Y_{\alpha}}{\partial x_i} \quad (2.12)$$

In arriving at equation (2.12) the following relations have been used: $c_p \equiv \left(\frac{\partial h}{\partial T} \right)_p$ and from equation (2.8), $\frac{\partial h}{\partial Y_{\alpha}} = h_{\alpha}$. Substituting equation (2.12) into (2.11) gives,

$$q_i = -\frac{\partial}{\partial x_i} \left\{ \frac{\mu}{Pr} \frac{\partial h}{\partial x_i} \right\} - \frac{\partial}{\partial x_i} \left\{ \mu \left(\frac{1}{Sc} - \frac{1}{Pr} \right) \sum_{\alpha=1}^N h_{\alpha} \frac{\partial Y_{\alpha}}{\partial x_i} \right\} \quad (2.13)$$

Term IV in equation (2.7) represents the energy exchanges associated with the absorption and emission of thermal radiation. Energy transfer due to radiation is not considered in this work. The reasons for this are partly related to the fact that the flames being investigated are 'small-scale', such that the heat loss by radiation is usually a negligible fraction of the total heat flow [78]. As far as term V is concerned, in low speed combustng flows the work done against viscous forces is negligible in comparison to the thermal energy changes brought about by chemical reaction; the same is true for terms associated with the work done against body forces. Taking ' U ' as a representative velocity of the mixture, the Mach number is defined as, $Ma \equiv \frac{U}{\sqrt{\gamma R T}}$ where $\gamma \equiv \frac{c_p}{c_v}$. Using $\gamma R = c_p(\gamma - 1)$, the following can be written,

$$\frac{1}{2} U^2 = \frac{1}{2} Ma^2 c_p (\gamma - 1) T \quad \Rightarrow \quad \frac{\frac{1}{2} U^2}{c_p T} = \frac{1}{2} Ma^2 (\gamma - 1) \quad (2.14)$$

¹This is known as the Dufour effect - the Soret effect is essentially the reciprocal of this, i.e. the presence of temperature gradients giving rise to species diffusional flux.

The quantity $\frac{\frac{1}{2}U^2}{c_p T}$ can be interpreted as the fractional rise in temperature ($\frac{\Delta T}{T}$) that would result from all the kinetic energy (per unit mass), at the point in question, being converted into heat. Therefore provided $Ma \ll 1$, the kinetic energy of the mixture at a point can be neglected in comparison to its enthalpy.

Term VI ($\frac{\partial p}{\partial t}$) in equation (2.7) is assumed to be negligible in many practically occurring situations, e.g. jets, gas turbines and furnaces, but cannot be neglected in the flows occurring, for example, in internal combustion engines. If the flow is statistically stationary, the term will be zero upon averaging. However, even if the flow is statistically non-stationary the term can still be neglected provided the boundary conditions imply that the reference pressure does not change significantly. Finally, the Lewis number, defined as the ratio of the Schmidt number to the Prandtl number is close to unity in combusting flows, and it is a useful assumption to take it as exactly unity, since the energy equation then simplifies to:

$$\rho \frac{\partial h}{\partial t} + \rho u_j \frac{\partial h}{\partial x_j} = \frac{\partial}{\partial x_i} \left(\frac{\mu}{Pr} \frac{\partial h}{\partial x_i} \right) \quad (2.15)$$

2.2.4 Species Conservation Equation

$$\rho \frac{\partial Y_\alpha}{\partial t} + \rho u_j \frac{\partial Y_\alpha}{\partial x_j} = - \frac{\partial J_{i,\alpha}}{\partial x_i} + \dot{S}_\alpha \quad \text{for } \alpha = 1, \dots, N \quad (2.16)$$

where $\dot{S}_\alpha$ is the net formation rate of specie α per unit volume. The mass conservation law requires that there can be no overall source/sink of mass, so that $\sum_{\alpha=1}^N \dot{S}_\alpha = 0$. $J_{i,\alpha}$ is the diffusional flux vector of specie α , and using Fick's law of diffusion this can be approximated as: $J_{i,\alpha} \approx -\frac{\mu}{Sc} \frac{\partial Y_\alpha}{\partial x_i}$. This approximation is strictly true for binary mixtures, and so the use of Fickian diffusion in the context of multi-component mixtures is only justified if the Schmidt number is the same for all species (in order to ensure that the mass fractions sum to unity). The assumption of uniform Sc (which is tantamount to assuming equal diffusivity) is known to be poor when species with widely differing molar masses are being considered. To obtain a general expression for $\dot{S}_\alpha$, consider the system involving N chemical species and R reaction steps, expressed in the following compact form [60]:

$$\sum_{\alpha=1}^N \nu'_{\alpha r} M_\alpha = \sum_{\alpha=1}^N \nu''_{\alpha r} M_\alpha \quad r = 1, \dots, R \quad (2.17)$$

where the rate constants for the forward and backward reactions are k_{f_r} and k_{b_r} , respectively, and M_α denotes the chemical symbol of specie α . If specie α does not participate in a particular reaction then its stoichiometric coefficients appearing in reaction r , referred to as $\nu'_{\alpha r}$ and $\nu''_{\alpha r}$, are zero. The rate constants can be expressed as:

$$k_{f_r} = B_r T^{a_r} \exp \left(-\frac{E_r}{RT} \right) \quad (2.18)$$

where B_r is the pre-exponential factor (for the r^{th} reaction step), a_r is a constant exponent, E_r is the activation energy and R is the universal gas constant. The net rate of formation of specie α is the sum of the net formation rates of specie α in all the individual reaction steps. In any particular reaction step, the net rate of formation of specie α is determined by the magnitudes of the forward and backward rates in reaction r . Mathematically this can be expressed as:

$$\dot{S}_\alpha = W_\alpha \sum_{r=1}^R (\nu''_{\alpha r} - \nu'_{\alpha r}) \left\{ k_{f_r} \rho^{m_r} \prod_{\zeta=1}^N \left(\frac{Y_\zeta}{W_\zeta} \right)^{\nu'_{\zeta r}} - k_{b_r} \rho^{l_r} \prod_{\zeta=1}^N \left(\frac{Y_\zeta}{W_\zeta} \right)^{\nu''_{\zeta r}} \right\} \quad (2.19)$$

where $m_r \equiv \sum_{\zeta=1}^N \nu'_{\zeta r}$ and $l_r \equiv \sum_{\zeta=1}^N \nu''_{\zeta r}$ are the orders of the forward and reverse reactions respectively. The term in curly brackets in equation (2.19) is effectively the *net forward rate* of the r^{th} reaction step (in $\text{Kmol m}^{-3}\text{s}^{-1}$). In order to close the system of equations, an expression for the density is required. This is given by an equation of state,

$$\rho = \rho(\underline{Y}, h, p) \quad (2.20)$$

where $\underline{Y}$ denotes the set of species mass fractions (i.e. Y_α where $\alpha = 1, \dots, N$). The assumption of low Mach number can be used to remove the dependence of ρ on pressure variations, Δp , caused by the flow. Essentially $Ma \ll 1$ ensures that the coupling of the momentum and scalar conservation equations is through the density, and that the linkage through the pressure is removed. To demonstrate this an estimate of the magnitude of Δp is required. At high Reynolds number,

$$\Delta p \sim \mathcal{O}(\rho U^2) \quad \text{where} \quad \rho U^2 = Ma^2(\rho a_s^2) \quad (2.21)$$

U is a characteristic velocity and a_s is the speed of sound ($a_s = \sqrt{\gamma RT} = (\frac{\gamma p}{\rho})^{\frac{1}{2}}$). Substituting for a_s into equation (2.21) yields,

$$\frac{\Delta p}{p} \sim \mathcal{O}(Ma^2) \quad (2.22)$$

Whilst ρ depends on the absolute level of pressure, if $Ma \ll 1$ its dependence on pressure fluctuations can be ignored. Under these circumstances p can be interpreted as a reference pressure, and in certain flows, such as jet flows, furnaces and gas turbine combustors in the steady-state, the reference pressure can be treated as a constant. The equation of state for an ideal gas is,

$$p = \rho RT \sum_{\alpha=1}^N \frac{Y_\alpha}{W_\alpha} \quad (2.23)$$

with the understanding that in the flows described above, p is treated as nominally constant ($= p_o$, the reference pressure) *as far as the evaluation of ρ is concerned*. Consequently the state relations can be re-expressed as $\rho = \rho(\underline{Y}, h, p_o)$. In internal combustion engines the reference pressure does change significantly, and in this situation the aforementioned relation is no longer valid.

2.3 The Averaging Process

The discussion so far has developed a closed set of equations and in principle they could be solved (numerically) with appropriate initial and boundary conditions. For the high Reynolds number flows of engineering interest, direct numerical simulation (DNS) is computationally prohibitive [83]; typically the cpu requirements for a DNS of inert mixing are of the order of the Reynolds number (Re) raised to the power three. For statistically homogeneous 'low' Re flows, DNS is feasible and as discussed in section 4 it can be very useful. The traditional approach for treating high Re turbulent flows has been to average the conservation equations, with the consequence that all the fine details of the flow do not require spatial and temporal resolution. Different types of averaging can be applied, but they all share a common feature; the closed set of instantaneous equations are transformed by averaging into an 'open' set of equations. Terms appear which cannot be accounted for in any known way via the dependent variables of the flow field. Modelling serves to approximate these terms, and the purpose of these introductory chapters is to address the principles behind the representation of such terms. The two commonly-employed types of

averaging are based on the Reynolds (unweighted) and Favre (density-weighted) approach, defined as follows:

$$\bar{A}(\mathbf{x}, t) \equiv \lim_{N_r \rightarrow \infty} \frac{1}{N_r} \sum_{i=1}^{N_r} A^{(i)}(\mathbf{x}, t) \quad \text{Reynolds averaging} \quad (2.24)$$

$$\tilde{A}(\mathbf{x}, t) \equiv \frac{1}{\bar{\rho}} \lim_{N_r \rightarrow \infty} \frac{1}{N_r} \sum_{i=1}^{N_r} \rho^{(i)} A^{(i)}(\mathbf{x}, t) \quad \text{Favre averaging} \quad (2.25)$$

where $A^{(i)}(\mathbf{x}, t)$ is the i^{th} sample amongst N_r realisations of an experiment. The standard practice is to decompose the instantaneous equations into mean and fluctuating parts and then to average them. With density-weighted averaging the velocity and scalars (but not the density and pressure) are decomposed according to $u_i = \tilde{u}_i + u_i''$, $\phi_\alpha = \tilde{\phi}_\alpha + \phi_\alpha''$. With the definition of Favre averaging in equation (2.25) it can be shown that $\overline{\rho \phi_\alpha''} = \overline{\rho u_i''} = 0$. For Reynolds averaging, $u_i = \bar{u}_i + u_i'$, $\phi_\alpha = \bar{\phi}_\alpha + \phi_\alpha'$ and where $\overline{u_i'} = \overline{\phi_\alpha'} = 0$. From the basic definitions, the following results can be established between weighted and unweighted statistics:

$$\bar{u}_i = \tilde{u}_i + \overline{u_i''}; \quad \bar{\phi}_\alpha = \tilde{\phi}_\alpha + \overline{\phi_\alpha''} \quad (2.26)$$

$$\overline{u_i''} = -\frac{\overline{\rho' u_i'}}{\bar{\rho}}; \quad \overline{\phi_\alpha''} = -\frac{\overline{\rho' \phi_\alpha'}}{\bar{\rho}} \quad (2.27)$$

$$\overline{\rho' u_i''} = \overline{\rho' u_i'}; \quad \overline{\rho' \phi_\alpha''} = \overline{\rho' \phi_\alpha'} \quad (2.28)$$

The reason for employing Favre averaging is that the resulting conservation equations do not include terms involving correlations of density fluctuations. Consequently these equations are easier to interpret (and hence to model) than their Reynolds averaged counterparts. Furthermore, Favre averaging leads to equations describing the mean of conserved quantities e.g., the resulting momentum equation is in terms of $\overline{\rho u_i}$ ($\equiv \bar{\rho} \tilde{u}_i$) rather than $\bar{\rho} \bar{u}_i$ which is obtained from Reynolds averaging and is not conserved. Despite the advantages of adopting Favre averaging, instruments may measure values close to density-weighted or unweighted averages². Clearly the ability to calculate both unweighted and density-weighted quantities is desirable. But $\overline{\rho u_i} \equiv \bar{\rho} \tilde{u}_i = \bar{\rho} \bar{u}_i + \overline{\rho' u_i'}$, and $\overline{\rho' u_i'}$ can be very difficult to calculate accurately; in terms of the pdf, unweighted and weighted statistics can be used interchangeably without approximation [6]. Applying Favre averaging to equations (2.1), (2.6), (2.15) and (2.16) yields:

$$\frac{\partial}{\partial t} (\bar{\rho}) + \frac{\partial}{\partial x_j} (\bar{\rho} \tilde{u}_j) = 0 \quad (2.29)$$

$$\frac{\partial}{\partial t} (\bar{\rho} \tilde{u}_i) + \frac{\partial}{\partial x_j} (\bar{\rho} \tilde{u}_j \tilde{u}_i) = -\frac{\partial \bar{p}}{\partial x_i} - \frac{\partial}{\partial x_j} (\bar{\rho} \widetilde{u_j'' u_i''}) + \bar{\rho} g_i \quad (2.30)$$

$$\frac{\partial}{\partial t} (\bar{\rho} \tilde{h}) + \frac{\partial}{\partial x_j} (\bar{\rho} \tilde{u}_j \tilde{h}) = -\frac{\partial}{\partial x_j} (\bar{\rho} \widetilde{u_j'' h''}) \quad (2.31)$$

$$\frac{\partial}{\partial t} (\bar{\rho} \tilde{Y}_\alpha) + \frac{\partial}{\partial x_j} (\bar{\rho} \tilde{u}_j \tilde{Y}_\alpha) = -\frac{\partial}{\partial x_j} (\bar{\rho} \widetilde{u_j'' Y_\alpha''}) + \bar{S}_\alpha \quad (2.32)$$

²An infinitely small thermocouple will detect an unweighted record of an instantaneous temperature, but some degree of density-weighting is associated with thermocouples of finite size. The extent of this weighting with 40 μm diameter thermocouples is unlikely to exceed around 50°C compared with differences between weighted and unweighted values which can exceed 300°C [49]. Sampling probes and LDV measure values very close to density-weighted concentrations and velocities, respectively.

where equation (2.32) holds for $\alpha = 1, \dots, N$. The above equations have been written for the limit of high Reynolds number; under these circumstances, order of magnitude analysis shows that the averaged molecular fluxes can be treated as negligible in comparison with the turbulent fluxes, namely the Reynolds stress ($\overline{\rho u_i'' u_j''}$) and the scalar flux ($\overline{\rho u_j'' \phi_\alpha''}$). Consequently the equations have a form identical to their constant density counterparts. The unknowns appearing in the equations are the Reynolds stress, scalar flux, mean density and the mean formation rate due to chemical reaction. In the following sections the turbulence models used to calculate $\overline{\rho u_i'' u_j''}$ are described since they form an essential part of the calculation method; in addition, for the purpose of notation ϕ_α ($\alpha = 1, \dots, N + 1$) is used to denote the entire scalar field, i.e. the species mass fractions ($\alpha = 1, \dots, N$) and the mixture enthalpy ($\alpha = N + 1$).

2.4 The Energy Cascade Viewpoint

Before discussing turbulence modelling it is useful to provide a brief overview of the 'energy cascade process'. This will demonstrate the complexity of the interactions taking place in a turbulent flow and help to exemplify the degree of simplification associated with current modelling approaches.

Turbulence can be thought of as a tangle of eddies of varying length scales interacting with one another. Only at the Kolmogorov scales of motion do the effects of viscosity become significant in comparison to the inertial effects. This means that for the larger scale motions the entangled eddies can interact with one another in a way which is essentially unaffected by the viscous stresses; this implies that for these eddies their angular momenta can be treated as constant.

Energy cascade arguments [8] suggest that energy is initially transferred from the mean flow to the 'large' eddies (constituting the fluctuating field) which have a preferred orientation. The fine scales of turbulent motion cannot interact directly with (and thereby extract energy from) the mean motion due to the disparity of the scales of motion. Through a process of 'vortex stretching' an eddy experiences a rate of strain along its axis of rotation and is extended with the consequence that its cross-sectional area reduces. Since it is presumed that the angular momentum of the eddy remains constant during this process, this implies that its kinetic energy of rotation increases. The energy required for the stretching comes from a neighbouring eddy whose axis of rotation has a component at right angles to the axis of the eddy being stretched. As an eddy reduces its (diametral) length scale it can, in turn, bring about the stretching of other eddies whose axes of rotation are at right angles to its own.

In this way energy can be transferred from the large scale (energy-containing) motions right down to the Kolmogorov scales of motion where, by virtue of the substantially larger velocity gradients, the kinetic energy of the eddies is dissipated into heat. It is argued that the small-scale eddies no longer share the preferred orientation of the mean rate of strain. It is postulated that at the fine scales, the turbulence has no preferred direction and is therefore *locally isotropic* - this implies that the statistics of the fine scale motions are invariant under the action of rotation and/or reflection of the coordinate system³.

The arguments presented in the above paragraphs suggest that the viscous and scalar dissipation rates are determined by the rate of energy transfer from the large to the small scales. This implies

³Some of the consequences of local isotropy are: $\overline{\frac{\partial u_1}{\partial x_j} \frac{\partial u_1}{\partial x_j}} = \overline{\frac{\partial u_2}{\partial x_j} \frac{\partial u_2}{\partial x_j}} = \overline{\frac{\partial u_3}{\partial x_j} \frac{\partial u_3}{\partial x_j}}$ and $\overline{\frac{\partial u_1}{\partial x_j} \frac{\partial u_2}{\partial x_j}} = 0$.

that when the Reynolds number of the flow changes, the fine scales re-adjust themselves in order to maintain the balance of energy transfer.

2.5 Turbulence Models

In turbulent combustion large fluctuations in fluid density arise and proper account must be taken of them. Most of the work to date concerning turbulence modelling is based on the consideration of constant density flows. The most common approach is to recast the models for constant density flows in terms of Favre averages *with the assumption that the density fluctuations are accounted for by the averaging*. Experimental verification of this assumption is very difficult owing to the level of detail required to quantify the contribution made by density fluctuations in real flames, and subsequently to translate this into a modelling criterion. However, the results based on such an approach have so far been encouraging [42].

An appropriate starting point for the discussion on turbulence modelling is the density-weighted Reynolds stress equation (derived from equation (2.6)):

$$\begin{aligned}
 & \underbrace{\bar{\rho} \frac{D}{Dt} (\widetilde{u_i'' u_j''})}_I + \underbrace{\bar{\rho} \widetilde{u_i'' u_j''} \frac{\partial \tilde{u}_j}{\partial x_k} + \bar{\rho} \widetilde{u_j'' u_i''} \frac{\partial \tilde{u}_i}{\partial x_k}}_{II} = - \underbrace{\frac{\partial}{\partial x_k} \{ \overline{\rho u_i'' u_j'' u_k''} \}}_{III} \\
 & - \underbrace{\left\{ \overline{u_j'' \frac{\partial \tau_{ik}}{\partial x_k}} + \overline{u_i'' \frac{\partial \tau_{jk}}{\partial x_k}} \right\}}_{IV} - \underbrace{\left\{ \overline{u_i'' \frac{\partial p'}{\partial x_j}} + \overline{u_j'' \frac{\partial p'}{\partial x_i}} \right\}}_V \\
 & - \underbrace{\left\{ \overline{u_i'' \frac{\partial \bar{p}}{\partial x_j}} + \overline{u_j'' \frac{\partial \bar{p}}{\partial x_i}} \right\}}_{VI} \quad (2.33)
 \end{aligned}$$

In the above, the operator $\frac{D}{Dt} \equiv \frac{\partial}{\partial t} + \tilde{u}_k \frac{\partial}{\partial x_k}$. The terms appearing in equation (2.33) have the following significance:

- I: Rate of change of $\widetilde{u_i'' u_j''}$ including convection by the mean flow
- II: Production of $\widetilde{u_i'' u_j''}$ by the interaction with the mean strain rate
- III: Diffusive transport of $\widetilde{u_i'' u_j''}$ by velocity fluctuations
- IV: Viscous destruction of $\widetilde{u_i'' u_j''}$
- V: Fluctuating velocity-pressure gradient correlation
- VI: Production/destruction of $\widetilde{u_i'' u_j''}$ by the mean pressure gradient

Terms I and II appear in exact form whilst the rest of the terms require closure approximations. Term IV can be simplified for high Re turbulence; first the following manipulation can be applied:

$$\overline{u_j'' \frac{\partial \tau_{ik}}{\partial x_k}} + \overline{u_i'' \frac{\partial \tau_{jk}}{\partial x_k}} = \underbrace{\frac{\partial \overline{u_i'' \tau_{jk}}}{\partial x_k} + \frac{\partial \overline{u_j'' \tau_{ik}}}{\partial x_k}}_{\text{transport}} - \underbrace{(\bar{\tau}_{ik} + \tau'_{ik}) \frac{\partial \overline{u_j''}}{\partial x_k} - (\bar{\tau}_{jk} + \tau'_{jk}) \frac{\partial \overline{u_i''}}{\partial x_k}}_{\text{dissipation}} \quad (2.34)$$

Order of magnitude analysis demonstrates that $\overline{\tau'_{ik}(\partial u''_j/\partial x_k)} + \overline{\tau'_{jk}(\partial u''_i/\partial x_k)}$ is the dominant term on the rhs of equation (2.34) and the other terms become negligible as $Re \rightarrow \infty$. Local isotropy implies that the off-diagonal components are zero and the dissipative term can be written as:

$$\overline{\tau'_{ik} \frac{\partial u''_j}{\partial x_k}} + \overline{\tau'_{jk} \frac{\partial u''_i}{\partial x_k}} \equiv \bar{\rho} \varepsilon_{ij} = \frac{2}{3} \delta_{ij} \bar{\rho} \varepsilon \quad \text{where} \quad \varepsilon \equiv \frac{1}{\bar{\rho}} \left(\overline{\tau'_{lk} \frac{\partial u''_l}{\partial x_k}} \right) \quad (2.35)$$

Following Lumley [64] the fluctuating velocity-pressure gradient correlations are split up into a redistributive and a nonredistributive part:

$$\overline{u''_j \frac{\partial p'}{\partial x_i}} + \overline{u''_i \frac{\partial p'}{\partial x_j}} = \underbrace{\left(\overline{u''_j \frac{\partial p'}{\partial x_i}} + \overline{u''_i \frac{\partial p'}{\partial x_j}} \right)}_{\text{redistributive}} - \frac{2}{3} \delta_{ij} \overline{u''_k \frac{\partial p'}{\partial x_k}} + \underbrace{\frac{2}{3} \delta_{ij} \overline{u''_k \frac{\partial p'}{\partial x_k}}}_{\text{nonredistributive}} \quad (2.36)$$

The redistributive part has no role in the generation or destruction of turbulence as it is constructed to vanish on contraction of the indices (i.e. it disappears in the turbulence kinetic energy equation (2.39)). The non-redistributive term can be further manipulated to yield:

$$\frac{2}{3} \delta_{ij} \overline{u''_k \frac{\partial p'}{\partial x_k}} = \underbrace{\frac{2}{3} \delta_{ij} \overline{\frac{\partial u''_k p'}{\partial x_k}}}_{\text{transport}} - \underbrace{\frac{2}{3} \delta_{ij} \overline{p' \frac{\partial u''_k}{\partial x_k}}}_{\text{noise}} \quad (2.37)$$

For $Ma \ll 1$ the noise production term can be neglected [4]. Using the results from equations (2.35) and (2.37), the equation for the turbulence kinetic energy, defined as,

$$k \equiv \frac{1}{2} \left(\overline{u''_1^2} + \overline{u''_2^2} + \overline{u''_3^2} \right) = \frac{\overline{u''^2}}{2} \quad (2.38)$$

can be obtained by contracting the indices of equation (2.33) and dividing by two:

$$\bar{\rho} \frac{Dk}{Dt} + \bar{\rho} \overline{u''_i u''_j} \frac{\partial \tilde{u}_j}{\partial x_i} = - \frac{\partial}{\partial x_j} \left\{ \frac{1}{2} \overline{\rho u''_j u''_i u''_i} + \overline{u''_j p'} \right\} - \bar{\rho} \varepsilon - \overline{u''_i} \frac{\partial \bar{p}}{\partial x_i} \quad (2.39)$$

2.5.1 Eddy-Viscosity Models

The simplest turbulence closures are based on the eddy-viscosity approach. Essentially they attempt to represent turbulent transport using relations similar in form to the molecular flux laws. For a constant density thin shear layer situation the Reynolds stress $\overline{u'v'}$ is approximated by $-\nu_t \frac{\partial \bar{u}}{\partial y}$. Though similar in form to Newton's constitutive relation for momentum diffusion, $\nu_t (\equiv \frac{\mu_t}{\rho})$ is not a fluid property but depends on the local properties of the turbulence. If the turbulence can be characterised in terms of a single representative length and time scale, L , u' (respectively), then $\nu_t \propto u' L$ becomes a dimensional necessity; this relation forms the basis for constructing a modelled expression. u' can be identified with $\sqrt{k}$, and L with the integral length scale. Therefore the closure is complete provided k and L can be specified. In flows with appreciable density fluctuations it is assumed that the reasoning previously suggested still applies. Thus, the Reynolds stress can be expressed as:

$$\bar{\rho} \overline{u''_i u''_j} = \underbrace{-\mu_t \left(\frac{\partial \tilde{u}_i}{\partial x_j} + \frac{\partial \tilde{u}_j}{\partial x_i} \right)}_{\text{I}} + \underbrace{\frac{2}{3} \bar{\rho} k \delta_{ij} + \frac{2}{3} \mu_t \frac{\partial \tilde{u}_l}{\partial x_l} \delta_{ij}}_{\text{II}} \quad (2.40)$$

Term I ensures the symmetric property of the Reynolds stress tensor ($\widetilde{u_i'' u_j''} = \widetilde{u_j'' u_i''}$) is preserved. Term II is included to ensure that the left and right hand sides of the equation are identical on contraction of the indices. The modelled scalar flux becomes:

$$\bar{\rho} \widetilde{u_i'' \phi''} = -\frac{\mu_t}{\sigma_t} \frac{\partial \tilde{\phi}}{\partial x_i} \quad (2.41)$$

where σ_t is a turbulent Prandtl/Schmidt number. To obtain the eddy-viscosity several levels of closure are possible, ranging from simple zero equation mixing length models to the widely used two equation models (Launder and Spalding [59]). Focusing attention on the latter (two-equation) specification, a popular choice of closure employs k and ε . Energy cascade arguments suggest that the large scale turbulent motions dictate the rate of energy transfer down to the fine scales. Consequently $\varepsilon \sim \mathcal{O}(\frac{k^{3/2}}{L})$ so that the eddy viscosity becomes, $\nu_t \propto \frac{k^2}{\varepsilon}$ and so $\mu_t \equiv C_\mu \bar{\rho} \frac{k^2}{\varepsilon}$. This is referred to as the ' k - ε ' model [45], where C_μ is a constant to be determined and where k and ε are obtained from the solution of modelled transport equations. In equation (2.39), the terms requiring closure appear on the rhs. The transport term and the mean pressure gradient term are approximated using a gradient transport hypothesis:

$$-\left\{ \frac{1}{2} \overline{\rho u_j'' u_i'' u_i''} + \overline{u_j'' p'} \right\} \approx \frac{\mu_t}{\sigma_k} \frac{\partial k}{\partial x_j} \quad (2.42)$$

$$-\overline{u_i'' \frac{\partial \bar{p}}{\partial x_i}} = \frac{\overline{\rho' u_i'}}{\bar{\rho}} \frac{\partial \bar{p}}{\partial x_i} = \frac{1}{\bar{\rho}^2} \left(\bar{\rho} \overline{\rho' u_i'} \frac{\partial \bar{p}}{\partial x_i} \right) \approx -\frac{1}{\bar{\rho}^2} \left(\frac{\mu_t}{\sigma_\rho} \frac{\partial \bar{\rho}}{\partial x_i} \right) \frac{\partial \bar{p}}{\partial x_i} \quad (2.43)$$

The approximation expressed in equation (2.43) was introduced by Jones and McGuirk [47], but in many flows it makes only a small contribution and is often neglected. Substituting the above into equation (2.39) yields the modelled form of the k -equation:

$$\bar{\rho} \frac{Dk}{Dt} + \bar{\rho} \widetilde{u_i'' u_j''} \frac{\partial \tilde{u}_j}{\partial x_i} = \frac{\partial}{\partial x_j} \left(\frac{\mu_t}{\sigma_k} \frac{\partial k}{\partial x_j} \right) - \frac{\mu_t}{\bar{\rho}^2} \frac{\partial \bar{\rho}}{\partial x_i} \frac{\partial \bar{p}}{\partial x_i} - \bar{\rho} \varepsilon \quad (2.44)$$

The derivation of a transport equation for ε , as defined in equation (2.35), is complicated and exceedingly laborious. For constant density flows, where $\tau'_{lk} = \mu(\partial u'_l/\partial x_k + \partial u'_k/\partial x_l)$, the viscous dissipation rate can be expressed as $\nu(\partial u'_l/\partial x_k)^2$ since by invoking local isotropy the contribution to ε from $\overline{(\partial u'_l/\partial x_k)(\partial u'_k/\partial x_l)}$ is equal to zero. A transport equation for this quantity can be obtained by straightforward manipulation of equation (2.6); the equation for ε in constant density flows can be written as,

$$\begin{aligned} \underbrace{\frac{d\varepsilon}{dt}}_{\mathcal{O}(1)} = & - \underbrace{\frac{\partial}{\partial x_k} \left\{ \nu u'_k \left(\frac{\partial u'_i}{\partial x_l} \right)^2 \right\}}_{\text{I} \sim \mathcal{O}(1)} - \underbrace{\frac{2\nu}{\rho} \frac{\partial}{\partial x_k} \left\{ \frac{\partial p'}{\partial x_l} \frac{\partial u'_k}{\partial x_l} \right\}}_{\text{II} \sim \mathcal{O}(Re^{-\frac{1}{2}})} + \underbrace{\nu \frac{\partial^2 \varepsilon}{\partial x_k \partial x_k}}_{\text{III} \sim \mathcal{O}(Re^{-1})} \\ & - \underbrace{2\nu \frac{\partial \bar{u}_i}{\partial x_k} \left\{ \frac{\partial u'_i}{\partial x_l} \frac{\partial u'_k}{\partial x_l} + \frac{\partial u'_l}{\partial x_i} \frac{\partial u'_l}{\partial x_k} \right\}}_{\text{IV} \sim \mathcal{O}(Re^{-\frac{1}{2}})} - \underbrace{2\nu u'_k \frac{\partial u'_i}{\partial x_l} \frac{\partial^2 \bar{u}_i}{\partial x_k \partial x_l}}_{\text{V} \sim \mathcal{O}(Re^{-\frac{1}{2}})} \\ & - \underbrace{2\nu \frac{\partial u'_i}{\partial x_k} \frac{\partial u'_i}{\partial x_l} \frac{\partial u'_k}{\partial x_l}}_{\text{VI} \sim \mathcal{O}(Re^{\frac{1}{2}})} - \underbrace{2\nu^2 \left(\frac{\partial^2 u'_i}{\partial x_l \partial x_k} \right)^2}_{\text{VII} \sim \mathcal{O}(Re^{\frac{1}{2}})} \end{aligned} \quad (2.45)$$

where the operator $\frac{d}{dt} \equiv \frac{\partial}{\partial t} + \bar{u}_k \frac{\partial}{\partial x_k}$, and where $Re \equiv \frac{u' L}{\nu}$. The maximum order of magnitude of each term is indicated in equation (2.45) and at high Reynolds numbers the dissipation rate equation reduces to,

$$\frac{d\varepsilon}{dt} = - \underbrace{\frac{\partial}{\partial x_k} \left\{ \overline{\nu u'_k \left(\frac{\partial u'_i}{\partial x_l} \right)^2} \right\}}_{I^*} - \underbrace{2\nu \frac{\partial u'_i}{\partial x_k} \frac{\partial u'_i}{\partial x_l} \frac{\partial u'_k}{\partial x_l}}_{II^*} - \underbrace{2\nu^2 \left(\frac{\partial^2 u'_i}{\partial x_l \partial x_k} \right)^2}_{III^*} \quad (2.46)$$

The terms appearing on the rhs require modelling. The order of magnitude estimates show that terms II^* and III^* individually vary as $Re^{1/2}$ compared with the other terms, and so in the limit, become infinite. But when taken together, they have to form a finite value in order for the equation to balance. Terms II^* and III^* should be modelled together in order to avoid their respective singular behaviour as $Re \rightarrow \infty$. However, the exact equation is of little help in the modelling partly because of the singular behaviour of terms II^* and III^* . Instead the modelling is based on the following form [42]:

$$\frac{d\varepsilon}{dt} = - \underbrace{\frac{\partial}{\partial x_k} \left\{ \overline{\nu u'_k \left(\frac{\partial u'_i}{\partial x_l} \right)^2} \right\}}_{I^*} + \varphi_o + \frac{\varepsilon}{k} \varphi_1 \quad (2.47)$$

The grouping $(\varphi_o + \frac{\varepsilon}{k} \varphi_1)$ is the modelled representation of the source terms (i.e. the net creation rate of ε), where φ_o is not an explicit function of ε . The appearance of $\frac{k}{\varepsilon}$ appears to be a natural choice for the time scale (which is representative of the energy-containing motions), though this does not imply that it is sufficient to characterise the evolution of ε .

The basic requirement imposed on the solutions of equation (2.47) are that $\varepsilon, k \geq 0$ and that as $k \rightarrow 0$, then $\varepsilon \rightarrow 0$. In the limit, if $k = 0$ then $\varepsilon = 0$ too, otherwise it is possible for k to become negative, as implied by equation (2.39). Based on these requirements, the following constraints apply to φ_o and φ_1 ,

- $\varphi_o \geq 0$: In order to illustrate why $\varphi_o \geq 0$, the turbulent transport term (I^*) in equation (2.47) is neglected and φ_1 is set to zero. Although this is not a physically meaningful test case it is a useful foundation upon which a robust modelled equation for ε can be constructed. If $\varphi_o \leq 0$ then from equation (2.47), $\frac{d\varepsilon}{dt} \leq 0$; consequently when the condition $\varepsilon = k = 0$ is attained, the modelled equation would allow $\varepsilon \leq 0$, which is unphysical. With eddy-viscosity closures $\varphi_o \geq 0$,

$$\varphi_o = \frac{C_{\varepsilon_1} C_\mu}{2} \left(\frac{\partial \bar{u}_i}{\partial x_j} + \frac{\partial \bar{u}_j}{\partial x_i} \right)^2 k \geq 0 \quad (2.48)$$

where equation (2.48) stems from the production term. In conventional second moment closures, $\varphi_o = 0$

- φ_1 must remain bounded as $\varepsilon \rightarrow 0$: This can be explained by considering the model equation with the transport and φ_o terms set to zero, leaving $\frac{d\varepsilon}{dt} = \frac{\varepsilon}{k} \varphi_1$. If φ_1 is unbounded and negative, as $\varepsilon, k \rightarrow 0$, $\frac{d\varepsilon}{dt}$ can become so large in magnitude that ε could undershoot and become negative.

- $\varphi_o + \frac{\varepsilon}{k}\varphi_1 \rightarrow -\infty$ as $k \rightarrow 0$. This condition is required so that $\varepsilon \rightarrow 0$ as $k \rightarrow 0$. In the limit, if $k = 0$, and ε is still finite, this condition ensures that $\varepsilon \rightarrow 0$ instantly.

A well-tested and widely-used form of the modelled ε equation for flows involving small density fluctuations is:

$$\frac{d\varepsilon}{dt} = \frac{\partial}{\partial x_j} \left(\frac{\nu_t}{\sigma_\varepsilon} \frac{\partial \varepsilon}{\partial x_j} \right) - C_{\varepsilon_1} \frac{\varepsilon}{k} \overline{u_i' u_j'} \frac{\partial \bar{u}_i}{\partial x_j} - C_{\varepsilon_2} \frac{\varepsilon^2}{k} + C_{\varepsilon_3} \frac{\varepsilon}{k} g_i \overline{\rho' u_i'} \quad (2.49)$$

Recast in terms of a variable density formulation, equation (2.49) becomes,

$$\bar{\rho} \frac{D\varepsilon}{Dt} = \frac{\partial}{\partial x_j} \left(\frac{\mu_t}{\sigma_\varepsilon} \frac{\partial \varepsilon}{\partial x_j} \right) - C_{\varepsilon_1} \frac{\varepsilon}{k} \overline{\bar{\rho} u_i' u_j'} \frac{\partial \bar{u}_i}{\partial x_j} - C_{\varepsilon_2} \bar{\rho} \frac{\varepsilon^2}{k} - C_{\varepsilon_3} \frac{\varepsilon}{k} \frac{\mu_t}{\bar{\rho}^2} \frac{\partial \bar{\rho}}{\partial x_i} \frac{\partial \bar{p}}{\partial x_i} \quad (2.50)$$

In discussing the k - ε closure several constants were introduced. These constants are calibrated in simple (constant density) flows, such as decaying grid turbulence ($\Rightarrow C_{\varepsilon_2}$) and near-wall flows ($\Rightarrow C_{\varepsilon_1}, C_\mu$). The following values are commonly employed [43]: $C_\mu = 0.09$; $C_{\varepsilon_1} = 1.44$; $C_{\varepsilon_2} = 1.92$; $C_{\varepsilon_3} \approx 1.0$; $\sigma_k = 1.0$; $\sigma_\varepsilon = 1.3$. For free flows $\sigma_t = 0.7$ and near-wall flows $\sigma_t = 0.9$. For a round jet issuing into stagnant surrounds improved spreading rates in the self-similar region are obtained with $C_{\varepsilon_2} = 1.85$ and $C_\mu = 0.065$.

The k - ε model is robust and has been (and still is) widely used; it can lead to useful results in many circumstances. However it is clearly a very simple representation of what is undoubtedly an exceedingly complex phenomenon and consequently it does have its limitations. Specifically, it is found to be incapable of accurately reproducing the effects of mean swirling motions on mixing rates, the stabilizing/de-stabilizing influences of buoyancy forces and countergradient diffusion effects (as observed in premixed flames). A more detailed and accurate description of turbulent transport is needed.

2.5.2 Second Moment Closures

Higher order closures are employed to provide a more accurate description of turbulent transport, and second-moment (single-point) closures represent about the simplest level at which this can be achieved. The equations to be solved are the Reynolds stress equation (2.33) and the scalar flux equation:

$$\underbrace{\bar{\rho} \frac{D}{Dt} \overline{u_i'' \phi_\alpha''}}_{\text{I}} + \underbrace{\overline{\bar{\rho} u_j'' \phi_\alpha''} \frac{\partial \bar{u}_i}{\partial x_j} + \overline{\bar{\rho} u_i'' u_j''} \frac{\partial \bar{\phi}_\alpha}{\partial x_j}}_{\text{II}} = \underbrace{- \frac{\partial}{\partial x_j} \left(\overline{\bar{\rho} u_i'' u_j'' \phi_\alpha''} \right)}_{\text{III}} - \underbrace{\overline{\phi_\alpha'' \frac{\partial \bar{p}'}{\partial x_i}}}_{\text{IV}} - \underbrace{\overline{\phi_\alpha'' \frac{\partial \bar{p}}{\partial x_i}}}_{\text{V}} + \underbrace{\overline{u_i'' \dot{S}_\alpha}}_{\text{VI}} \quad (2.51)$$

All terms on the rhs of equations (2.33) and (2.51) require closure. In equation (2.51) term IV is referred to as 'pressure scrambling' and term VI is a velocity-reaction source correlation term. As in the discussion of equation (2.33), order of magnitude and local isotropy arguments can be employed to neglect terms like $\overline{\phi_\alpha'' (\partial \tau_{ik} / \partial x_k)}$ and $\overline{u_i'' (\partial J_{k,\alpha} / \partial x_k)}$. The use of equation (2.51) has so far been restricted to situations where $\overline{u_i'' \dot{S}_\alpha} = 0$ (even in combustions flows) by making a suitable choice of ϕ_α , i.e. a strictly conserved scalar (section 3.2). For the time being

a conserved scalar, ξ , can be viewed as a normalized element mass fraction which is, of course, neither consumed nor produced. This is to be distinguished from the concept of a *passive* scalar which is a quantity whose evolution does not affect the dynamics of the flow⁴. In order to complete the picture for combustng flows within the context of second-moment closures, the scalar variance equation is also required:

$$\bar{\rho} \frac{D\phi''^2}{Dt} + \underbrace{2\bar{\rho}u_j''\phi'' \frac{\partial \phi''}{\partial x_j}}_{\text{I}} = - \underbrace{\frac{\partial}{\partial x_j} \left(\bar{\rho}u_j''\phi''^2 \right)}_{\text{II}} - \underbrace{2\bar{\rho}\tilde{\chi}_\alpha}_{\text{III}} + \underbrace{\phi''_\alpha \dot{S}_\alpha}_{\text{IV}} \quad (2.52)$$

The terms appearing in equation (2.52) are as follows: I - generation rate of scalar fluctuations; II - turbulent 'diffusion'; III - dissipation rate of ϕ''^2 ; IV - source rate of production/consumption of ϕ''^2 due to chemical reaction. Note that the scalar dissipation $\tilde{\chi}_\alpha$ in equation (2.52) is defined, for the case of Fickian diffusion, as $(\bar{\rho})^{-1}(\mu/Sc)(\nabla \phi''_\alpha)^2$. The closure problem is now set; all terms appearing on the rhs of equations (2.33), (2.51) and (2.52) require modelling. The work described in this thesis treats the scalar field using a transported pdf approach, therefore a detailed discussion of the modelling of the scalar flux and variance equations is omitted. With regards to the velocity field, only the Reynolds stress closure proposed by Jones and Musonge [48] is discussed since this is used in the calculations. Consequently only a brief discussion of second moment closures will be given here, whilst a more detailed review can be found in the works of Jones [41, 42] and Launder [58].

2.5.2.1 Transport Terms

The simplest level of approximation is a gradient transport hypothesis, originally proposed by Daly and Harlow [18] for the triple correlation $\overline{\rho u_i'' u_j'' u_k''}$; in density-weighted form the results can be expressed as:

$$\left(\overline{\rho u_i'' u_j'' u_k''} + \frac{2}{3} \delta_{ij} \overline{u_k'' p'} \right) = -C_s \bar{\rho} \frac{k}{\varepsilon} u_k'' u_m'' \frac{\partial}{\partial x_m} \left(u_i'' u_j'' \right) \quad (2.53)$$

where $C_s \approx 0.22$. The appearance of $\frac{2}{3} \delta_{ij} \overline{u_k'' p'}$ arises from the decomposition described in equation (2.37); this contribution to the turbulent transport is sometimes referred to as pressure diffusion. More elaborate (and complicated) models have been proposed than that expressed in equation (2.53), for example where the pressure diffusion term is represented separately; however, in constant density thin shear layer flows the transport terms do not provide a major contribution to the second moment balance equations. This serves to justify the use of a gradient transport hypothesis for the transport terms.

2.5.2.2 Viscous Destruction of $u_i'' u_j''$

The treatment of term IV in equation (2.33) has already been discussed and its evaluation requires knowledge of ε , as exemplified by equation (2.35). In the context of the eddy-viscosity closure, ε can be obtained in terms of equation (2.50); the form of the modelled ε -equation for second

⁴The study of passive scalars is of considerable importance, for example in the discharge of fluids into the environment (water discharges into rivers and lakes involving a 'small' temperature difference, or gas releases into the atmosphere). The mixture fraction is a strictly conserved scalar, however, in variable density situations (inert or non-reacting), it is not a passive scalar.

moment closures is slightly different in relation to the turbulent diffusion term. A gradient transport hypothesis is still employed, and the form originally proposed by Hanjalić and Launder [34] is commonly used,

$$\bar{\rho} \frac{D\varepsilon}{Dt} = \frac{\partial}{\partial x_i} \left(C_\varepsilon \bar{\rho} \frac{k}{\varepsilon} \widetilde{u_i'' u_j''} \frac{\partial \varepsilon}{\partial x_j} \right) - C_{\varepsilon_1} \frac{\varepsilon}{k} \bar{\rho} \widetilde{u_i'' u_j''} \frac{\partial \bar{u}_j}{\partial x_i} - C_{\varepsilon_2} \bar{\rho} \frac{\varepsilon^2}{k} - C_{\varepsilon_3} \frac{\varepsilon}{k} \bar{\rho} \frac{\partial \bar{p}}{\partial x_i} \quad (2.54)$$

Note that since $\widetilde{u_i'' u_j''}$ is the subject of the Reynolds stress equation, this information can be used to calculate the turbulence kinetic energy without making recourse to a modelled k -equation.

2.5.2.3 Fluctuating Pressure Terms

A principal difficulty in constant and variable density flows is the fluctuating pressure terms. These include the fluctuating velocity-pressure gradient correlation (term V in equation (2.33)) responsible for the transfer amongst the components of $\widetilde{u_i'' u_j''}$, and the pressure scrambling (term IV in equation (2.51)). For constant density flows Chou [16] has shown that they can be related to volume integrals involving two-point correlations:

$$\begin{aligned} \frac{\overline{u_i \frac{\partial p}{\partial x_j}}}{\rho} = & \frac{1}{4\pi} \int \underbrace{\frac{\partial^3 \overline{u'_l \mathbf{x}' u'_m \mathbf{x}' u'_n \mathbf{x}'} }{\partial r_m \partial r_l \partial r_j}}_{(a)} + 2 \underbrace{\frac{\partial \bar{u}_l \mathbf{x}}{\partial x_m} \frac{\partial^2 \overline{u'_m \mathbf{x}' u'_n \mathbf{x}'} }{\partial r_l \partial r_j}}_{(b)} + g_l \underbrace{\frac{\partial^2 \overline{u'_l \mathbf{x}' \rho \mathbf{x}'} }{\partial r_l \partial r_j}}_{(c)} \frac{d\text{Vol}}{|\mathbf{x}' - \mathbf{x}|} \\ & + \text{Surface integral} \end{aligned} \quad (2.55)$$

The terms on the rhs with subscript $\mathbf{x}' (\equiv \mathbf{x} + \mathbf{r})$ are evaluated at that position, with the integrations carried over $\mathbf{r}$ space. The surface integral is important only when the typical size of the energy-containing eddies is of the same order as the distance from a wall. In many combustion systems this contribution is ignored, partly because the ability to reproduce very accurately the wall friction and turbulence structure close to solid surfaces is often of secondary importance [42]. Equation (2.55) contains three terms labelled as (a), (b) and (c). Term (a) involves turbulence quantities only, term (b) is affected by mean strain rates and term (c) represents the buoyancy contribution. In the decay of homogeneous, strain-free and initially isotropic turbulence which has been distorted so that $\overline{u'^2} \neq \overline{v'^2} \neq \overline{w'^2}$, the high Re (constant density) stress equations become:

$$\frac{d\overline{u'^2}}{dt} = 2 \frac{\bar{p}}{\rho} \frac{\partial \bar{u}'}{\partial x} - \frac{2}{3} \varepsilon; \quad \frac{d\overline{v'^2}}{dt} = 2 \frac{\bar{p}}{\rho} \frac{\partial \bar{v}'}{\partial x} - \frac{2}{3} \varepsilon; \quad \frac{d\overline{w'^2}}{dt} = 2 \frac{\bar{p}}{\rho} \frac{\partial \bar{w}'}{\partial x} - \frac{2}{3} \varepsilon \quad (2.56)$$

It is assumed that this type of turbulence 'returns to isotropy', an assumption which is supported by experiment [93]. Consequently term (a) is referred to as the return component. The following definition is introduced:

$$A_{ij} \equiv - \left\{ \overline{u_i'' \frac{\partial \bar{p}}{\partial x_j}} + \overline{u_j'' \frac{\partial \bar{p}}{\partial x_i}} - \frac{2}{3} \delta_{ij} \overline{u_k'' \frac{\partial \bar{p}}{\partial x_k}} \right\} \quad (2.57)$$

The three contributions ((a), (b) and (c)) towards A_{ij} are denoted as $A_{ij}^{(1)}$, $A_{ij}^{(2)}$, $A_{ij}^{(3)}$ respectively. One popular approach has been to provide closure approximations for each of these terms separately. The modelling of the buoyancy term is not discussed since it is not of primary importance to the work described in this thesis. The basis for modelling $A_{ij}^{(1)}$ is the concept of return to

isotropy. Rotta [84] assumed that the rate of return to isotropy, $A_{ij}^{(1)}$, is proportional to the degree of anisotropy, $b_{ij} (\equiv (\overline{u_i'' u_j''}/k) - (2/3)\delta_{ij})$. In density-weighted form, the model is:

$$A_{ij}^{(1)} = -C_1 \bar{\rho} \varepsilon b_{ij} \quad (2.58)$$

where C_1 is the constant of proportionality obtained from measurements (e.g. [93]). It is found that the degree of scatter in the measurements leads to the following popular choices for C_1 : 1.5, 1.8 and 3.0; the various values of C_1 that can be specified demonstrate the inadequacy of the linear theory, however its simplicity and the fact that it leads to reasonable results in practical flows justifies its use. More recently it has been argued that there is a 'tendency to isotropy' such that the return is non-linear as the anisotropy goes to zero. This leads to refinements (with added complexity) in the modelled expression and are not discussed here.

Jones and Musonge [48] point out that an exact balance equation for the 'return-to-isotropy' part of the velocity-pressure gradient correlation in homogeneous turbulence contains mean velocity gradients. Consequently it is argued that because terms (a) and (b) in equation (2.55) are both affected by the mean strain, they can be modelled collectively rather than separately. To formulate a model for $A_{ij}^* (\equiv A_{ij}^{(1)} + A_{ij}^{(2)})$, it is first assumed that it can be expressed as a linear function of the Reynolds stress tensor. This is because models involving non-linear terms may not generate unique steady-state solutions - at least for homogeneous flows. Consequently the following approximation is employed (recast in density-weighted form):

$$A_{ij}^* = -C_1 \bar{\rho} \varepsilon \underbrace{\left(\frac{\overline{u_i'' u_j''}}{k} - \frac{2}{3} \delta_{ij} \right)}_{b_{ij}} + a_{ijlm} \frac{\partial \tilde{u}_l}{\partial x_m} \quad (2.59)$$

In the absence of mean strain the model is identical to that initially proposed by Rotta. The second term involves a_{ijlm} , a linear fourth-rank tensor function of the Reynolds stress tensor. The properties of this tensor (which follow from those of A_{ij}^* , and hence $\overline{u_i'' u_j''}$) are: $a_{ijlm} = a_{jilm}$, this preserves the symmetry of the Reynolds stress tensor; $a_{iilm} = 0$, this ensures that the fluctuating pressure terms are redistributive as originally defined in the decomposition of equation (2.36). The most general fourth-rank linear tensor function of the Reynolds stress, satisfying the above-mentioned properties, is:

$$\begin{aligned} a_{ijlm} = & C_2 \bar{\rho} \delta_{ij} \overline{u_l'' u_m''} + C_3 \bar{\rho} (\delta_{li} \overline{u_m'' u_j''} + \delta_{lj} \overline{u_m'' u_i''}) \\ & + C_4 \bar{\rho} k (\delta_{mi} \delta_{lj} + \delta_{mj} \delta_{li}) + C_5 \bar{\rho} \delta_{lm} \overline{u_i'' u_j''} \\ & + C_6 \bar{\rho} (\delta_{mi} \overline{u_l'' u_j''} + \delta_{mj} \overline{u_l'' u_i''}) + C_7 \bar{\rho} k \delta_{ij} \delta_{lm} \end{aligned} \quad (2.60)$$

Contracting the indices ($i = j$) and equating the coefficients of the terms involving $\overline{u_l'' u_m''}$ and $\delta_{lm} k$ yields two relations which allow the elimination of $C_6 (= -\frac{3}{2}C_2 - C_3)$ and $C_7 (= -\frac{2}{3}C_4 - \frac{2}{3}C_5)$. The equation for A_{ij}^* then becomes:

$$\begin{aligned} A_{ij}^* = & - C_1 \bar{\rho} \varepsilon b_{ij} + C_2 \bar{\rho} \delta_{ij} \overline{u_l'' u_m''} \frac{\partial \tilde{u}_l}{\partial x_m} + C_3 \bar{\rho} \left(\overline{u_m'' u_j''} \frac{\partial \tilde{u}_i}{\partial x_m} + \overline{u_m'' u_i''} \frac{\partial \tilde{u}_j}{\partial x_m} \right) \\ & + C_4 \bar{\rho} k \left(\frac{\partial \tilde{u}_i}{\partial x_j} + \frac{\partial \tilde{u}_j}{\partial x_i} \right) + C_5 \bar{\rho} \overline{u_i'' u_j''} \frac{\partial \tilde{u}_l}{\partial x_l} \\ & - \left(\frac{3}{2}C_2 + C_3 \right) \bar{\rho} \left(\overline{u_l'' u_j''} \frac{\partial \tilde{u}_l}{\partial x_i} + \overline{u_l'' u_i''} \frac{\partial \tilde{u}_l}{\partial x_j} \right) - \frac{2}{3} (C_4 + C_5) \bar{\rho} \delta_{ij} k \frac{\partial \tilde{u}_l}{\partial x_l} \end{aligned} \quad (2.61)$$

$\widetilde{u''v''}$	$C_s = 0.22$	$C_1 = 1.5$	$C_2 = -0.53$	$C_3 = 0.67$	$C_4 = -0.12$
$\widetilde{u''v''}^a$	$C_s = 0.22$	$C_1 = 3.0$	$C_2 = -0.44$	$C_3 = 0.46$	$C_4 = -0.23$
ε	$C_\varepsilon = 0.18$	$C_{\varepsilon_1} = 1.40^b$	$C_{\varepsilon_2} = 1.90$		

Table 2.1: Model constants appearing in the Reynolds stress and viscous dissipation rate equations.

^aThe constants appearing in this row have been re-calibrated using $C_1 = 3.0$, and are now the recommended set of values.

^b $C_{\varepsilon_1} = 1.44$ and $C_{\varepsilon_2} = 1.92$ are used in the context of the k - ε closure. The term involving C_{ε_3} is quite often ignored, as in the present calculations.

The terms involving $\frac{\partial \tilde{u}_l}{\partial x_l}$ make no contribution in constant density flows. In variable density boundary layer type flows, the components of $|\frac{\partial \tilde{u}_l}{\partial x_l}|$ are negligible in comparison to $\frac{\partial \tilde{u}_1}{\partial x_2}$, where '2' denotes the direction normal to the mainstream flow. Consequently, under these circumstances, knowledge of the magnitude of C_5 is immaterial. The form of the modelled Reynolds stress equation can be written as,

$$\begin{aligned} \bar{\rho} \frac{D}{Dt} (\widetilde{u''_i u''_j}) + \bar{\rho} \widetilde{u''_i u''_k} \frac{\partial \tilde{u}_j}{\partial x_k} + \bar{\rho} \widetilde{u''_j u''_k} \frac{\partial \tilde{u}_i}{\partial x_k} &= C_s \frac{\partial}{\partial x_k} \left\{ \bar{\rho} \frac{k}{\varepsilon} \widetilde{u''_k u''_m} \frac{\partial}{\partial x_m} (\widetilde{u''_i u''_j}) \right\} \\ &- \frac{2}{3} \delta_{ij} \bar{\rho} \varepsilon + A_{ij}^* \end{aligned} \quad (2.62)$$

The mean pressure gradient term has not been included in equation (2.62). In the calculations reported in this thesis such an omission is justifiable, since for a turbulent jet issuing into unconfined surrounds the mean pressure gradient is zero. Therefore, equation (2.62) can be considered as the modelled transport equation for the Reynolds stress in the work to be described in subsequent chapters.

The Reynolds stress model of Jones and Musonge [48] involves four independent constants ($C_l, l = 1, \dots, 4$) whose values remain to be determined. C_1 has already been discussed in relation to Rotta's hypothesis, leading to equation (2.58). The remaining constants are associated with the mean strain and experimental data from an homogeneous turbulent field subjected to a constant mean shear strain is used to estimate their value. In addition to the flows considered by Jones and Musonge [48], the model has been applied with good results to a range of flows including confined swirling flows both with and without density variation and a single jet issuing into a cross-flow. However, the values of the 'constants' have been re-calibrated according to $C_1 = 3.0$ in order to improve the robustness of the model predictions [42]. These constants, in addition to the others encountered in the discussion of the dissipation rate equation for second moment closures, are presented in table 2.1.

The modelling of the pressure scrambling term (IV in equation (2.51)) is not described here since a scalar flux closure is not employed. Closure approximations for this term can be found in the aforementioned review papers.

2.5.2.4 Scalar Variance Dissipation Rate

The scalar dissipation rate, appearing in equation (2.52), plays an important role in combustion modelling. $\tilde{\chi}_\alpha$ can be interpreted as the rate at which fluctuations in $\phi_\alpha(\mathbf{x}, t)$ are reduced by

molecular diffusion. Following Spalding's [88] idea, the simplest method of approximating the scalar dissipation term is to presume that the ratio of the scalar to mechanical turbulence time scale is constant, so that:

$$\tilde{\chi}_\alpha \approx \frac{C_D}{2} \frac{\varepsilon}{k} \widetilde{\phi_\alpha'^2} \quad (2.63)$$

where $(\frac{C_D}{2})^{-1}$ is the time scale ratio with $C_D \approx 2.0$. In fact studies in decaying grid turbulence suggest that the time scale ratio can vary from 0.75 to 3.3. Newman et al. [70] point out that there is good reason to expect that $(\frac{C_D}{2})^{-1}$ should vary in different types of flows. It is argued that since $\frac{k}{\varepsilon}$ and $(\widetilde{\phi_\alpha'^2} \cdot \tilde{\chi}_\alpha^{-1})$ are proportional to the local turnover times of the energy-containing eddies, and that these motions are significantly influenced by the production mechanisms of the respective velocity and scalar fields, the magnitude of $(\frac{C_D}{2})^{-1}$ may well depend on the production mechanisms; this may even be true for a flow where there is a varying influence of the production mechanism. Nevertheless, the approximation expressed in equation (2.63) has been widely used and gives reasonable results in many thin shear layer flows. A more general way of obtaining $\tilde{\chi}_\alpha$ is from a modelled transport equation [48].

The treatment of second moment closures is now complete. The model is considerably more elaborate than their eddy-viscosity counterparts and the resulting improvement in the ability to predict complex flow fields has been amply demonstrated, particularly in situations where k - ε is inadequate. Though the computational cost is larger than that required by the k - ε model, it is not prohibitive and the use of second moment closures is currently the most detailed approach feasible for complex and practical problems.

2.6 Mean Reaction Rates

This chapter is concluded with a brief description of the other source of difficulty in the averaged conservation equations. This is associated with the thermochemical field and manifests itself through the appearance of the terms $\dot{\bar{S}}_\alpha$ and $\bar{q}$. In principle,

$$\dot{\bar{S}}_\alpha \equiv \lim_{N_r \rightarrow \infty} \frac{1}{N_r} \sum_{i=1}^{N_r} \dot{S}_\alpha^{(i)}(T, \varrho, \phi_1, \dots, \phi_N) \quad \text{for} \quad \alpha = 1, \dots, N \quad (2.64)$$

The form of $\dot{S}_\alpha^{(i)}$ is given by equation (2.19). The non-linearity of the expression precludes the evaluation of the mean reaction rate in terms of the averaged scalar properties. In fact such a practice would lead to substantial errors, since in turbulent combustion the fluctuations in scalar properties are comparable (in magnitude) to the mean properties. A mathematically rigorous and computationally tractable way of calculating the mean formation rates is through the use of a joint probability density function (pdf) of the species mass fractions and the temperature.

The approaches which use the concept of a joint scalar pdf are presented in the subsequent chapters. Initially those methods where the shape of the pdf is presumed are described, followed by a discussion of the transported pdf approach. In this latter technique, the exact evolution equation for the joint pdf is modelled and then solved.

Chapter 3

Combustion Models for Non-Premixed Configurations

3.1 Introduction

This chapter is concerned with combustion modelling in non-premixed configurations. The discussion will centre around the conserved scalar formalism, hence the emphasis on 'non-premixed' combustion. In the next chapter, the transported pdf approach will be discussed and whilst the method will be applied to non-premixed systems, it is important to bear in mind that this particular formalism can be used to describe premixed combustion too.

The application of the averaging process to the specie transport equation leads to the mean reaction rate term. Direct attempts at closure are unfeasible for the general case of highly exothermic combustion. In fact, to date, no suitable deterministic approximation for evaluating this term has been obtained, and consequently it seems natural to resort to probabilistic approaches to achieve closure.

Based on the assumptions invoked in section 2, it is clear that the mean reaction rate term, $\bar{\dot{S}}_\alpha$, is in general a function of the scalar field $\underline{\phi} = \phi_\alpha$ ($\alpha = 1, \dots, N + 1$) where ϕ_α includes the species mass fractions as well as the enthalpy. By introducing the pdf of the scalar field $P_{\underline{\phi}}(\underline{\psi})$, where $\underline{\psi}$ denotes the sample space of $\underline{\phi}$, closure for the mean reaction rate (and for the entire scalar field) can be obtained from the following basic property of a pdf:

$$\bar{\dot{S}}_\alpha(\underline{\phi}) = \int_{\underline{\psi}} \dot{S}_\alpha(\underline{\psi}) P_{\underline{\phi}}(\underline{\psi}) d\underline{\psi} \quad ; \quad \bar{\phi}_\alpha(\mathbf{x}, t) = \int_{\underline{\psi}} \psi_\alpha P_{\underline{\phi}}(\underline{\psi}) d\underline{\psi} \quad (3.1)$$

Note that $P_{\underline{\phi}}(\underline{\psi})$ is formally referred to as the single point, single time, Eulerian joint pdf of the scalar field. In writing the above, several features remain outstanding. Firstly, a thermochemical model is required so that $\dot{S}_\alpha$ is known, on an instantaneous basis, as a function of the scalar domain, $\underline{\psi}$. Secondly, the pdf $P(\underline{\psi})$ (where the subscript on $\underline{\phi}$ is dropped for conciseness) needs to be specified. Assuming these quantities are known, the next stumbling block is the integration over the scalar space. The integrals written in equation (3.1) are multi-dimensional and the cpu requirements for their computation are considerable. It is therefore desirable to reduce the dimensionality of the problem, ideally to a single scalar which uniquely characterizes the thermochemistry of the reacting mixture.

3.2 The Conserved Scalar Formalism

The basic framework upon which most of the traditional theory of non-premixed combustion has been developed is the conserved scalar formalism. Under this formalism, a *strictly conserved scalar* is a quantity whose instantaneous evolution equation is given by:

$$\frac{\partial}{\partial t}(\rho z_\alpha) + \frac{\partial}{\partial x_k}(\rho u_k z_\alpha) = \frac{\partial}{\partial x_k} \left(\rho D \frac{\partial z_\alpha}{\partial x_k} \right), \quad \alpha = 1, \dots, N+1 \quad (3.2)$$

No source terms appear in equation (3.2). This requirement is naturally satisfied by quantities such as element mass fractions, which are not created nor destroyed by chemical reaction, but are simply convected and diffused. Note, however, that chemical reaction influences the evolution of z_α through the density, ρ . The next important feature of equation (3.2) is the presence of a single diffusion coefficient. Since z_α can represent elemental concentrations as well as enthalpy, the use of a single diffusion coefficient implies uniform Prandtl and Schmidt numbers as well as unity Lewis number. The effects of differential diffusion are clearly ignored in this situation.

With the assumption of equal diffusivity, the element mass fraction transport equations all satisfy the same form of differential equation, differing only in the initial and boundary conditions. The assumptions of low Mach number, constant thermodynamic pressure and negligible radiative heat losses to the surroundings (guaranteed by a no heat flux condition on all bounding surfaces) implies that the enthalpy also satisfies a conserved scalar transport equation. Following Bilger [3], for a *two-stream* mixing process, with each stream containing a uniform, constant and different composition, the following normalization can be applied:

$$\xi_\alpha(\mathbf{x}, t) \equiv \frac{z_\alpha(\mathbf{x}, t) - z_{\alpha,2}(\mathbf{x}, t)}{z_{\alpha,1}(\mathbf{x}, t) - z_{\alpha,2}(\mathbf{x}, t)} \quad (3.3)$$

where z_α can denote element mass fraction or enthalpy and ξ_α is referred to as the *mixture fraction for specie α* . Through this normalization the transport equations for the various ξ_α are of the same form and their boundary conditions are identical too. It can then be demonstrated that after an initial transient ($t > t_o$) all ξ_α at any place and time are equal, even on an instantaneous basis. Thus,

$$\xi_{\alpha,1}(\mathbf{x}, t) = \xi_{\alpha,2}(\mathbf{x}, t) = \dots \equiv \xi(\mathbf{x}, t) \quad (3.4)$$

In summary, subject to certain assumptions, all conserved scalars are linearly related. Consequently the problem of calculating several conserved scalars, z_α , can be circumvented by solving for a single normalized conserved scalar, ξ , constructed from any one of the individual conserved z_α . Multiple feed problems can be considered in the same manner as the above provided the streams have the same composition and temperature as in streams '1' and '2'. Any relaxation of the assumptions presented above requires the specification of extra scalars. For example, if adiabatic flow is no longer assumed, enthalpy would have to be included as an extra independent scalar since it would no longer be 'conserved' due to the heat losses to the surroundings.

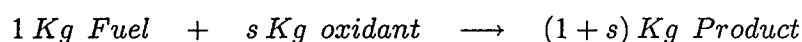
In combustion the characterization of the conserved scalar field in a flow domain is only of secondary importance and obtaining the concentration field of the reactive species is the main objective. Consequently thermochemical models are required which are compatible with the conserved scalar formalism, i.e. where knowledge of ξ instantaneously and uniquely specifies the value of the other scalars. The thermochemical models that satisfy this requirement all share the common assumption of 'fast chemistry'. In the case of the flame-sheet model (to be described in

section 3.3.1), the flame approximates to a geometric surface 'flapping' through the mixture as a result of the influence of turbulent fluctuations. However, on average the reaction zone is of finite thickness, and often described as a turbulent flame brush. For the more realistic proposal based on chemical equilibrium and laminar flamelets, 'thin' flame burning is inherently presumed; with this assumption the transport (aerodynamic) time scales are much longer than the chemical time scales (i.e the Damköhler number, Da , is considerably greater than unity).

3.3 Thermochemical Models

3.3.1 The Flame Sheet Approximation

This model stems from the initial work of Burke and Schumann [9]. One step irreversible global kinetics is employed in the following form:



It is assumed that the reaction proceeds at an infinite rate, implying that as soon as the reactants mix at the molecular scale, they burn instantly to form fully-burned products. As a result, fuel and oxidant cannot co-exist instantaneously except within an infinitesimally thin flame sheet. However, on a time-averaged basis, fuel and air can be present in finite concentrations at a particular point; this is commonly referred to as 'unmixedness' [36].

The representation of a flame zone as a geometric surface is clearly unrealistic, as substantiated by experimental studies in laminar and turbulent flames. Furthermore, in hydrocarbon flames this model precludes the determination of any intermediate species (e.g. CO and H₂) since the global kinetics only allows for the presence of fuel, oxidant and products (CO₂ and H₂O). However, the model is capable of reproducing gross flame characteristics (e.g. mean temperature) reasonably well provided the combustion is unhindered by significant finite-rate effects. The flame sheet methodology has now been superseded by more realistic thermochemical models.

3.3.2 The Chemical Equilibrium Approach

Studies in chemical kinetics demonstrate that one-step irreversible chemistry is a gross simplification. Nearly all reactions are reversible to some degree. The concept of chemical equilibrium can be put to use in the conserved scalar formalism in the following manner. At equilibrium, if the elemental composition and two state variables for the mixture (e.g. the thermodynamic pressure and the enthalpy) are known, then the species concentrations can be determined [32]. Based on the discussion above relating to the conserved scalar concept, it is clear that the elemental composition and the enthalpy are known if the mixture fraction is specified. Because the thermodynamic pressure is presumed constant, knowledge of ξ is sufficient to (instantaneously) characterize the mixture composition at equilibrium. In principle, if the reactions are reversible, fuel and oxidant can co-exist; in this case it can no longer be assumed that the reactions are confined to an infinitely thin flame sheet, but occur where necessary to keep the mixture in equilibrium. It must be stressed that in the chemical equilibrium approach, no input is required from a chemical mechanism. Identifying the participating elemental species and specifying ξ is sufficient.

The chemical equilibrium approach has been widely used, and like the flame sheet model, reasonable temperature and velocity field characteristics can be obtained. The major shortcoming of the approach is related to the prediction of CO levels in hydrocarbon-air flames. For

hydrocarbon-air mixtures it is found [41] that in the case of lean mixtures, negligible quantities of CO are predicted. The proportion of CO increases progressively with mixture strength and for equivalence ratios greater than about 1.2 the CO mole fraction is greater than that of CO₂; at equivalence ratios of around three, CO mole fractions of about 20% can be achieved. These levels are wholly unrealistic in laminar and turbulent hydrocarbon flames. Therefore, although the presence of minor species may not affect the gross flame features significantly, their levels are very poorly reproduced if it is assumed that the fluid at a particular mixture strength is at its equilibrium composition. In reality, mixtures which are too lean or too rich will simply not burn and the same applies if the temperature is too low. Based on these facts, certain ad-hoc modifications have been applied to the chemical equilibrium approach in order to yield more realistic CO levels [31, 24].

3.3.3 The Laminar Flamelet Approach

In this section the basic ideas behind the laminar flamelet technique are outlined. For a more detailed review of the theory, the papers by Williams [102] and Peters [75] should be consulted. At the end of section 3.2, it was pointed out that the laminar flamelet formulation was entirely consistent with (and restricted to) 'thin' flame burning. In fact the thickness of the burning zone must be less than or equal to the Kolmogorov length scale - this fundamental requirement for the existence of flamelets (i.e. laminar-like burning zones in a turbulent medium) is referred to as the Klimov-Williams criterion.

The flame stretch rate, s_r , is a quantity of central importance to laminar flamelet theory and can be defined as the fractional time rate of increase of the flame-zone area,

$$s_r = \frac{1}{\Lambda} \frac{d\Lambda}{dt} \quad (3.5)$$

where Λ represents an area of flame surface. Effects of curvature on the flame front are usually ignored, i.e. locally the flame front is presumed planar.

A turbulent flame is viewed as an ensemble of strained laminar flamelets. The experimental observation that sections of a flame can break away from the bulk structure upstream and possess a highly contorted structure, demonstrates that the flame zone can be a multiply-connected region. However, it is argued that even under these circumstances, provided $Da \gg 1$ and $Ka \ll 1$, the reaction zone is sufficiently thin so that it can instantaneously consume reactants in a 'laminar fashion'. If the degree of flame stretch is too large the flamelet will be quenched, leading to local extinction. Extinction studies of laminar diffusion flames show that the characteristic controlling parameter is Da . However, since a unique quantitative definition of Da cannot be given, it is generally unsuitable for quantifying the flame stretch. The fact that the spatial gradients of scalars increase/decrease in magnitude depending on whether flame stretch/compression is operative [102], suggests that the instantaneous scalar dissipation rate, χ , may be used as a quantitative parameter. If it is assumed that only the flame zone is critically affected by stretch, then the external stretch imposed on the flamelet by the hydrodynamic field has the form of an instantaneous scalar dissipation rate evaluated at stoichiometric conditions $\chi_{st} = \left(\frac{\nu}{Sc}\right)_{st} \left(\frac{\partial \xi}{\partial x_j}\right)_{st}^2$

If the local flame stretch increases, χ_{st} follows suit. As the local scalar gradients steepen, more fresh reactants can enter the flame zone (and enhance the heat release), however at the same time the heat conduction from the reaction zone will increase. If χ_{st} goes above a critical value, χ_q , heat conduction away from the flame zone can no longer be balanced by heat production due to chemical reaction and the flamelet is extinguished. χ_q is affected by thermodynamic and

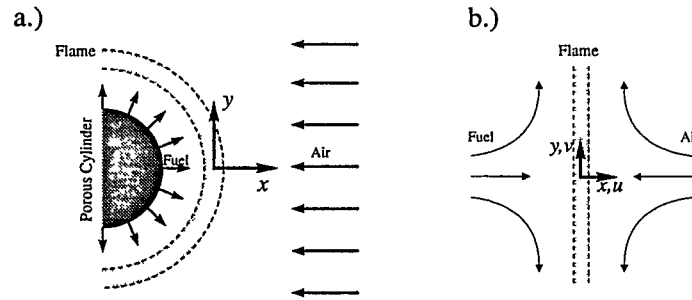


Figure 3.1: a.) Schematic of a laminar counterflow diffusion flame geometry. The width of the flame is exaggerated for clarity. b.) Locally the flame is treated as planar.

chemical-kinetic parameters, and as such it can be evaluated without consideration of turbulence. The basic geometry that is frequently used to investigate flame extinction is a laminar counterflow diffusion flame (figure 3.1) which is established in the forward stagnation region of a porous cylinder. In figure 3.1, the flame thickness is sufficiently thin so that it can be treated as locally planar. Measurements suggest that where the fuel and air merge to form the reaction zone, the flow can be viewed as an opposed jet.

In the above paragraphs the basic ideas behind the laminar flamelet formulation have been expounded. In order to make use of such a formulation in turbulent flame calculations, a thermochemical model is required. The following observation clarifies how this can be achieved. Bilger [5] examined data on laminar counterflow diffusion flames; it was observed that when the results were re-plotted as a function of $\xi^\dagger$ they roughly fell onto a single curve, indicating that the flame configuration satisfied a self-similarity solution. The relations, $\phi_\alpha = \phi_\alpha(\xi)$, can therefore be used as a suitable thermochemical input for the modelling of turbulent diffusion flames. Its significant advantage over the previous models, described in sections 3.3.1 and 3.3.2, is that more realistic chemistry can be introduced into a turbulent flame calculation; differential diffusion effects and their interaction with the flame zone can be accounted for in an indirect manner through $\phi_\alpha(\xi)$. The thermochemical relations ($\phi_\alpha(\xi)$) can be obtained by solving the exact conservation equations for a laminar flame employing detailed kinetics and transport. The most favoured configurations are the steady two-dimensional counterflow diffusion flame geometry and the transient one-dimensional laminar flame. In this manner flamelet 'libraries' may be constructed on an once and for all basis and expressed in the instantaneous form, $\phi_\alpha(\xi, \chi_{st})$. It should be pointed out that one of the disadvantages of the flamelet technique is that for many fuels of engineering interest (e.g. kerosene and petrol) the details of the reaction mechanisms are not adequately known. Furthermore, certain devices operate under fuel lean conditions (e.g. gas turbine combustors) and in such situations the difference in the results between an equilibrium calculation and one based on flamelet thermochemistry is not significantly different.

3.4 The Presumed-Shape Pdf Method

In this section the calculation of the averaged species concentrations, $\tilde{\phi}_\alpha(\mathbf{x}, t)$, is considered. In the present case, $P(\psi)$ is interpreted as $P(\hat{\xi})$. For calculating density-weighted averages, the

[†]For the data of Tsuji and Yamaoka [92], Bilger [5] reported that curves for ξ_C , ξ_H , ξ_O and ξ_N generally fell onto a single curve.

following relation can be employed:

$$\tilde{\phi}_\alpha(\mathbf{x}, t) = \int_0^1 \phi_\alpha(\hat{\xi}) \tilde{P}(\hat{\xi}) d\hat{\xi} \quad (3.6)$$

where the density-weighted pdf is defined as:

$$\tilde{P}(\hat{\xi}) \equiv \frac{\varrho(\hat{\xi}) P(\hat{\xi})}{\bar{\varrho}(\mathbf{x}, t)} \quad \text{and} \quad \bar{\varrho} = \left\{ \int_0^1 \frac{\tilde{P}(\hat{\xi})}{\varrho(\hat{\xi})} d\hat{\xi} \right\}^{-1} \quad (3.7)$$

The non-linearity of the instantaneous relations, $\phi_\alpha(\xi)$, implies that $\tilde{\phi}_\alpha(\mathbf{x}, t) \neq \phi_\alpha(\tilde{\xi})$. In calculation methods the most popular approach is the presumed-shape pdf method. In this approach a two parameter form of the pdf of ξ is chosen and subsequently constructed in terms of its mean and variance; the statistical moments are obtained from the solution of their respective modelled transport equations. Since for combusting flows these equations are usually solved in density-weighted form, the resulting pdf that is constructed from these moments will be the density-weighted pdf. The transport equations for the mean and variance of ξ are very similar in form to equations (2.32) and (2.52) with $\dot{S}_\alpha = 0$. The modelled forms are written below for completeness.

$$\bar{\varrho} \frac{\partial \tilde{\xi}}{\partial t} + \bar{\varrho} \tilde{u}_j \frac{\partial \tilde{\xi}}{\partial x_j} = \frac{\partial}{\partial x_j} \left(\frac{\mu_t}{\sigma_t} \frac{\partial \tilde{\xi}}{\partial x_j} \right) \quad (3.8)$$

$$\bar{\varrho} \frac{\partial \tilde{\xi}''^2}{\partial t} + \bar{\varrho} \tilde{u}_j \frac{\partial \tilde{\xi}''^2}{\partial x_j} = \frac{\partial}{\partial x_j} \left(\frac{\mu_t}{\sigma_t} \frac{\partial \tilde{\xi}''^2}{\partial x_j} \right) + 2 \frac{\mu_t}{\sigma_t} \left(\frac{\partial \tilde{\xi}''^2}{\partial x_j} \right)^2 - \bar{\varrho} C_D \frac{\varepsilon}{k} \tilde{\xi}''^2 \quad (3.9)$$

The turbulent fluxes appearing in the exact equations ($\tilde{u}_j'' \tilde{\xi}''$ and $\tilde{u}_j'' \tilde{\xi}''^2$) have been modelled using a gradient transport hypothesis, and the scalar dissipation rate is approximated using equation (2.63). In order to evaluate μ_t the k - ε model is usually employed; alternatively a full second moment closure can be used in place of equations (3.8) and (3.9), as discussed in section 2.5.2.

Several different forms of the two-parameter pdf have been proposed. For example a form constituting two delta functions located at points ξ^+ and ξ^- has been applied by Khalil et al. [51]:

$$\tilde{P}(\hat{\xi}) = a \delta(\hat{\xi} - \xi^+) + (1 - a) \delta(\hat{\xi} - \xi^-) \quad (3.10)$$

where a , ξ^+ and ξ^- are determined from $\tilde{\xi}$ and $\tilde{\xi}''^2$. On comparison with the measurements of Kent and Bilger [50] in H_2 -air diffusion flames, Jones [41] concluded that such a pdf was unsatisfactory since it led to radial profiles of temperature in which two maxima appeared. Lockwood and Naguib [63] proposed a more realistic clipped Gaussian pdf. The clipping was required since the mixture fraction is a bounded random variable. An unweighted pdf was employed in their formulation, defined by:

$$P(\hat{\xi}) = \begin{cases} \frac{1}{\sigma\sqrt{2\pi}} \int_{-\infty}^0 \exp \left[-\frac{1}{2} \left(\frac{\hat{\xi}-\mu}{\sigma} \right)^2 \right] d\hat{\xi} & \text{if } \hat{\xi} = 0 \\ \frac{1}{\sigma\sqrt{2\pi}} \exp \left[-\frac{1}{2} \left(\frac{\hat{\xi}-\mu}{\sigma} \right)^2 \right] & \text{if } 0 < \hat{\xi} < 1 \\ \frac{1}{\sigma\sqrt{2\pi}} \int_1^{\infty} \exp \left[-\frac{1}{2} \left(\frac{\hat{\xi}-\mu}{\sigma} \right)^2 \right] d\hat{\xi} & \text{if } \hat{\xi} = 1 \end{cases} \quad (3.11)$$

where μ and σ were not obtained explicitly; an iterative procedure employing $\tilde{\xi}$ and $\overline{\xi'^2}$ had to be used. A pdf which does not require arbitrary clipping in order to satisfy the bounds of the mixture fraction is the beta pdf:

$$\tilde{P}(\hat{\xi}) = \frac{\hat{\xi}^{a-1}(1-\hat{\xi})^{b-1}}{\int_0^1 x^{a-1}(1-x)^{b-1}dx} \quad (3.12)$$

$$\text{where } a = \tilde{\xi} \left[\tilde{\xi} \frac{(1-\tilde{\xi})}{\xi'^2} - 1 \right] \quad \text{and} \quad b = (1-\tilde{\xi}) \left[\tilde{\xi} \frac{(1-\tilde{\xi})}{\xi'^2} - 1 \right] \quad (3.13)$$

The integral in the denominator of equation (3.12) is referred to as the β function. This form of pdf has been widely used for engineering calculations of reacting turbulent flow. The implementation of equation (3.6) is particularly straightforward in the context of the β -pdf. The instantaneous relations, $\phi_\alpha(\xi)$, can usually be curve-fitted using a least-squares polynomial (of order ≈ 20) and the relations can then be expressed in the form,

$$\phi_\alpha = \sum_{j=0}^{N_f} \mathcal{X}_{j,\alpha} \xi^j \quad (3.14)$$

where N_f denotes the order of the polynomial fit. By substituting (3.14) into (3.6) and employing the relations pertaining to the β - and Γ -functions:

$$\beta(a, b) = \frac{\Gamma(a)\Gamma(b)}{\Gamma(a+b)} \quad \text{and} \quad \Gamma(a+1) = a\Gamma(a) \quad (3.15)$$

the following analytical expression for the mean of a dependent scalar variable is obtained:

$$\tilde{\phi}_\alpha = \sum_{j=0}^{N_f} \mathcal{X}_{j,\alpha} \frac{\prod_{i=0}^{j-1} (a+i)}{\prod_{i=0}^{j-1} (a+b+i)} \quad (3.16)$$

The numerator and denominator are defined for $j \geq 1$. According to comparisons made by Jones [41] of the clipped Gaussian and the beta pdf, it was observed that the results were quite insensitive to the precise shape of the pdf once the mean and variance were determined.

3.5 Applications of the Flamelet Technique with Flame Stretch

In section 3.3.3 the laminar flamelet technique was briefly discussed and its implementation in turbulent flame calculations using the presumed-shape pdf method (section 3.4) focused attention on flamelet thermochemistry at a fixed strain rate.

Liew [61] employed unstrained ($\chi_{st} = 0$) as well as strained flamelets, though in his work, χ_{max} was used in place of χ_{st} ; χ_{max} was evaluated in a region where the temperature was at a maximum. This modification was introduced in order to account for the translation of the reaction zone to higher values of ξ ($> \xi_{st}$) as the degree of flame stretch increased. For $\phi_\alpha(\xi, \chi_{max})$ a bivariate pdf ($\tilde{P}(\hat{\xi}, \hat{\chi}_{max})$) is required to evaluate the mean scalar properties. The conventional approach for obtaining mean properties is to assume statistical independence of the random variables, ξ and χ_{max} . Without recourse to measurements it is difficult to estimate the accuracy

of this assumption and how strongly it influences the results. It was pointed out in section 3.4 that the statistics of the flow are often not strongly affected by the choice of the pdf, $\tilde{P}(\hat{\xi})$. However, it is not clear that this follows for $\tilde{P}(\hat{\xi}, \hat{\chi}_{max})$, since ξ is a variable influenced primarily by the mixing patterns, whereas χ_{st} (or χ_{max}) is influenced by the finite-rate effects that may be present. Consequently, in flows with appreciable finite-rate effects, the choice of the shape for $\tilde{P}(\hat{\xi}, \hat{\chi}_{max})$ may well be an important consideration. Statistical independence implies that:

$$\tilde{\phi}_\alpha = \int_0^\infty \int_0^1 \phi_\alpha(\hat{\xi}, \hat{\chi}_{max}) \tilde{P}(\hat{\xi}) \tilde{P}(\hat{\chi}_{max}) d\hat{\xi} d\hat{\chi}_{max} \quad (3.17)$$

where $\phi_\alpha(\xi, \chi_{max})$ can be obtained from a library of stretched laminar flamelets. In order to avoid computing the double integral in equation (3.17), Liew [61] modelled the burning process by introducing two phases represented by undisturbed flamelet burning, $\phi_{\alpha,f}(\xi)$ and a post-extinction phase $\phi_{\alpha,I}(\xi)$. Consequently,

$$\tilde{\phi}_{\alpha,f} = \int_0^{\chi_q} \tilde{P}(\hat{\chi}_{max}) d\hat{\chi}_{max} \int_0^1 \phi_{\alpha,f}(\hat{\xi}) \tilde{P}(\hat{\xi}) d\hat{\xi} \quad (3.18)$$

The upper limit of χ_{max} in equation (3.17) has been replaced by (the physically correct value of) χ_q in (3.18). The quantity $\int_0^{\chi_q} \tilde{P}(\hat{\chi}_{max}) d\hat{\chi}_{max} (\equiv \wp_c)$ was regarded as the probability of undisturbed flamelet burning. Although the integral in (3.18) involves the undisturbed flamelet parameter, $\phi_{\alpha,f}(\xi)$, the effect of stretch appears through the term $\wp_c$; as stretch increases, $\wp_c$ decreases. The probability of finding the alternative phase of non-burning is $(1 - \wp_c)$. Thus for the post-extinction phase:

$$\tilde{\phi}_{\alpha,I} = (1 - \wp_c) \int_0^1 \phi_{\alpha,I}(\hat{\xi}) \tilde{P}(\hat{\xi}) d\hat{\xi} \quad (3.19)$$

The relation $\phi_{\alpha,I}(\xi)$ was modelled as a pure mixing process. The quantity of interest, $\tilde{\phi}_\alpha$, was obtained from:

$$\tilde{\phi}_\alpha(\xi, \chi_{max}) = \tilde{\phi}_{\alpha,f}(\xi) + \tilde{\phi}_{\alpha,I}(\xi) \quad (3.20)$$

$\tilde{P}(\hat{\xi})$ was assumed to be a β -pdf. In order to evaluate $\wp_c$, $\tilde{P}(\hat{\chi}_{max})$ had to be specified; by invoking Kolmogorov's third hypothesis (originally proposed for the viscous dissipation rate), $\tilde{P}(\hat{\chi})$ was assumed to be log-normal with the additional assumption that $\chi \approx \chi_{max}$. $\tilde{\chi}$ was modelled using equation (2.63) and the variance of the pdf was given by the third hypothesis directly and its evaluation relied considerably on empiricism with the appearance of geometry dependent constants.

Chapter 4

The Transported Pdf Approach

4.1 Introduction

The closure problem associated with the evaluation of the mean reaction rate can be avoided by assuming that the instantaneous thermochemical state of the flow is a function of one (or more) scalar(s) and averages are obtained when the instantaneous relations are weighted by a presumed-shape pdf. In this section an approach is reviewed which involves the solution of a transport equation for the pdf. Attention will focus on the scalar pdf, $P(\psi; \mathbf{x}, t)$, referred to in section 3.1.

Transport pdf modelling has been investigated for well over two decades. The emergence of this methodology from its rather deep-rooted theoretical background to a potential tool for engineering calculations is mainly due to the development of Monte Carlo algorithms for solving the pdf equation. From the statistical point of view, the specification of $P(\psi; \mathbf{x}, t)$ makes available all single point, single time moments of the scalar field. In contrast, moment closures yield only those moments for which modelled transport equations are solved. Where pdf methods appear to be particularly advantageous is in the description of turbulent reacting flows where the thermochemistry is a non-linear function of several independent scalars. In the conserved scalar approach (section 3.2) the pdf of a single, strictly conserved scalar is employed; however, in order to reproduce finite-rate effects (e.g. emissions and extinction) accurately, more independent scalars need to be incorporated in the characterization of the pdf. The extension of the presumed-shape pdf method to multi-scalar problems is difficult because:

- the number of modelled transport equations required to construct the pdf rapidly rises with the number of independent scalars and,
- presuming a shape for a multi-dimensional pdf which satisfies the appropriate bounds becomes increasingly difficult and uncertain as the number of independent scalars rises.

Such difficulties are averted through the use of pdf transport methods.

4.2 Derivation of the Scalar Pdf Equation

The transport equation for $P(\psi)$ (the dependence on $\mathbf{x}$ and t is omitted for conciseness) is an exact, unclosed equation and its derivation relies on the use of the conservation equations. Consequently the equation has a physical foundation. The most widely-used method of derivation is based on

the concept of a fine-grained density function $\check{p}(\underline{\psi}, \underline{\phi}; \mathbf{x}, t)$, Lundgren [65], and is defined such that $\check{p}(\underline{\psi}, \underline{\phi})d\underline{\psi}$ is the probability that at position $\mathbf{x}$ and time t , $\psi_\alpha \leq \phi_\alpha(\mathbf{x}, t) \leq \psi_\alpha + d\psi_\alpha$ for all $\alpha = 1, \dots, N+1$. Note that ψ_α denotes an independent variable (like $\mathbf{x}$ and t) and represents the sample space of the dependent random variable $\phi_\alpha(\mathbf{x}, t)$; $\underline{\psi}$ denotes the set of ψ_α for $\alpha = 1, \dots, N+1$. Clearly $\check{p}(\underline{\psi}, \underline{\phi})$ is a multi-dimensional quantity and it represents the pdf for a single realization of a turbulent flow, so that if $\psi_\alpha \leq \phi_\alpha(\mathbf{x}, t) \leq \psi_\alpha + d\psi_\alpha$ is not satisfied for all α , then $\check{p}(\underline{\psi}, \underline{\phi}) = 0$, otherwise $\check{p}(\underline{\psi}, \underline{\phi}) = 1$. Consequently the fine-grained density can be expressed in the following manner:

$$\check{p}(\underline{\psi}, \underline{\phi}) \equiv \prod_{\alpha=1}^{N+1} \delta\{\phi_\alpha(\mathbf{x}, t) - \psi_\alpha\} \quad (4.1)$$

In contrast, the conventional pdf $P(\underline{\psi})$ is representative of an ensemble of realizations, such that its integral over a finite region of its sample space represents the likelihood of occurrence for an event. It can be shown [81] that:

$$P(\underline{\psi}) = \langle \check{p}(\underline{\psi}, \underline{\phi}) \rangle \quad (4.2)$$

A detailed derivation of the transport equation for $P(\underline{\psi})$ is presented in appendix A; the works of O'Brien [72] and Kollmann [54] also discuss the derivation. Pope [81] describes a method for obtaining the scalar pdf equation without invoking the concept of the fine-grained density. Kollmann [52, 53] derives the pdf equation in terms of the characteristic function, which is the Fourier transform of the pdf. Whatever the method of derivation the transport equation for the density-weighted scalar pdf is given by:

$$\begin{aligned} \underbrace{\bar{\rho} \frac{\partial \tilde{P}(\underline{\psi})}{\partial t}}_{\text{I}} + \underbrace{\bar{\rho} \tilde{u}_k \frac{\partial \tilde{P}(\underline{\psi})}{\partial x_k}}_{\text{II}} + \underbrace{\bar{\rho} \sum_{\alpha=1}^{N+1} \frac{\partial}{\partial \psi_\alpha} \{ \dot{S}_\alpha^*(\underline{\psi}) \tilde{P}(\underline{\psi}) \}}_{\text{III}} = & - \underbrace{\frac{\partial}{\partial x_k} \{ \bar{\rho} \langle u_k'' | \underline{\phi} = \underline{\psi} \rangle \tilde{P}(\underline{\psi}) \}}_{\text{IV}} \\ & + \underbrace{\sum_{\alpha=1}^{N+1} \frac{\partial}{\partial \psi_\alpha} \left\{ \left\langle \frac{\partial J_{i,\alpha}}{\partial x_i} \right| \underline{\phi} = \underline{\psi} \right\rangle P(\underline{\psi}) \right\}}_{\text{V}} \end{aligned} \quad (4.3)$$

where $\dot{S}_\alpha(\underline{\phi}) \equiv \rho(\underline{\phi}) \dot{S}_\alpha^*(\underline{\phi})$. The terms in equation (4.3) have the following significance:

- Term I: Rate of change in physical space
- Term II: Mean convection in physical space
- Term III: Chemical source production in compositional space
- Term IV: Turbulent transport in physical space
- Term V: 'Molecular mixing' in compositional space

When $J_{i,\alpha}$ is expressed in terms of Fick's and Fourier's laws, the 'molecular mixing' term can be written as:

$$- \sum_{\alpha=1}^{N+1} \sum_{\beta=1}^{N+1} \frac{\partial^2}{\partial \psi_\alpha \partial \psi_\beta} \left\{ \left\langle \frac{\mu}{Sc} \frac{\partial \phi_\alpha}{\partial x_k} \frac{\partial \phi_\beta}{\partial x_k} \right| \underline{\phi} = \underline{\psi} \right\rangle P(\underline{\psi}) \right\} \quad (4.4)$$

Due to the unity Lewis number assumption ($Sc = Pr$), the form of equation (4.4) is applicable to both the species concentrations and the enthalpy. With the assumption of constant μ and ρ the molecular mixing term can be expressed in terms of a two-point pdf [54]:

$$-\frac{\mu}{Sc} \sum_{\beta=1}^{N+1} \frac{\partial}{\partial \psi_{\beta}} \lim_{\mathbf{x}' \rightarrow \mathbf{x}} \frac{\partial^2}{\partial x'_k \partial x'_k} \left\{ \int \psi'_{\beta} P(\underline{\psi}, \underline{\psi}'; \mathbf{x}, \mathbf{x}', t) d\underline{\psi}' \right\} \quad (4.5)$$

where the primes denote terms based on conditions at $\mathbf{x}'$.

4.3 Terms Appearing in the Pdf Equation

Inspection of equation (4.3) reveals that the chemical source production term appears in exact and closed form. In other words it is known in terms of $\underline{\psi}$ and $\tilde{P}(\underline{\psi})$. This feature of equation (4.3) is one of the main reasons for adopting the transported pdf approach in turbulent combustion modelling. The implication is that realistic chemistry involving finite-rate kinetics can be incorporated explicitly in turbulent flow calculations without an assumed flame structure to characterize the thermochemistry. In comparison to other forms of thermochemical closure this is clearly a novel, advantageous and unique feature. However, the molecular mixing term appears in an unclosed form due to the conditional expectation. To evaluate this term the joint pdf of the scalars and their gradient at the single point level is required; alternatively the two point pdf of the scalars would have to be specified. Needless to say that transport equations for such higher-order pdf's contain further unknown terms and increase the dimensionality and/or computational cost of the problem dramatically.

The molecular mixing term involves diffusion coefficients and composition gradients which stem directly from the molecular diffusion term in equation (2.16) and an order of magnitude analysis demonstrates that it cannot be neglected, even at high Re . Since molecular diffusion is important at the small scales of turbulent motion, an accurate representation of these scales is clearly essential for the determination of the pdf and hence of the averaged scalar field. In hydrocarbon combustion, this is particularly true for species such H_2 and H which are formed and consumed by reaction (in the fine scales of turbulent motion) and which possess much larger diffusivities than other scalars. As a consequence, it can be argued that the closure problem which manifests itself as a mean reaction rate in moment closures is simply re-expressed in terms of the molecular mixing term in pdf closures. In fact the appearance of the chemical source term in closed form in the pdf formalism is merely a consequence of the mathematical manipulations employed to derive such an equation. As pointed out by Kollmann [54], in pdf methods nonlinear terms are transformed into linear terms with variable coefficients. This is achieved by converting the associated dependent variables (which make the nonlinear terms unclosed upon averaging) into independent variables of the pdf; one outcome of this is that the number of independent variables is substantially increased. The results of such a manipulation do not overcome the basic physical problems inherent in any statistical approach to turbulence, whether it be Reynolds/Favre averaging, spatial averaging or transported pdf modelling.

The pdf $\tilde{P}(\underline{\psi})$ is only a function of the scalar field and includes no velocity information; consequently the turbulent diffusion of the pdf (term IV) appears in unclosed form. In the joint velocity-composition pdf equation [81] transport appears in closed form, but at the expense of introducing further unknowns and extra independent variables to the level of closure. In the scalar

pdf formalism $\tilde{u}_k$ is unknown. This quantity is conventionally evaluated through the use of a turbulence model (section 2.5).

4.4 General Solution Features of Pdf Transport Equations

At this stage it may appear somewhat premature to discuss the solution features of pdf transport equations, particularly since the modelling of the unclosed terms has not yet been considered. However, based on this formalism the method of numerical solution and the modelling are strongly interconnected. For problems involving a single scalar dimension and in simple geometries, solutions of pdf equations using conventional finite-difference techniques have been obtained (Janicka et al. [39, 40]). However, in order to exploit the full potential of pdf equations it is necessary to solve for a multi-dimensional pdf involving several scalars. Pope [79] estimated that with a finite-difference procedure the number of computer operations at each node (in compositional space) and at each time step was of the order of $\exp(6N^*)$, where N^* is the number of scalar variables. In the same paper he described a solution algorithm for performing a Monte Carlo (particle-method) simulation of the scalar pdf equation¹. In this approach the computational cost rises only linearly with N^* and the pdf is simulated via an ensemble of stochastic particles. The idea that a continuous pdf, $\tilde{P}(\psi)$, can be represented in a discrete manner is commonly employed; for example equations (2.24) and (2.25) demonstrate how an average can be constructed from an ensemble of samples. If a histogram was constructed from $A^{(i)}(\mathbf{x}, t)$, this would constitute an approximate discrete representation of the pdf. In the limit of an infinite number of realizations the histogram tends to the pdf - this result is formalized by equation (4.2).

With regards to the modelling of the mixing term in equation (4.3), the above paragraph implies that given a suitable approximation (written in a continuous pdf form), an equivalent stochastic (particle) representation needs to be constructed. Thus in the section dealing with the mixing models, both interpretations will be given for the case of a single scalar. For certain mixing models the extension to multi-scalar problems is simple; in other models it is still not clear how this can be achieved in a rigorous and/or tractable manner. The existence of a link between the particle method and the continuous pdf approach is essential if the advantages of the former method are to be exploited. Such a link and the necessary criteria for its use have been investigated by Pope [77, 79, 81]; the fundamental basis for 'equivalence' of these two methodologies is that the statistical moments (mean, variance, etc. . .) arising from the particle-method approach should evolve in an identical manner to its deterministic counterpart in the limit of an infinite number of particles.

¹Handscorn and Hammersley [33] define Monte Carlo methods as "*that branch of experimental mathematics which is concerned with experiments on random numbers*". Given the generality of the definition, it is clear that Monte Carlo method is a generic name for an approach to solving problems. These methods can range from the simulation of neutron diffusion in fissile material, to the estimation of π based on an experiment where a needle is thrown in a random manner onto a board ruled with parallel lines. For simple problems Monte Carlo methods are inefficient compared with standard numerical techniques, but for multi-dimensional problems they are practicable relative to the other methods which become computationally prohibitive.

4.5 Basic Modelling Requirements of the Molecular Mixing Term

Proposals for closing the molecular mixing term are invariably tested in simple flows e.g. inert, homogeneous and isotropic turbulence. Under these circumstances the scalar pdf is no longer a function of position and simply evolves in time, with the result that the scalar variance decays to zero. It is argued that if pdf methods cannot reproduce the statistics of the decay process accurately in this simple flow, then the claim that such a formalism provides a more complete statistical representation of fluid flows would be unjustified. In the situation described above, the most basic requirements of a mixing model are that it leaves the mean of the scalar unchanged (i.e. $\Delta\langle\phi(\mathbf{x}, t)\rangle \equiv \langle\phi(\mathbf{x}, t + \delta t) - \phi(\mathbf{x}, t)\rangle = 0$) and predicts a variance decay rate in accordance with experimental observations:

$$\frac{d}{dt}\langle\phi'^2\rangle = -C_D \frac{\varepsilon}{k}\langle\phi'^2\rangle \quad (4.6)$$

where $C_D \approx 2.0$. All models are designed to satisfy the above (minimum) requirements. In discussing the statistical behaviour of the various models, reference will be made to the following parameters: skewness, flatness and superskewness. These are the normalized moments of the pdf, defined as follows:

$$\Omega_n = \frac{\int_{\psi} (\psi - \langle\phi\rangle)^n P(\psi) d\psi}{\left\{ \int_{\psi} (\psi - \langle\phi\rangle)^2 P(\psi) d\psi \right\}^{\frac{n}{2}}} \quad \left\{ \begin{array}{l} n = 3 \Rightarrow \text{skewness} \\ n = 4 \Rightarrow \text{flatness} \\ n = 6 \Rightarrow \text{superskewness} \end{array} \right. \quad (4.7)$$

The most revealing test of a mixing model lies in its ability to predict correctly the evolution of the scalar pdf. The test case utilised by most workers is based on initial conditions corresponding to segregated fluid parcels (in composition space), idealized as a double δ -function distribution. Here it is to be expected that the process of mixing/homogenization will result in the initial pdf to relax to a smooth, bell-shaped distribution and in the limit of $t \rightarrow \infty$, converging to a δ function at the mean composition. It is conventional to assume that the bell-shaped distribution is Gaussian, although there is no formal proof for this. Tavoularis and Corrsin [91] investigated (experimentally) the evolution of a scalar (temperature) in a turbulent flow with uniform mean velocity and temperature gradients, and nearly homogeneous fluctuating velocity and temperature fields. The overheat of the fluid by heating rods was considered small enough to have negligible effect on the turbulence, and hence the temperature can be viewed as a passive scalar. It was found that the typical values of the skewness and flatness factors of the temperature measured on the centre-line of the tunnel, far downstream, were indistinguishable from those for a normally distributed random variable (which are zero and three, respectively); it was found that a Gaussian pdf with the same mean and variance fitted the measured pdf's nearly perfectly. The velocity-scalar joint pdf also closely resembled a jointly-normal distribution.

Eswaran and Pope [25] performed a DNS of decaying passive scalar fields in inert, homogeneous and isotropic turbulence. A statistically stationary velocity field was maintained by 'forcing' the simulations i.e. energy was added to the velocity field at low wave numbers, allowing the simulations to reach a quasi-equilibrium state where the rate at which energy was dissipated at the small scales was equal to the rate at which energy was added at the large scales. The results

demonstrated that as time progressed, $P(\psi; t)$ evolved from a double δ function distribution to an inverted parabola and subsequently to a bell-shaped distribution. The evolution of the pdf shapes appeared to be independent of the initial conditions, such as the ratio of the mechanical and scalar turbulence length scales (which had a significant effect on the scalar variance decay rate). It was argued that this observation should considerably simplify the task of constructing a realistic mixing model and it was found that after about eight eddy turnover times (a relatively long period of time), the skewness, flatness and superskewness of ϕ tended very closely to the Gaussian values of zero, three and fifteen respectively. The tendency to Gaussianity was accompanied by an ever increasing lack of correlation between χ and $\phi(\mathbf{x}, t)$. This is an important result since the molecular mixing term in equation (4.4) is effectively χ conditioned on $\phi(\mathbf{x}, t)$. As a modelling recommendation it was claimed that the assumption of χ being independent of $\phi(\mathbf{x}, t)$, although valid at long times, is not valid in the short-term range of mixing and this raises questions concerning the validity of the statistical independence assumption invoked in section 3.5.

Despite the above comments concerning the evolution of a decaying scalar field to Gaussianity, it must be borne in mind that the scalars considered here are bounded quantities and so their pdf's are bounded too; therefore such a scalar pdf cannot be exactly Gaussian. The implication is that the pdf approaches a Gaussian distribution in the limit of zero variance where most of the interacting fluid 'parcels' are very close to the mean composition. However the distribution near the bounds of the scalar are unlikely to resemble Gaussian behaviour, but this is immaterial given the degree of homogenization at that stage of the mixing process. A number of proposals have been made for closing the molecular mixing term and these are discussed below.

4.6 The Linear Mean Square Estimation (LMSE) Closure

Equation (4.5) and the concept of conditional probability, $P(\psi, \psi') = P(\psi' | \phi(\mathbf{x}, t) = \psi)P(\psi)$, can be used to write the pdf equation in homogeneous, isotropic turbulence as:

$$\begin{aligned} \frac{\partial P(\psi; t)}{\partial t} &= -\frac{\nu}{Sc} \sum_{\beta=1}^{N+1} \frac{\partial}{\partial \psi_{\beta}} \lim_{\mathbf{x}' \rightarrow \mathbf{x}} \frac{\partial^2}{\partial x'_k \partial x'_k} \left\{ \int \psi'_{\beta} P(\psi'_{\beta} | \phi = \psi) d\psi'_{\beta} P(\psi) \right\} \\ &= -\frac{\nu}{Sc} \sum_{\beta=1}^{N+1} \frac{\partial}{\partial \psi_{\beta}} \lim_{\mathbf{x}' \rightarrow \mathbf{x}} \frac{\partial^2}{\partial x'_k \partial x'_k} \left[E(\phi'_{\beta} | \phi = \psi) P(\psi) \right] \end{aligned} \quad (4.8)$$

Dopazo [19, 21] and Dopazo and O'Brien [22] consider the case of the single scalar, where $E(\phi' | \phi = \psi)$ is the expectation of the random variable $\phi(\mathbf{x}', t)$ at a point $\mathbf{x}'$, conditioned on the event that $\phi(\mathbf{x}, t) = \psi$ at a point $\mathbf{x}$. If it is assumed that $P(\psi' | \phi = \psi)$ is conditionally Gaussian, the theory of stochastic processes implies that:

$$E(\phi' | \phi = \psi) = \langle \phi \rangle + r(\mathbf{x}, \mathbf{x}', t) (\psi - \langle \phi \rangle) \quad (4.9)$$

where $\langle \phi \rangle$ is the expectation (mean) of the random variable $\phi(\mathbf{x}, t)$ and $r(\mathbf{x}, \mathbf{x}', t)$ is the correlation coefficient of $\phi(\mathbf{x}, t)$ and $\phi(\mathbf{x}', t)$,

$$r(\mathbf{x}, \mathbf{x}', t) = \frac{\langle [\phi(\mathbf{x}, t) - \langle \phi(\mathbf{x}, t) \rangle] [\phi(\mathbf{x}', t) - \langle \phi(\mathbf{x}', t) \rangle] \rangle}{\sigma_{\phi}(\mathbf{x}, t) \sigma_{\phi}(\mathbf{x}', t)} \quad (4.10)$$

where σ_{ϕ} denotes the square root of the variance of ϕ . Dopazo and O'Brien [23] point out that the above assumption does not imply that $\phi(\mathbf{x}, t)$ and $\phi(\mathbf{x}', t)$ are normally distributed; it is claimed

that it constrains the expected value of $\phi(\mathbf{x}', t)$, given $\phi(\mathbf{x}, t)$, to a certain prescribed behaviour. More recently O'Brien [72] reformulated the problem in terms of linear mean square estimation, which involves less restrictive assumptions but gives rise to the same result. The requirement of homogeneity and isotropy together with equations (4.9) and (4.10) leads to the following result:

$$\frac{\partial P(\psi; t)}{\partial t} = A \frac{\partial}{\partial \psi} [(\psi - \langle \phi \rangle) P(\psi; t)] \quad (4.11)$$

where $A = \frac{6\nu}{Sc\lambda_\phi^2}$, $\lambda_\phi^2 \equiv -2(\partial^2 r / \partial \ell^2)_{\ell=0}^{-1}$ and $\ell = |\mathbf{x}' - \mathbf{x}|$. Multiplying equation (4.11) by $(\psi - \langle \phi \rangle)^2$ and integrating over ψ space leads to:

$$\frac{d\langle \phi'^2 \rangle}{dt} = -2A\langle \phi'^2 \rangle \quad (4.12)$$

Comparing the result in equation (4.12) with that of equation (2.52) (in the case of constant density, homogeneous, isotropic and inert turbulence) implies the following identity:

$$\bar{\chi}_\phi = \frac{\nu}{Sc} \frac{\partial \phi'}{\partial x_i} \frac{\partial \phi'}{\partial x_i} = A\langle \phi'^2 \rangle \quad (4.13)$$

Referring to equation (4.11) as the LMSE model, the particle method simulation corresponding to this closure becomes:

$$\frac{d\phi^{(p)}}{dt} = -\frac{C_D}{2} \frac{\varepsilon}{k} (\phi^{(p)} - \langle \phi \rangle) \quad (4.14)$$

where A has been replaced with $C_D \frac{\varepsilon}{k}$, and by selecting $C_D \approx 2$ a variance decay rate in conformity with measurements can be predicted as suggested by equation (4.6). LMSE constitutes a linear, deterministic simulation of the mixing phenomenon and the model is continuous in time. Analytical solutions for equation (4.11) are discussed in [23, 72, 81]; the results of such analysis show that if the initial condition is a normal distribution, the model preserves the Gaussianity which is considered as a satisfactory result. However, the drawback is that the initial functional form of the pdf is never relaxed and consequently, in the case of an initial double δ function specification for the pdf, the subsequent time evolution remains as two δ functions which approach each other in composition space. Pope [81] points out that such behaviour is to be expected since the model contains no information about the shape of the distribution - only the mean appears in equation (4.14). Despite this drawback, Dopazo [21] argues that the modelling is applicable provided the turbulence is "given a chance" to smear out the initial discontinuous state. In other words it is applicable if the pdf is continuous. Predictions based on LMSE were compared against measurements in [21] for grid-generated homogeneous turbulence with an imposed asymmetric mean temperature profile. In the far downstream region good agreement was found for the conditional expected value of temperature at a point, when the temperature at a neighbouring point was prescribed.

4.7 Coalescence-Dispersion Mixing Models

4.7.1 Curl's Mixing Model

The work of Curl [17] was concerned with the analysis of the mixing of clouds of droplets in a two-phase chemical reactor. It was assumed that all the droplets in the system were of the same

size and that coalescence took place between two droplets having a solute concentration of c_1 and c_2 . Redispersion occurred immediately to produce two equal droplets of concentration $\frac{1}{2}(c_1 + c_2)$. The total concentration range was divided into L intervals of concentration width δc and $P(l)\delta c$ was defined as the fraction of the droplets with concentrations within the l^{th} ($1 \leq l \leq L$) interval. The basic balance equation constructed for $P(c)$ stated that the rate of change of the number of droplets in a particular interval was influenced by:

- the rate of creation of new droplets in the interval $c, c + \delta c$ due to collisions of droplets between symmetrically and asymmetrically located concentration intervals
- the rate of loss of droplets by collision of droplets from $c, c + \delta c$ with those from all other concentration intervals

and this led to:

$$\frac{\partial P(c)}{\partial t} = 4Q \left\{ \int_0^c P(c + \alpha)P(c - \alpha) d\alpha - P(c) \right\} \quad (4.15)$$

Curl defined Q as twice the collision rate divided by the total number of droplets in the system and solved equation (4.15) for the case of inert and reacting flows using conventional finite-difference techniques. Subsequently Spielman and Levenspiel [89] proposed a particle (Monte Carlo) method for the solution of such an equation and used the methodology to investigate the flow characteristics of an ideal stirred tank reactor involving a dispersed phase system. Curl's mixing closure has also been used in the context of continuous (single-phase) systems, to describe the turbulent mixing of gases. The work of Flagan and Appleton [28] and the references cited therein may be consulted as examples of such applications.

Dopazo [21] pointed out that since Curl's model was originally intended for interactions of droplets, the direct extrapolation of this model to the mixing of single-phase gaseous eddies was questionable. The implication of such an extrapolation would be that two fluid 'parcels' at different temperatures, say, would mix instantaneously to form fluid parcels with a temperature equal to the average of the two original temperatures. This feature of the model is clearly unrealistic and has also been highlighted by Pope [80] on performing the stochastic simulations. An initial condition corresponding to a double δ -function was specified in the following manner: of the $\mathcal{N}$ particles in the system, $\mathcal{N}/2$ were assigned $\phi^{(p)} = 1$ and the remainder as $\phi^{(p)} = -1$ (where $1 \leq p \leq \mathcal{N}$). In any particular time step, pairs of particles were selected, and the number of pairs chosen was the nearest integer to:

$$\mathcal{N}_s = \mathcal{B} \delta t \frac{\varepsilon}{k} \mathcal{N} \quad (4.16)$$

This expression was chosen to ensure a variance decay rate consistent with equation (4.6) and for Curl's model $\mathcal{B} \approx 2$. The particles were then allowed to interact along the straight line connecting them in composition space, such that their new concentrations 'jump' from their initial values to their joint mean value. The values of particles not selected remain unaltered. The results demonstrated that after the first time step ϕ took the values $-1, 0$ and 1 ; after the second step they were $-1, -\frac{1}{2}, 0, \frac{1}{2}$ and 1 ; after the j^{th} step they were all integer multiples of 2^{1-j} . Therefore a continuous distribution was never achieved since certain values of the scalar sample space (those not corresponding to multiples of 2^{1-j}) could not be reached. In spite of this, after a sufficient period of time, a bell-shaped (but non-Gaussian) distribution was obtained. Therefore, in contrast to the LMSE closure, the pdf does relax from its initial shape.

4.7.2 Modified Curl

Dopazo [21] and Janicka et al. [40] independently proposed modifications to Curl's model in order to avoid some of its shortcomings. Although the starting point for motivating their modelling was different, the end results were very similar indeed largely because their approach followed the ideas behind the derivation of Curl's formulation. Essentially the difference between Curl's model and the modified forms lies in the treatment of the dispersion of the material point properties. In discussing these differences it is useful to introduce the concept of the transition pdf. The transition pdf is defined such that $T_r(\varphi, \Psi|\psi) d\psi$ is the probability that the interaction of a material point $\phi = \varphi$ with a point $\phi = \Psi$ produces $\psi \leq \phi \leq \psi + d\psi$ and $(\varphi + \Psi - \psi) \leq \phi \leq (\varphi + \Psi - \psi) + d\psi$. The transition pdf has to satisfy the following properties:

- In coalescence-dispersion models the mixing process is effectively represented by the pairwise interaction of material points. These material points are only allowed to mix along the line joining them in composition space and this is consistent with the notion of physical mixing. Therefore, if ψ is outside the interval (φ, Ψ) , it follows that $T_r(\varphi, \Psi|\psi) d\psi = 0$. This implies that the interval (φ, Ψ) denotes a local convex domain for ψ .
- $T_r(\varphi, \Psi|\psi) = T_r(\varphi, \Psi|\varphi + \Psi - \psi)$. This effectively states that after pairwise interaction, the new scalar properties ψ and $\varphi + \Psi - \psi$ are formed with the same likelihood. This condition, together with the previous requirement ensure that the mean of the dependent scalar, ϕ , remains unchanged by the molecular mixing process.
- $T_r(\varphi, \Psi|\psi)$ is necessarily a pdf with respect to ψ . Therefore the normalization condition follows; the integration over the domain of ψ yields: $\int_{\psi} T_r(\varphi, \Psi|\psi) d\psi = 1$

In terms of the transition pdf, coalescence-dispersion models can be expressed in the following general form (using a density-weighted pdf) [44]:

$$\bar{\rho} \frac{\partial \tilde{P}(\psi)}{\partial t} \approx \bar{\rho} C_d \frac{\varepsilon}{k} \left\{ \int_{\varphi} d\varphi \int_{\Psi} d\Psi \tilde{P}(\varphi) \tilde{P}(\Psi) T_r(\varphi, \Psi|\psi) - \tilde{P}(\psi) \right\} \quad (4.17)$$

where C_d is a constant which can be evaluated by multiplying equation (4.17) by $(\psi - \langle \phi \rangle)^2$, integrating over ψ space, and then comparing the result with equation (4.6). In relation to equation (4.16), $C_d = 2B$.

In terms of Curl's original specification, where material points/particles coalesce and disperse with properties involving their mean value, the transition pdf takes the following form:

$$T_r(\varphi, \Psi|\psi) = \delta \left[\psi - \frac{1}{2}(\varphi + \Psi) \right] \quad (4.18)$$

In modified forms of Curl's model the material points that interact are not constrained to mix to their common mean values. In fact the degree of mixing can vary between no mixing and complete mixing (up to the mean composition) and it can be assumed that all composition states can be attained with equal likelihood between these limits. Thus, the transition pdf will then be a uniform distribution [40] over the range of the local convex domain (φ, Ψ) :

$$T_r(\varphi, \Psi|\psi) = \begin{cases} \frac{1}{|\varphi - \Psi|} & \psi \in [\varphi, \Psi] \\ 0 & \psi \notin [\varphi, \Psi] \end{cases} \quad (4.19)$$

In this case discontinuous pdf's cannot arise (in the sense that Curl's original model produces) since all possible composition states can be reached. However, the mixing is still represented as a jump process and such closures are essentially discontinuous representations of the mixing phenomenon.

The proposal of Dopazo [21] and Janicka et al. [40] has been examined further by Pope [80]. In the particle method analogy of the generalised coalescence-dispersion model, a random selection of a pair of particles $\phi^{*(p)}(t)$ and $\phi^{*(q)}(t)$ is followed by 'mixing' such that the new concentrations are given by:

$$\begin{aligned}\phi^{*(p)}(t + \delta t) &= \phi^{*(p)}(t) + \frac{1}{2}x \left(\phi^{*(q)}(t) - \phi^{*(p)}(t) \right) \\ \phi^{*(q)}(t + \delta t) &= \phi^{*(q)}(t) + \frac{1}{2}x \left(\phi^{*(p)}(t) - \phi^{*(q)}(t) \right)\end{aligned}\quad (4.20)$$

x controls the extent of mixing ($x \in [0, 1]$), and the result is unaffected by the time step; with $x = 0$ no mixing occurs, whilst Curl's model is recovered with $x = 1$. x has to be a random variable with a continuous distribution, $\mathcal{A}(x)$, otherwise the problems associated with Curl's original formulation (given by $\mathcal{A}(x) = \delta(x - 1)$) arise. The simplest choice is to make $\mathcal{A}(x)$ a uniform distribution, i.e. $\mathcal{A}(x) = 1$. With regards to equation (4.16), $\mathcal{B}$ changes according to the choice of $\mathcal{A}(x)$. Pope [80] shows that:

$$\mathcal{B} = \left(a_1 - \frac{1}{2}a_2 \right)^{-1} \quad \text{where} \quad a_n = \int_0^1 x^n \mathcal{A}(x) dx \quad (4.21)$$

According to equation (4.21), if $\mathcal{A}(x) = 1$ then $\mathcal{B} \approx 3$. Calculations of scalar mixing in inert, homogeneous, isotropic turbulence fields demonstrate that starting from a double δ function distribution, continuous pdf's can be obtained with the generalised coalescence-dispersion model. However, the flatness (equation (4.7)) and higher *even* moments of the resulting pdf rise exponentially with time, whilst for a Gaussian pdf, these values are constant. Curl [17] demonstrated this analytically for his particular specification and Kosaly [55] has generalized the result regardless of the form of $\mathcal{A}(x)$.

Kosaly [55] argued that the 'proper' choice of $\mathcal{A}(x)$ was a potential method of incorporating more physics into coalescence-dispersion mixing models. In the initial stages of mixing in homogeneous, isotropic turbulence the pdf resembles an inverted parabola, and so a rational choice for $\mathcal{A}(x)$ should peak at $x = 0$ and monotonically decrease for increasing values of x . This ensures that the degree of mixing is controlled so that concentrations at the mean value do not appear in the early stages. Most proposals for $\mathcal{A}(x)$ do not satisfy this requirement in the early stages of mixing (e.g. $\mathcal{A}(x) = 1$) and furthermore, the question of how fast $\mathcal{A}(x)$ falls with x is important; Kosaly demonstrates analytically that with the condition $\frac{a_n}{a_1} \ll 1, n > 1$, coalescence-dispersion modelling yields an identical result to LMSE plus terms $\mathcal{O}(\mathcal{B}a_2)$. It should not be inferred from this that LMSE is simply the limiting behaviour of coalescence-dispersion models. Both of these models are fundamentally different; LMSE is a linear, deterministic and continuous representation of mixing, whilst coalescence-dispersion models are nonlinear, jump processes whose derivations are somewhat phenomenological. The idea of improving the modelling of the coalescence-dispersion closure through more elaborate specifications of $\mathcal{A}(x)$ has not received widespread attention since it is not regarded as the primary factor affecting the performance of the model. Kosaly and Givi [56] investigated the sensitivity of their predictions

to the choice of $\mathcal{A}(x)$ and considered LMSE and Curl's model to represent opposite extremes in mixing behaviour. The former was, in fact, represented through the following specification: $\mathcal{A}(x) = \delta(x - \epsilon)$, $\epsilon \rightarrow +0$ though in practice, $\epsilon \approx 0.1$ was employed. Calculations of an axisymmetric, non-swirling, turbulent jet discharging into stagnant surrounds demonstrated that the pdf's arising out of the use of $\mathcal{A}(x) = \delta(x - \epsilon)$, $\mathcal{A}(x) = 1$ and $\mathcal{A}(x) = \delta(x - 1)$ were quite similar at about 10 diameters downstream of the nozzle.

Due to the shortcomings of the coalescence-dispersion models with regard to the predictions of the normalized moments, much effort has been devoted towards their 'improvement' so that finite values of flatness and superskewness can be obtained.

4.7.3 Age-Biased Coalescence-Dispersion Model

Pope [80] formulated an age-biased model such that continuous pdf's with finite flatness and superskewness could be obtained. In this model the selection of elements for mixing was biased towards the age of the element, whereas in the conventional coalescence-dispersion modelling, the likelihood of sampling (without replacement) a particular element is equal for all particles. The age of an element was defined as the normalized time since it mixed with another element. Denoting $t^{(n)}$ as the last time that the element n was selected for mixing, the age of the n^{th} element was defined as: $\tau^{(n)} = \frac{\epsilon}{k}(t - t^{(n)})$ where t is the current time. An age distribution $\mathcal{T}(s)$ was assigned as the pdf of τ , where s is the sample space of τ . The age distribution can be changed by biasing the sampling according to the age of the elements. This was achieved through the sample bias, $\mathcal{Z}(s)$, normalized so that $\int_0^\infty \mathcal{T}(s)\mathcal{Z}(s)ds = 1$. $\mathcal{T}(s)$ was obtained from the following evolution equation:

$$\mathcal{T}(s + \frac{\epsilon}{k}\delta t; t + \delta t) \approx \mathcal{T}(s; t) + 2\mathcal{B}\frac{\epsilon}{k}\delta t [\delta(s) - \mathcal{T}(s)\mathcal{Z}(s)] \quad (4.22)$$

The argument $s + \frac{\epsilon}{k}\delta t$ in equation (4.22) reflects the fact that an element (not selected for mixing) ages an amount $\frac{\epsilon}{k}\delta t$ in a time interval δt . The fraction of elements selected is $2\mathcal{B}\frac{\epsilon}{k}\delta t$ and the effect is to remove an amount $\mathcal{T}(s)\mathcal{Z}(s)$ from $\mathcal{T}(s)$ and to add it to $\mathcal{T}(0)$; in other words the age of the mixed elements is reset to zero. $\mathcal{Z}(s)$ is interpreted as the relative probability of sampling the element n , of age $\tau = s$; for modified Curl $\mathcal{Z}(s) = 1$, so that $\mathcal{T}(s) = 2\mathcal{B}\exp(-2\mathcal{B}s)$. The proposed mixing model was defined by two pdf's, $\mathcal{A}(x)$ and $\mathcal{Z}(s)$. A stochastic model algorithm, simulating the behaviour of the resulting pdf equation was proposed and solved for the binary mixing problem. Six different $\mathcal{A}(x)$ specifications were employed with six different $\mathcal{T}(s)$. The results demonstrated that the 'best' combination of $\mathcal{A}(x)$ and $\mathcal{T}(s)$, in terms of yielding normalized moments closest to the Gaussian value, involved (paradoxically) $\mathcal{A}(x) = \delta(x - 1)$ (i.e. Curl's original form) with an age distribution $\mathcal{T}(s) = 2\mathcal{B}$. It was found that a quartic function yielded the best compromise between good overall behaviour (in terms of pdf evolution) and acceptable values of the normalized moments. It is clear why this approach should yield finite higher-order moments; in homogeneous turbulence test cases using conventional coalescence-dispersion models, the main factor which contributes to the monotonically rising higher order moments is the presence of scalar 'parcels' or particles near the bounds of the composition space which have not yet been selected for mixing; with age-biased modelling and LMSE, particles near the bounds are constrained to move towards the mean.

Chen and Kollmann [14] investigated the application of Pope's age-biased model and chose a power law variation for the biasing function. They calculated joint pdf's of mixture fraction and

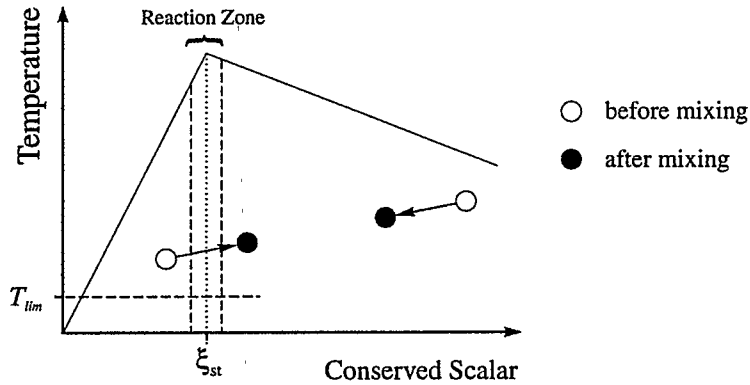


Figure 4.1: Schematic of the mixing process based on coalescence-dispersion closures showing how a particle mixes and thereby crosses the reaction zone without reacting. T_{lim} is the temperature below which the reaction rates are negligibly low.

temperature for reacting and non-reacting turbulent jets. Very simplified kinetics was employed so that the temperature was the only indicator of the progress of the reaction. The results of their investigation suggested that age-biasing had little influence on the statistics of the mixture fraction up to fourth moment for the non-reacting jet flows. Examination of the joint pdf's of temperature and mixture fraction (in the form of scatter diagrams) also demonstrated little sensitivity to the choice of the mixing model. An implication of this result is that convection and turbulent transport may moderate any improvements which may otherwise arise from an improved mixing model in simple (e.g. homogeneous, isotropic) flows.

4.7.4 Reaction Zone Conditioning

Coalescence-dispersion closures represent jump processes i.e. they are discontinuous in time. This feature of the model is problematic partly because mixing is a continuous process and 'interacting' eddies do not suddenly jump to new concentrations. Furthermore in the flamelet regime of combustion, where chemical reaction is confined to a thin region of both physical and compositional space, a jump process allows for the possibility of two material points (or particles) to be mixed through the reaction zone *without* any reaction (figure 4.1). In this sense, the process of combustion is never 'felt' by the interacting particles.

The problem highlighted by figure 4.1 is of concern if the particles have a sufficiently high temperature for the reaction rates to be effective. With finite-rate kinetics 'cold' particles ($T < T_{lim}$) can, in principle, mix across the stoichiometric ξ -plane without appreciable reaction; this is inconsistent with the flame sheet regime where the 'mixed equals burned' criterion applies. Chen and Kollmann [14] have proposed a form of reaction zone conditioning which can account for the combined effects of chemical reaction and mixing, and requires no substantial modifications to current coalescence-dispersion proposals. With reference to figure 4.1, the conditioning involves performing mixing and reaction sequentially, in sub-steps, such that a particle is forced to pass through the flame zone, react and then resume mixing.

As discussed in section 4.7.3 with reference to [14], joint pdf's of temperature (T) and mixture fraction (ξ) were examined for turbulent jet diffusion flames. The mixing models used were the modified form of Curl's model with and without reaction zone conditioning. Two very simplified forms of reaction kinetics were specified; a flame sheet type reaction zone where $\frac{dT}{dt} = 10^{13}$

K/s inside the 'thin' reaction zone (until T reached its maximum value at a fixed ξ), and slow chemistry where $\frac{dT}{dt} = 10^6$ K/s. Outside the reaction zone the rate was set to zero. For the slow chemistry case, conditioning the mixing process on the reaction zone did not alter the joint pdf (T, ξ) noticeably relative to the pdf predicted by the unconditioned model. In the fast chemistry test case, reaction zone conditioning showed a very large effect, such that all the particle properties were situated on the equilibrium $T = T(\xi)$ line. In contrast, without conditioning, a significant number of points fell below the $T = T(\xi)$ line, demonstrating the deficient behaviour of the original model.

Norris and Pope [71] also addressed the problem of reaction zone conditioning but followed a different approach. They identified the criteria under which the effect is important: when particles are selected with values of ξ that lie on either side of ξ_{st} , and whose difference in ξ -property ($\Delta\xi$) is large compared to the width of the reaction zone. It was therefore proposed to re-order the initial sub-ensemble of particles, that were randomly selected (without replacement), in ascending values of ξ and to pair off adjacent particles - in this manner, $\Delta\xi$ would be minimized between any particle pair prior to mixing. It was claimed that this approach has a physical justification since the instantaneous conservation equation for ξ , $\frac{D\xi}{Dt} = \Gamma \nabla^2 \xi$, indicates that $\xi(\mathbf{x})$ is affected by the ξ field within an infinitesimal neighbourhood of $\mathbf{x}$, where the surrounding ξ differs infinitesimally from $\xi(\mathbf{x})$. The application of adjacent pairing of particles in ascending order of ξ in homogeneous turbulence with flame-sheet thermochemistry led to far fewer 'jumps' across ξ_{st} . However, in the non-reacting homogeneous test case it was found that such re-ordering led to pdf's with bi-modal shapes, and consequently bore no qualitative resemblance to a Gaussian distribution. Consequently a compromise pairing scheme, which effectively involved trying out different combinations of ordered-pairing sequences, was performed. At the expense of introducing extra 'jumps' across the flame zone, improved pdf shapes were obtained. It does not appear possible to extend the ordered-pairing approach to multi-scalar pdf equations without ambiguity - against which scalar property does one choose to re-order the randomly selected set of particles? The choice of a particular scalar will lead to different sequences of particles.

How much improvement can be gained in practice through the use of reaction zone conditioning has yet to be demonstrated, and since its inception little development appears to have been undertaken. Such modifications to coalescence-dispersion closures are important, however, if only because they highlight that mixing models developed in non-reacting flows may not be applicable when chemical reaction is present.

4.8 Langevin Models

Non-linear integral models (such as coalescence-dispersion closures) can be interpreted (stochastically) as Poisson processes; these form the basis of particle interaction models, Pope [81]. Random diffusion processes can also be used to simulate the behaviour of molecular mixing, and these form the basis of Langevin models. Pope [81] proposed a model for the mixing term using a Langevin equation:

$$\delta\phi(t) \equiv \phi(t + \delta t) - \phi(t) = G [\phi(t) - \langle\phi(t)\rangle] \frac{\delta t}{\tau} + \left(B \frac{\langle\phi'^2\rangle}{\tau} \right)^{\frac{1}{2}} \delta W_t \quad (4.23)$$

The rhs contains a linear deterministic term followed by a random diffusive term. G and B are constants to be determined, and B is a non-negative diffusion coefficient for this process. τ is a

characteristic turbulent time scale and δW_t is the increment of a Weiner process which is defined by the properties: $\langle \delta W_t \rangle = 0$ and $\langle \delta W_t^2 \rangle = \delta t$ where $\delta W_t = W_{t+\delta t} - W_t$. A function which satisfies these properties is: $W_t \equiv (\delta t)^{1/2} \sum_{i=1}^{\mathcal{N}_s} v_i$, where v_i ($i = 1, \dots, \mathcal{N}_s$) represents a set of independent standardized (i.e. zero mean, unity variance) normal random variables. W_t is a continuous and non-differentiable function of time. The mean is preserved in this formulation, and the requirement for the correct variance decay rate leads to a relation between G and B leaving one constant to be specified. Pope points out that from any initial condition and in the absence of any disturbing influences, the pdf relaxes to Gaussian with this model. The rate at which the pdf relaxes depends on the value of B ; for $B = 0$, LMSE is recovered for which there is no relaxation. A drawback of the model is that the term involving B can lead to the violation of scalar bounds and for this reason, the expression shown in equation (4.23) is not considered as useful for representing scalar mixing. However, for the velocity field which is not bounded, such a Langevin model can be employed.

4.9 Binomial Models

Valiño and Dopazo [94] proposed a sequential model involving LMSE and a random jump process capable of producing a Gaussian relaxation for bounded scalars. During a time step δt , two additive subprocesses take place: first, all $\mathcal{N}$ particles modify their values according to a LMSE specification and then $\mathcal{N}B'\delta t/\tau$ of the particles are randomly selected out of the $\mathcal{N}$ stochastic particles (where τ is a characteristic time scale)². The $\mathcal{N}(1 - B'\delta t/\tau)$ remaining particles do not alter their values, whilst those randomly sampled modify their values according to:

$$\phi^{(p)}(t + \delta t) - \langle \phi(t) \rangle = \varsigma^{(p)} \left\langle (\phi - \langle \phi \rangle)^2 \right\rangle^{\frac{1}{2}} \quad (4.24)$$

where $\varsigma^{(p)}$ is either a standardized Gaussian random variable if ϕ is an unbounded scalar, or a standardized binomially distributed random variable if ϕ is bounded. Since the latter case is relevant to reactive scalars, attention will focus on its representation:

$$\varsigma^{(p)} = \frac{\eta - M\mathcal{P}}{\sqrt{M\mathcal{P}(1 - \mathcal{P})}} \quad (4.25)$$

where η is a binomially distributed *integer* random variable with mean $M\mathcal{P}$ and variance $M\mathcal{P}(1 - \mathcal{P})$. $\mathcal{P}$ is a real number in $[0, 1]$ and η can take integer values between zero and M . To ensure boundedness, the minimum and maximum values of ϕ correspond to η equal to zero and M , respectively. With this requirement it can be shown:

$$M = \frac{(\langle \phi \rangle - \phi_{min})(\phi_{max} - \langle \phi \rangle)}{\langle \phi'^2 \rangle} \quad (4.26)$$

$$\mathcal{P} = \frac{(\langle \phi \rangle - \phi_{min})}{(\phi_{max} - \phi_{min})} \quad (4.27)$$

² B' is a positive dimensionless constant whose value strongly affects the relaxation rate of the pdf and its value requires calibration against measurements. Note that B' is not the same as the B appearing in equations (4.16) and (4.21), since the second operation that is applied in the binomial model affects neither the mean nor the variance (which are only affected by the application of LMSE in the first step). In equations (4.16) and (4.21), B is chosen so as to yield the correct variance decay rate.

In an homogeneous turbulence $\langle \phi'^2 \rangle$ decreases with time and thus, for fixed bounds, equation (4.26) implies that M increases. Furthermore as $M \rightarrow \infty$, the *de Moivre-Laplace* theorem shows that $\varsigma^{(p)}$ tends to a normal continuous random variable; thus a distribution close to Gaussian at long times is implied. Monte Carlo simulations of homogeneous turbulence demonstrated that the moments of the pdf evolved correctly; i.e. to within statistical error (based on finite sample size) the mean and skewness were invariant, the variance decayed exponentially and the flatness tended asymptotically to 3 - the Gaussian value. However, the rate at which the moments evolved were strongly affected by B' ; for $B' = 0.5$, the pdf's evolved towards a bell-shaped distribution, but with the appearance of two persistent spikes corresponding to the initial δ functions. With $B' = 2$ qualitatively better pdf evolution was obtained.

Subsequently Valiño and Dopazo [95] combined a LMSE and a binomial diffusion process to obtain a modified Langevin model:

$$\delta\phi(t) = \underbrace{\left\{ \mathcal{K} \left[1 - \left(\frac{\phi'(t)}{\phi'_*} \right)^2 \right] \frac{\delta t}{\frac{1}{2}\tau} \langle \phi'^2 \rangle \right\}^{\frac{1}{2}} \varsigma^{(p)}}_{\text{Random diffusion term}} - \underbrace{\left[1 + \mathcal{K} \left(1 - \frac{\langle \phi'^2 \rangle}{\phi'^2_*} \right) \right] \frac{\delta t}{\tau} (\phi(t) - \langle \phi \rangle)}_{\text{Linear deterministic term}} \quad (4.28)$$

where ϕ'^2_* is equal to ϕ'^2_{max} whenever $\phi^{(p)}(t) > 0$ and equal to ϕ'^2_{min} for $\phi^{(p)}(t) < 0$. ϕ'^2_{max} and ϕ'^2_{min} are the maximum (positive) and minimum (negative) fluctuations allowed, respectively. $\mathcal{K}$ is a constant to be adjusted to yield the correct evolution of the pdf shape and to be consistent with the scalar variance decay rate, equation (4.6). It is worth noting that the binomial model described earlier on is sequential but is neither continuous nor differentiable; the present (non-sequential) model is continuous but non-differentiable, and is therefore a more physically realistic representation of mixing. Equation (4.28) is similar to the proposal of Pope [81], equation (4.23), except that the Weiner process (which may generate unbounded scalar values) is now replaced by a standardized binomially distributed random variable, $\varsigma^{(p)}$, and defined so as to ensure boundedness as in the previous binomial model. As time increases $\langle \phi'^2 \rangle$ decreases, so that $\langle \phi'^2 \rangle / \phi'^2_* \rightarrow 0$, $\phi'(t) / \phi'_* \rightarrow 0$ and for large values of t , equation (4.28) asymptotically yields relaxation to a Gaussian pdf. Comparison of the pdf forms (using $\mathcal{K} = 2.1$) with the DNS data of Eswaran and Pope [25] demonstrated very good agreement. However, the sensitivity of the results to the value of $\mathcal{K}$ was not discussed.

Binomial models have been demonstrated to work well for pdf's of single scalars with fixed bounds. However, the ability of the model to handle multi-scalar pdf's is not clear. The bounds of a reactive scalar are not fixed; each scalar property (e.g. the specific mole number of H_2 , CO, etc. . .) has its own bounds fixed by conservation principles and the allowable scalar domain is a complicated hypervolume in multi-dimensional composition space. Now, in Monte Carlo simulations, equations (4.26) and (4.27) would be used to select η from a random number generator. For a given stochastic particle, a binomial distribution characteristic of each scalar property (in terms of M and P) is required from which η is to be selected. Furthermore the bounds for each property can differ from one particle to another, and these bounds evolve as a result of mixing and reaction. Thus, in principle it is possible for different values of M and P to be specified for each property and for each particle over any particular time step. This has the implication that only one sample would be selected from any given binomial distribution characterised by M and P . The consequences of this are at present unclear, but it appears inconsistent with the idea of sampling

random numbers with a fixed distribution.

4.10 Mapping Closures

In the literature dealing with mixing models, the quest for predicting Gaussian pdf's in homogeneous turbulence appears to have culminated in the proposal for mapping closures. This closure has received a considerable amount of attention over a relatively short period of time, partly because of its ability to predict (in analytical form in certain cases) the correct evolution and asymptotic behaviour of passive scalar mixing in homogeneous fields. Chen et al. [11] addressed the problem of closure for the mixing term by constructing $\phi(\mathbf{x}, t)$ as a time dependent mapping of a Gaussian reference field; the evolution of the mapping characterizes the evolution of the scalar field for which the pdf is sought. The mapping technique is capable of handling strongly non-Gaussian $\phi(\mathbf{x}, t)$. The pdf equation for homogeneous turbulent scalar fields is considered in the following form:

$$\frac{\partial P(\psi; t)}{\partial t} + \frac{\partial}{\partial \psi} \left[E \{ D \nabla^2 \phi | \psi \} P(\psi; t) \right] = 0 \quad (4.29)$$

where D is the diffusivity and $E \{ D \nabla^2 \phi | \psi \}$ is the expectation of $D \nabla^2 \phi$ conditioned on $\phi(\mathbf{x}, t) = \psi$. The cumulative distribution function (cdf) of $P(\psi; t)$ is defined as:

$$F(\psi; t) \equiv \int_{-\infty}^{\psi} P(\hat{\psi}; t) d\hat{\psi} \quad (4.30)$$

Equation (4.30) taken together with (4.29) leads to the transport equation for the cdf:

$$\frac{\partial F(\psi; t)}{\partial t} + E \{ D \nabla^2 \phi | \psi \} \frac{\partial F(\psi; t)}{\partial \psi} = 0 \quad (4.31)$$

The closure establishes the following mapping relation [82, 54]:

$$\phi_s(\mathbf{x}, t) \equiv X[\theta(\mathbf{z}), t] \quad (4.32)$$

$\phi(\mathbf{x}, t)$ is the dependent scalar field and $\theta(\mathbf{z})$ is a standardized Gaussian reference field residing at a spatial location $\mathbf{z}$. $\phi_s(\mathbf{x}, t)$ is referred to as the surrogate field [82] and is effectively defined through equation (4.32). The mapping X does not determine the relation between the locations of $\phi(\mathbf{x}, t)$ and $\phi(\mathbf{z})$. These are related by a time-dependent stretching transformation given by [54]:

$$\frac{\partial z_\beta}{\partial x_\alpha} = \delta_{\alpha\beta} m(t) \quad (4.33)$$

The mapping, X , is constructed as follows: the value of the cdf of a turbulent field at $X(\eta, \mathbf{x}, t)$ (η is the sample space of the Gaussian reference field) is equal to the value of the standardized Gaussian cdf at $\theta(\mathbf{z}) = \eta$:

$$F(X(\eta, \mathbf{x}, t)) = F_G(\eta) \quad (4.34)$$

implying the relation $\psi = X(\eta, \mathbf{x}, t)$. A direct consequence of the monotonicity of the cdf's is that the mapping increases monotonically too, i.e. $X(\eta_1) < X(\eta_2)$ for $\eta_1 < \eta_2$. Differentiating equation (4.34) with respect to η :

$$\frac{\partial F}{\partial \psi} \frac{\partial X}{\partial \eta} = \frac{\partial F_G}{\partial \eta} \Rightarrow P(\psi; t) = P_G \left(\frac{\partial X}{\partial \eta} \right)^{-1} \quad (4.35)$$

Performing chain differentiation of equation (4.34) with respect to time yields:

$$\frac{\partial F}{\partial t} + \frac{\partial F}{\partial \psi} \frac{\partial X}{\partial t} = 0 \quad (4.36)$$

Substitution of equation (4.31) into (4.36) coupled with the fundamental assumption [82, 30] that *the unknown statistics of the turbulent field are the same as the known statistics of the surrogate field*, leads to:

$$\frac{\partial}{\partial t} X(\eta, \mathbf{x}, t) = E \left\{ D \nabla^2 \phi_s | \psi_s = X(\eta, \mathbf{x}, t) \right\} \quad (4.37)$$

Further manipulations are described in Kollman [54], and lead to the following form for the evolution equation of the mapping function:

$$\frac{\partial X}{\partial t} = m^2 D \left\langle \frac{\partial \theta}{\partial z_i} \frac{\partial \theta}{\partial z_i} \right\rangle \left(X'' - \frac{\eta}{\langle \theta^2 \rangle} X' \right) \quad (4.38)$$

where $\langle \theta^2 \rangle = 1$. D is treated as spatially uniform. Since equation (4.29) makes use of the mixing term employing Fickian diffusion, this strictly implies that D cannot vary from one specie to another (refer to equation (2.16))- thus it appears that this level of closure is constrained to an equal diffusivity assumption. The mapping closure described above relates to single point statistics. As in all other one-point, one-time closures, length scale information is not available; $\langle \frac{\partial \theta}{\partial z_i} \frac{\partial \theta}{\partial z_i} \rangle$ in equation (4.38) requires external information and is similar in form to a scalar dissipation. Valiño and Gao [97] suggest the use of a characteristic time k/ε , which is what is used in other mixing models. Another possibility is the modelling of the joint scalar and its gradient [96]. In this case $\tau_\phi \equiv \langle \phi'^2 \rangle / \langle D \nabla \phi' \nabla \phi' \rangle$ can be obtained directly from the joint pdf. How easily the latter option can be implemented is still not clear.

Gao [29] presented a closed form solution to equation (4.38). Two important features of the solution were that it preserved the boundedness of the scalar field and also predicted relaxation to a Gaussian distribution. Pope [82] and Valiño and Dopazo [95] obtained analytical solutions to equation (4.38) for the case of $P(\psi; 0) = \frac{1}{2}[\delta(\psi + 1) + \delta(\psi - 1)]$. Excellent agreement was found with the relaxation of pdf's arising from DNS. Monte Carlo simulations of the mapping closure for this test case [98, 97] led to equally good agreement.

How useful mapping closures will be for prediction methods in practical flows is still an open question and the problems associated with incorporating extra scalars (inert and reactive) in a mathematically and computationally tractable manner need to be resolved. Furthermore, although such a technique yields excellent results in homogeneous turbulence, it is not clear whether it will necessarily yield better results in inhomogeneous flows where the turbulent time scales can vary and are not known. The use of k/ε as the only characteristic time scale may limit its usefulness relative to simpler and cruder models previously discussed.

4.11 Closing Remarks Concerning Mixing Models

Several techniques for closing the molecular mixing term have been discussed, each with varying degrees of success in simple configurations. For practical applications to inhomogeneous reactive flows involving several scalars and incorporating realistic chemistry, LMSE and coalescence-dispersion models appear to be the most widely used. This is essentially because they can be extended without any significant additional effort to more complicated situations. In this regard,

many of the models previously discussed are still in the early development stages and their capabilities have yet to be demonstrated.

4.12 Models for the Turbulent Transport of the Pdf

Traditional modelling practice suggests that in many flows turbulent transport can be adequately represented through a gradient transport hypothesis. For many of the applications of the pdf method to thin shear flows it appears to yield acceptable results for quantities relating to the mixing patterns (e.g. mixture fraction). So term IV in equation (4.3) can be approximated as:

$$-\underbrace{\frac{\partial}{\partial x_k} \left\{ \bar{\rho} \langle u_k'' | \phi = \psi \rangle \tilde{P}(\psi) \right\}}_{\text{IV}} \approx \frac{\partial}{\partial x_k} \left[\frac{\mu_t}{\sigma_t} \frac{\partial \tilde{P}(\psi)}{\partial x_k} \right] \quad (4.39)$$

where $\sigma_t = 0.7$ and μ_t is given by the k - ε model (section 2.5.1). With a second moment closure being used for the velocity field, Chen and Kollmann [12] used the following approximation:

$$-\underbrace{\frac{\partial}{\partial x_k} \left\{ \bar{\rho} \langle u_k'' | \phi = \psi \rangle \tilde{P}(\psi) \right\}}_{\text{IV}} \approx \frac{\partial}{\partial x_k} \left[c_s \bar{\rho} \frac{k}{\varepsilon} \widetilde{u_k'' u_j''} \frac{\partial \tilde{P}(\psi)}{\partial x_j} \right] \quad (4.40)$$

with $c_s = 0.3$.

In the past other proposals for closing the turbulent diffusion term have been suggested. Dopazo [20] tentatively proposed a LMSE type approximation and such an expression can be expected to work well in nearly Gaussian cases. Janicka et al. [39] used a modelled transport equation for $\langle u_k' | \phi = \psi \rangle$ (i.e Reynolds averaging and for a single conserved scalar) - the modelling involved a formidable task. Certain terms involving the pressure were either neglected through ad-hoc assumptions or through order of magnitude analysis and conditional Reynolds stresses were modelled using gradient transport hypothesis. However, these methods have now been largely replaced by the techniques referred to in the previous paragraph.

The modelling of the turbulent diffusion and molecular mixing (representative of the mixing process at the energy-containing and fine scales of turbulent motion, respectively) make use of $\frac{k}{\varepsilon}$ as the *only* characteristic turbulent time scale. This is true regardless of the dependent scalar variable, and consequently the fundamental implication is that equal diffusivity of the scalars is being assumed since there is no information available on the distribution of scalar time scales.

4.13 The Monte Carlo Method for Solving the Scalar Pdf Equation

Having discussed in some detail the modelling of the scalar pdf equation, the problem of solving the equation has to be addressed. In doing so, no assumptions are made concerning the form of the mixing model to be employed, although it is assumed that a particle method can be constructed to represent it. However, the turbulent diffusion of pdf is approximated by gradient modelling as discussed in section 4.12. The specific details of how the Monte Carlo procedure is implemented, for the purpose of the predictions reported in chapter 6, is fully described in appendix E. Presently, only a brief overview of the general underlying principles are discussed.

The solution formalism for the scalar pdf transport equation is based on the method of fractional steps. First equation (4.3) is expressed in the following form:

$$\frac{\partial \tilde{P}}{\partial t} + \tilde{u}_k \frac{\partial \tilde{P}}{\partial x_k} + \frac{\partial}{\partial \psi_\alpha} (\tilde{P} \dot{S}_\alpha) = \frac{1}{\bar{\rho}} \frac{\partial}{\partial k} \left\{ \frac{\mu_t}{\sigma_t} \frac{\partial \tilde{P}}{\partial x_k} \right\} + \Xi(\underline{\psi}; \mathbf{x}, t) \quad (4.41)$$

where summation is implied over repeated indices (including greek subscripts). Equation (4.41) is then discretised using an explicit forward time finite-difference approximation which may be written in the following form:

$$\tilde{P}(\underline{\psi}; \mathbf{x}, t + \Delta t) \approx \{\mathbf{I} + \Delta t(\mathbf{T} + \Xi + \Omega)\} \tilde{P}(\underline{\psi}; \mathbf{x}, t) \quad (4.42)$$

where $\tilde{P}$ is the solution vector, $\mathbf{I}$ is the identity matrix and $\mathbf{T}$, Ξ and Ω are difference coefficient matrices associated with transport (convection and diffusion), molecular mixing and chemical reaction. For numerical stability the convective contribution to $\mathbf{T}$ must either correspond to upwind differencing or alternatively hybrid (upwind-central) differencing can be used for the complete term. Performing an approximate factorization on equation (4.42) yields:

$$\tilde{P}(\underline{\psi}; \mathbf{x}, t + \Delta t) \approx \underbrace{(\mathbf{I} + \Omega \Delta t)}_{\text{Source}} \underbrace{(\mathbf{I} + \Xi \Delta t)}_{\text{Mixing}} \underbrace{(\mathbf{I} + \mathbf{T} \Delta t)}_{\text{Transport}} \tilde{P}(\underline{\psi}; \mathbf{x}, t) \quad (4.43)$$

Equation (4.43) can now be solved via a sequence of fractional steps. First the effect of transport is calculated from: $\tilde{P}(\underline{\psi}; \mathbf{x}, t + \Delta t_T) = (\mathbf{I} + \mathbf{T} \Delta t) \tilde{P}(\underline{\psi}; \mathbf{x}, t)$ where Δt_T is a notional time step indicating that transport has been applied. The mixing operation is applied to $\tilde{P}(\underline{\psi}; \mathbf{x}, t + \Delta t_T)$ and the result of this is then acted upon by the chemical reaction operator. On completion of these steps an approximation to $\tilde{P}(\underline{\psi}; \mathbf{x}, t + \Delta t)$ is obtained. In the work of Pope [79] a slightly different factorisation was applied whereby the convection and diffusion operators were represented separately. There appears to be no advantage in adopting such a practice since convection would then have to be represented using (first-order accurate) upwind differencing. In the approach outlined in appendix E, second-order accurate central differencing is used for cell Peclet numbers (Pe) less than two, with upwinding being applied for larger Pe , giving a somewhat more accurate representation of transport.

In order to minimise the computational effort, a particle method (or Monte Carlo simulation) is employed. For mixing this has already been discussed and the simulation of transport will now be described. In the present context the Monte Carlo method is not a statistical simulation of the pdf transport equation, but of the finite-difference representation of the pdf equation. The simulation is performed on a finite-difference grid and rather than considering $\tilde{P}(\underline{\psi}; \mathbf{x}, t)$ explicitly, an ensemble of $\mathcal{N}$ 'representative' particles are located at each grid node. Each particle can carry with it $N + 1$ properties corresponding to the random variables of the pdf. The ensemble $\Phi(\mathbf{x}, t)$ at a point can be compactly represented in terms of its individual particles $\phi_\alpha^{(i)}$ as follows:

$$\Phi(\mathbf{x}, t) = \begin{pmatrix} \underline{\phi}^{(1)} \\ \vdots \\ \underline{\phi}^{(\mathcal{N})} \end{pmatrix} = \begin{pmatrix} \phi_1^{(1)}, & \dots, & \phi_{N+1}^{(1)} \\ \vdots & & \vdots \\ \phi_1^{(\mathcal{N})}, & \dots, & \phi_{N+1}^{(\mathcal{N})} \end{pmatrix} \quad (4.44)$$

From such a representation, average values of any function of $Q[\underline{\phi}(\mathbf{x}, t)]$ are given by:

$$\langle Q[\underline{\phi}(\mathbf{x}, t)] \rangle = \frac{1}{\mathcal{N}} \sum_{i=1}^{\mathcal{N}} Q[\underline{\phi}^{(i)}] \quad (4.45)$$

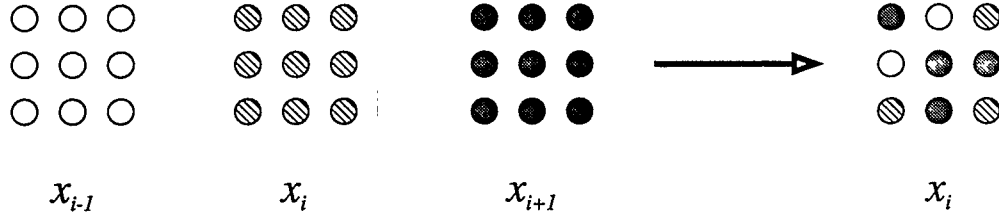


Figure 4.2: Schematic of representative particles. Random sampling without replacement from x_{i-1} , x_i and x_{i+1} leads to the new ensemble at x_i . The shading of the particles only identifies where the particles originate from. In this notation, x refers to the coordinate direction, and the subscript i identifies the mesh position in that direction ($i = 1, \dots, N+1$).

An important consideration is whether the simulation constitutes a correct representation of the pdf equation. Pope [79] defined "equivalence" as follows: If for all functions Q , the ensemble average $\langle Q \rangle$ converges in probability to the expected value $\tilde{Q}$, then $\Phi(\mathbf{x}, t)$ and $\tilde{P}(\mathbf{x}, t)$ are equivalent. Convergence in probability means [73] that for a number $\epsilon > 0$, the probability of the event $\{|\langle Q \rangle - \tilde{Q}| > \epsilon\}$ tends to zero as $\mathcal{N} \rightarrow \infty$.

The Monte Carlo method simulates the process of transport (convection plus diffusion) in physical space by shifting element properties (chosen at random) from node to node; this means that node $\mathbf{x}$ would interact with its nearest neighbours and figure 4.2 shows schematically the nature of such interactions along one particular direction (in a Cartesian frame of reference).

The groups of particles on the lhs of the arrow are representative of the situation at time t , whilst the application of the transport operation leads to the new ensemble at node x_i at time $t + \Delta t_T$. The number of representative elements chosen from x_{i-1} and x_{i+1} , referred to as $\mathcal{N}_{x_{i-1}}$ and $\mathcal{N}_{x_{i+1}}$ (respectively), is very important if equivalence is to be achieved between the Monte Carlo simulation and the finite-difference solution. The analysis of Pope [79] demonstrated that $\mathcal{N}_{x_{i\pm 1}} = \mathcal{N} \times \text{finite-difference coefficient for the transport term (contributing to the effect on } x_i \text{ from } x_{i\pm 1})$. Clearly the two methods (fde and Monte Carlo) are complementary i.e. even though fde's are not solved, the coefficients of the fde's have to be computed prior to the application of the diffusion and convection simulation using the Monte Carlo technique. Thus the new ensemble at node x_i will comprise $\mathcal{N}_{x_{i-1}}$ particles from node x_{i-1} , $\mathcal{N}_{x_{i+1}}$ from node x_{i+1} and $(\mathcal{N} - \mathcal{N}_{x_{i-1}} - \mathcal{N}_{x_{i+1}})$ from x_i . In order to form the new ensemble at x_{i+1} , the neighbouring nodes x_i and x_{i+2} (not shown in figure 4.2) are employed. In the case of x_i , the newly formed ensemble is *not* used for the purpose of randomly selecting particles, but the original ensemble prior to modification (as illustrated in figure 4.2 on the lhs of the arrow) is employed. This avoids the possibility of returning the same particles that were copied over from x_{i+1} in the previous step. Note that in accordance with hybrid differencing, if the cell Peclet number is greater than two, central differencing switches to upwind differencing; under these circumstances, x_i would interact with x_{i+1} or x_{i-1} only (depending on the direction of the local velocity field) in order to form the new ensemble at x_i .

The source term for chemical reaction is simulated deterministically. In principle, each particle reacts independently for an interval of time Δt according to a system of ordinary differential equations (ODE's),

$$\frac{d}{dt} (Y_\alpha^{(i)}) = \frac{\dot{S}_\alpha(\phi^{(i)})}{\varrho(\phi^{(i)})} \quad \text{where} \quad \alpha = 1, \dots, N \quad \text{and} \quad 1 \leq i \leq \mathcal{N} \quad (4.46)$$

For combusting flows these ODE's correspond to a stiff and coupled set of equations and their solution is non-trivial [38]. For the oxidation of hydrocarbons up to propane it is claimed [76] that typically one hundred reactions and thirty species are necessary. The implementation of a detailed reaction scheme in a transported pdf calculation would be prohibitively expensive due to the large number of independent scalars that need to be specified. Fortunately, many (laminar) flame characteristics do not require such a detailed level of description; reduced reaction mechanisms involving a 'handful' of species can simulate the salient features of their 'parent' detailed mechanism and reproduce measurements in *laminar flames* with acceptable accuracy.

Despite the limitations of computer speed and memory, which preclude the use of detailed kinetics, Chen et al. [15] have estimated that 1000 cpu hours on a Cray XMP-48 computer would be required for the simulation of jet flames with a reduced scheme involving five scalars and $N \approx 50000$. One strategy for significantly reducing the very large computational demand is the calculation of the integrals of the reaction rates for all possible chemical states and then storing the results in an *once and for all* 'look-up' table. During Monte Carlo simulations the changes of the scalar properties, as a result of reaction, are obtained from multi-linear interpolations based on the previously generated look-up table; this replaces the repeated time integration of the stiff ODEs. This strategy is computationally very efficient, but, the overriding constraint becomes the size of the 'look-up' table. As the complexity of the reduced mechanism increases, the number of independent³ scalars required to specify the thermochemistry increases. For interpolation tables including more than six scalars the demands on memory can become quite unmanageable if a sufficiently fine resolution of the chemistry is required; coarse grids may lead to solutions casting doubts on the accuracy of the results [86]. The scalar bounds for the interpolation tables are constructed using mass conservation principles and the assumption of constant carbon-to-hydrogen (C/H) and oxygen-to-nitrogen (O/N) atom ratios [15, 86]; this latter assumption is consistent with the idea of equal diffusivity⁴ which is implied both by the turbulence models and the mixing models. Based on these requirements it is possible to construct a table where a substantial part of the scalar domain would be redundant i.e. reaction would never occur in those regions, because the temperature is too low. For this reason, Sion and Chen [86] clipped some of the scalar bounds (e.g. for H-radicals) based on an arbitrary factor (typically three to four) of the maximum concentration of the scalar which would be found in a typical laminar flame. In very recent work [2] the concept of the interpolation table has been replaced by direct solution of the ODE's during Monte Carlo simulations and the computational effort is made manageable through the use of parallelization techniques.

³The meaning of 'independent scalar' needs to be clarified since it is frequently referred to in subsequent chapters. The scalar field is comprised of the species concentrations and the enthalpy. Independent scalars characterise the thermochemical field, such that once their value is known, the remainder of the scalars can be calculated.

⁴Equal diffusivity implies that the (C/H) and (O/N) ratios are constant if there is a two-stream mixing process where the fuel and air are in separate streams.

Chapter 5

Details of the Calculation Procedure

5.1 Introduction

In this chapter the main features of the calculation method for predicting the flows considered in chapter 6 will be discussed. These flows include jet flames (laminar and turbulent) and homogeneous, isotropic turbulence. The discussion of the solution procedure for jet flames will encompass the treatment of the hydrodynamic field (in terms of the momentum conservation equation) and the scalar pdf transport equation.

5.2 The Patankar-Spalding Method

5.2.1 The Basic Geometry of the Jet Configuration

The Patankar-Spalding method [74] is now a well-established method of solving the equations describing boundary layer flows and was originally embodied in a computer code referred to as GENMIX¹. A schematic of the typical jet flow geometry is shown in figure 5.1.

The fundamental requirement imposed by the boundary layer approximation is: $\frac{\delta}{L} \ll 1$, i.e a thin shear layer. Using figure 5.1, the following order of magnitude estimates can be expressed: $\frac{\partial \bar{u}}{\partial x} \sim \mathcal{O}(\frac{u'}{L})$ and $\frac{\partial \bar{u}}{\partial r} \sim \mathcal{O}(\frac{u'}{\delta})$. The boundary layer approximation implies that $|\frac{\partial \bar{u}}{\partial x}| \ll |\frac{\partial \bar{u}}{\partial r}|$. The use of the mass continuity equation leads to, $\bar{v} \sim \mathcal{O}(\frac{u'\delta}{L})$, so that $|\bar{v}| \ll |\bar{u}|$. Since a non-swirling, statistically stationary and two-dimensional axisymmetric flow is considered, $\bar{w} = 0$. Consequently the velocity field can be calculated in terms of mass continuity and the x -component momentum equation.

5.2.2 The General form of the Equations

The equations used to describe the evolution of the axisymmetric, high Reynolds number jet flow are given in appendix B. The momentum equation is closed using either an eddy viscosity (k - ϵ) or full Reynolds stress model. The mean density is required as an input from the scalar field calculation; the mean density and the species concentration field are obtained from the solution of the modelled pdf transport equation. In order to solve the pdf equation, $\tilde{u}$, k and ϵ are needed; this illustrates how the velocity and scalar fields are coupled at this level of closure. Appendix B

¹GENMIX is an abbreviation for 'GENeral MIXing'

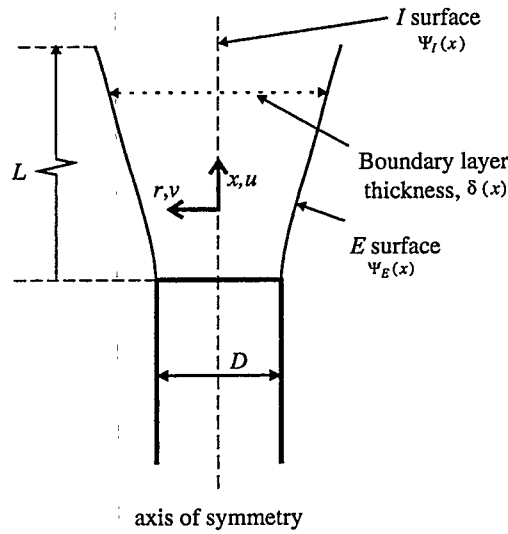


Figure 5.1: Schematic of a jet flow. Ψ denotes the stream function and since only the half-jet is calculated (due to the axisymmetry of the flow), the I and E surfaces represent the required solution domain.

shows that the boundary layer equations (excluding mass continuity) all share a common form, and can be expressed as:

$$\bar{\rho}\tilde{u}\frac{\partial\tilde{\phi}}{\partial x} + \bar{\rho}\tilde{v}\frac{\partial\tilde{\phi}}{\partial y} = \frac{1}{r}\frac{\partial}{\partial y}\left(r\Gamma_{\phi}\frac{\partial\tilde{\phi}}{\partial y}\right) + S_{\phi} \quad (5.1)$$

In cylindrical polar coordinates the variable y can be interpreted as r , and so $\tilde{u}$, $\tilde{v}$ are the axial and radial velocities, respectively. Equation (5.1) and the subsequent finite-difference discretisation are equally applicable to two-dimensional plane flows where Cartesian coordinates would be employed. In this case, $r = 1$ and $\tilde{u}$, $\tilde{v}$ are then interpreted as velocities in a Cartesian frame of reference. $\tilde{\phi}$ can represent $\tilde{u}$, k , ε , $\overline{u'^2}$, $\overline{v'^2}$, $\overline{w'^2}$, $\overline{u'v'}$ or $\tilde{P}(\psi)$; and S_{ϕ} is representative of the source of ϕ . For example if $\tilde{\phi} \equiv \tilde{u}$ and a second moment closure is used then, $S_{\tilde{u}} = -\frac{1}{r}\frac{\partial}{\partial r}(r\bar{\rho}\tilde{u}'') - \frac{\partial p}{\partial x} + \bar{\rho}g_x$ and $\Gamma_{\tilde{u}} = \mu$.

5.2.3 The Choice of Coordinate System

The motivation in the development of the original GENMIX code [74] was to solve the boundary layer equations in an efficient and economical manner. An important consideration in this respect is the choice of the coordinate system, since this affects the efficiency with which the grid nodes (used in the finite-difference simulation of the pde's) would be distributed. For example, a common choice of coordinate system is the Cartesian (x, y) frame of reference. This is inefficient since the developing boundary layer grows in thickness in the y -direction as it is convected in the mainstream (x) direction; now if the grid spacing is such that a certain maximum number of grid points lie within the thickness of the layer at the upstream end, then the distribution will be excessively fine (and hence wasteful) at the downstream end. Similarly, a spacing designed for the downstream layer will be too coarse for the upstream region, and so the accuracy there will be poor, as shown in figure 5.2.

A coordinate system where y is replaced by Ψ , the stream function, is no less wasteful since these two quantities are related by $\delta\Psi = \rho ur \delta y$. The *ideal* solution would be to have a grid

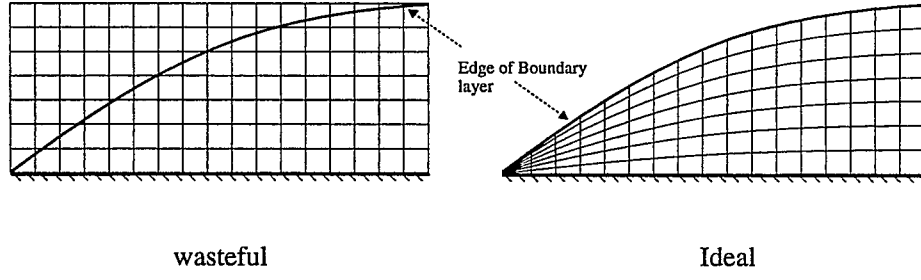


Figure 5.2: Schematic of the two possible coordinate systems which can be used to track the evolution of the boundary layer.

which always fits the boundary layer. The requirement for this is that the cross-stream variable is allowed to vary between *fixed* limits, and not one which increases with the x -direction; in addition it should vary monotonically across the mixing layer. Patankar and Spalding proposed the use of a *dimensionless stream function*, ω , as the appropriate cross-stream variable.

The mass continuity equation can be expressed as,

$$\frac{\partial}{\partial x}(\bar{\rho}\tilde{u}r) + \frac{\partial}{\partial y}(\bar{\rho}\tilde{v}r) = 0 \quad (5.2)$$

where $y = r$ for cylindrical polar coordinates and $r = 1$ in Cartesian coordinates. The stream function, Ψ , can be defined so as to satisfy equation (5.2),

$$\left(\frac{\partial\Psi}{\partial y}\right)_x \equiv \bar{\rho}\tilde{u}r \quad \left(\frac{\partial\Psi}{\partial x}\right)_y \equiv -\bar{\rho}\tilde{v}r \quad (5.3)$$

The dimensionless stream function, ω , is defined by,

$$\omega \equiv \frac{\Psi - \Psi_I}{\Psi_E - \Psi_I} \quad (5.4)$$

where Ψ_I and Ψ_E are the values of the stream function at the I and E boundaries (as portrayed in figure 5.1); thus, ω equals zero and unity at the I and E boundaries, respectively. Ψ_I and Ψ_E are chosen so that they are only functions of x , i.e. $\Psi_I = \Psi_I(x)$ and $\Psi_E = \Psi_E(x)$. Such a specification is suitable provided all the important variations in the variables of interest take place between $0 \leq \omega \leq 1$. The transformation from (x, Ψ) to (x, ω) coordinates is performed because the use of (x, ω) combines the advantages of the stream function coordinates (i.e. the mass continuity equation is automatically satisfied), *and* restricts the boundary layer to a finite range in ω .

From equation (5.3) and the fact that $\Psi_{I,E} = \Psi_{I,E}(x)$,

$$\left(\frac{\partial\Psi}{\partial x}\right)_I = \frac{d\Psi_I}{dx} = -(\bar{\rho}\tilde{v})_I r_I \equiv -r_I \dot{m}_I \quad (5.5)$$

A similar equation can be written for the E -boundary. The main result is,

$$\frac{d\Psi_I}{dx} \equiv -r_I \dot{m}_I \quad \frac{d\Psi_E}{dx} \equiv -r_E \dot{m}_E \quad (5.6)$$

The quantities $r_I \dot{m}_I$ and $r_E \dot{m}_E$ represent the mass entrainment rates (i.e. the rate of addition of fluid) *into the solution domain* encompassed by $0 \leq \omega \leq 1$. Their values are chosen, using

expressions which are to a certain degree ad-hoc, so that the solution domain just covers the region of interest; this is the region where the variable ϕ in equation (5.1) shows its most significant variation. The I boundary in the present configuration is an axis of symmetry, consequently there is a zero gradient condition there and there can be no mass flux across it. This means that $\frac{d\Psi_I}{dx} = 0$. In contrast, the E boundary is a free boundary, and physically the jet spreads by entraining fluid from the surroundings. Consequently the solution domain needs to expand in order to resolve the behaviour of the spreading jet. Patankar and Spalding [74] demonstrate that just inside the boundary layer, $r_E \dot{m}_E$ can be approximated by:

$$r_E \dot{m}_E \approx \lim_{\omega \rightarrow 1} \left\{ \frac{\frac{\partial}{\partial \omega} \left[\frac{\rho u r^2 \mu}{(\Psi_E - \Psi_I)} \frac{\partial u}{\partial \omega} \right]}{\left(\frac{\partial u}{\partial \omega} \right)} \right\} \quad (5.7)$$

The specification used for $r_E \dot{m}_E$ in this thesis follows equation (5.7) quite closely. The value of $r_E \dot{m}_E$ affects the location of the edge grid node. If the grid does not spread fast enough, the effect will show up as discontinuities in the radial profiles of ϕ since the free-stream properties will appear abruptly; furthermore, if the grid expands too quickly per integration step, this will lead to a wasteful distribution of nodes in regions where there is very little variation (if any) of the variable of interest, and the regions of significant change would be confined to a small part of the solution domain. The *physical spreading* of the jet and the *grid expansion* are two separate quantities; the former is an outcome of the physics of the problem and is reproduced through a correct representation of the governing differential equations, whilst the latter is a parameter chosen by the analyst in order to capture the significant features of the flow. Consequently, increasing $r_E \dot{m}_E$ increases the entrainment of fluid into the solution domain by expanding the finite-difference grid more rapidly. However, this does not mean that the jet spreads any faster.

Going from (x, y) to (x, Ψ) coordinates is referred to as the *Von Mises* transformation. The details of the transformation from (x, y) to (x, Ψ) and subsequently to (x, ω) coordinates can be found in appendix C. The important result is equation (C.13) which expresses the general form of the governing pde's:

$$(\Psi_E - \Psi_I) \frac{\partial \phi}{\partial x} + (a + b\omega) \frac{\partial \phi}{\partial \omega} = \frac{\partial}{\partial \omega} \left[\frac{\rho u r^2 \Gamma_\phi}{(\Psi_E - \Psi_I)} \frac{\partial \phi}{\partial \omega} \right] + \frac{(\Psi_E - \Psi_I)}{\rho u} S_\phi$$

where $a = r_I \dot{m}_I$, $b = r_E \dot{m}_E$ and $(\Psi_E - \Psi_I)$ is a function of x only.

5.2.4 The Finite-Difference Approximations and Numerical Solution Method

Equation (C.13) needs to be solved numerically, and the finite-difference procedure employed for this purpose is discussed in considerable detail in appendix D. The method of discretisation that is employed is not entirely the same as that used in the original version of GENMIX.

In summary, the control-volume approach is used to discretise the pde; the axial convection term is approximated using a backward-space Euler difference and the cross-stream *transport* is represented by hybrid differencing. The 'source' terms (i.e. those terms which do not fit the form of the axial and radial transport in equation (C.13)) are decomposed using a quasi-linearisation. The fde is formulated in a manner which allows solution via Gaussian elimination (also referred to as the tri-diagonal matrix algorithm [87]):

$$\mathcal{E}_i \phi_i^n = \mathcal{P}_i \phi_{i+1}^n + \mathcal{Q}_i \phi_{i-1}^{n-1} + \mathcal{J}_i \quad (5.8)$$

where the definitions of coefficients $\mathcal{E}$, $\mathcal{P}$, $\mathcal{Q}$ and $\mathcal{J}$ are given in appendix D. One particular detail not described in the appendix is that in the discretisation of the Reynolds stress equation, $(\widetilde{u''v''})$ is staggered in the ω -direction relative to the nodes at which the $\tilde{u}$ velocity and the other variables (including the normal stresses) are located². The pressure gradient term is not treated as an unknown, particularly for jets issuing into unconfined surrounds where $\frac{\partial \bar{p}}{\partial x} = 0$; however, even for confined (thin shear layer) flows the Patankar-Spalding method [74] calculates the pressure gradient in a direct (and somewhat ad-hoc) manner so that iterative calculations are avoided.

5.3 The Treatment of the Scalar Field

In order to calculate turbulent jet diffusion flames, the scalar field is evaluated using $\tilde{P}(\psi; x, \omega)$. The thin shear layer form of the governing equation is given in appendix B. Equation (4.43) demonstrates that a solution for advancing $\tilde{P}$ can be achieved in terms of an operator splitting technique followed by an approximate factorisation. In the present context, the relevant form of this equation would be:

$$\tilde{P}(\psi; x + \Delta x, \omega) \approx \underbrace{(\mathbf{I} + \mathbf{\Omega}\Delta t)}_{\text{Source}} \underbrace{(\mathbf{I} + \mathbf{\Xi}\Delta t)}_{\text{Mixing}} \underbrace{(\mathbf{I} + \mathbf{T}\Delta t)}_{\text{Transport}} \tilde{P}(\psi; x, \omega) \quad (5.9)$$

The approximate factorisation is performed so that the physical operations (i.e. transport, molecular mixing and chemical reaction) can be applied separately rather than simultaneously. In appendix E the application of each of these processes (except chemical reaction which is described in section 5.3.1) and their associated Monte Carlo representation is discussed in detail; whilst important aspects of the analysis have already been described in section 4.13, the details of the implementation remain to be clarified.

The Monte Carlo method is employed simply because the finite-difference approach would be prohibitively demanding for the computation of a multi-dimensional pdf. At each of the finite-difference mesh points particles are located and the evolution of their ensemble approximates the evolution of the pdf according to the fde. The particles are *notional*; in other words a physical interpretation cannot be conferred upon them. Therefore these particles, at the level of the single point scalar pdf, should not be confused with fluid elements being transported through the fluid medium. Furthermore, as described in section 4.13, the Monte Carlo simulation for transport is performed by moving (or shifting) particle properties between neighbouring nodes. This movement is not indicative of the turbulent fluctuations in the flow, or even of the apparent 'randomness' of turbulence; it is purely a technique designed to approximate part of the finite-difference representation of the pdf equation which involves only the convective and diffusive terms.

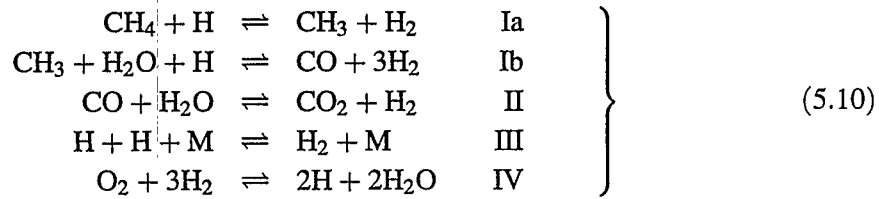
5.3.1 The Details of the Thermochemistry

A significant part of the work undertaken during this project was related to the implementation of two reduced reaction mechanisms. They differ in their degree of complexity and in the manner of their derivation. The schemes are the five-step *systematically reduced* mechanism of Chelliah et al. [10] for CH_4/air combustion, and the four-step *global* hydrocarbon mechanism of Jones and Lindstedt [46].

²The normal stresses are not staggered because they do not appear explicitly in the $\tilde{u}$ momentum equation.

5.3.1.1 The Systematically Reduced Scheme of Chelliah, Seshadri and Law

The reduced mechanism of Chelliah et al. [10] has been constructed from a *systematic reduction* of a detailed CH₄/air reaction scheme (known as the C₁-mechanism, [76]). The systematic reduction process centres around *steady-state* approximations for some of the species. This approximation implies that over the range of the mixture fraction relevant to the flame zone (typically, $0 \leq \xi \leq 0.3$) the production and consumption terms in the evolution equation for the species in question are of the same magnitude, and the transport terms are negligible³. By introducing steady-state approximations for the species CH₂, CH, CH₂O, HCO, OH, O, HO₂ and H₂O₂, the following five-step mechanism was deduced from the starting mechanism:



'M' denotes a third body and its molar concentration is defined as [76]:

$$[\text{M}] \equiv 6.5 [\text{CH}_4] + 6.5 [\text{H}_2\text{O}] + 1.5 [\text{CO}_2] + 0.75 [\text{CO}] + 0.4 [\text{O}_2] + 0.4 [\text{N}_2] + 1.0 [\text{other}] \quad (5.11)$$

where the square brackets surrounding the chemical symbol represents a molar concentration (Kmol/m³). The net forward reaction rates for steps Ia-IV in equation (5.10) can be expressed in terms of *elementary* reaction rates:

$$\left. \begin{array}{ll} \dot{\omega}_{\text{Ia}} &= -\dot{\omega}_{34} + \dot{\omega}_{38} + \dot{\omega}_{39} + \dot{\omega}_{40} \\ \dot{\omega}_{\text{Ib}} &= \dot{\omega}_{33} + \dot{\omega}_{35} + \dot{\omega}_{37} \\ \dot{\omega}_{\text{II}} &= \dot{\omega}_{18} - \dot{\omega}_{20} + \dot{\omega}_{28} \\ \dot{\omega}_{\text{III}} &= \dot{\omega}_5 + \dot{\omega}_{12} + \dot{\omega}_{15} + \dot{\omega}_{16} + \dot{\omega}_{17} + \dot{\omega}_{21} + \dot{\omega}_{22} + \dot{\omega}_{23} - \dot{\omega}_{32} + \dot{\omega}_{34} \\ \dot{\omega}_{\text{IV}} &= \dot{\omega}_1 + \dot{\omega}_6 + \dot{\omega}_9 - \dot{\omega}_{12} + \dot{\omega}_{13} - \dot{\omega}_{17} - \dot{\omega}_{20} + \dot{\omega}_{25} + \dot{\omega}_{27} + \dot{\omega}_{28} + \dot{\omega}_{37} \end{array} \right\} \quad (5.12)$$

where $\dot{\omega}_r$ is the reaction rate of the r^{th} chemical step (Kmol m⁻³s⁻¹, $r = 1, \dots, R$). The numbers appearing as subscripts on the rhs of equation (5.12) refer to the elementary steps of the detailed scheme presented in [76]; the data pertaining to the rate constants can also be found in this reference⁴. The net forward rate of the elementary reaction steps is given by the term in curly brackets appearing in equation (2.19), namely,

$$\dot{\omega}_r = k_{f,r} \varrho^{m_r} \prod_{\zeta=1}^N \left(\frac{Y_{\zeta}}{W_{\zeta}} \right)^{\nu'_{\zeta r}} - k_{b,r} \varrho^{l_r} \prod_{\zeta=1}^N \left(\frac{Y_{\zeta}}{W_{\zeta}} \right)^{\nu''_{\zeta r}} \quad (5.13)$$

Once these reaction rates are known, the stoichiometry of the mechanism, expressed in reaction steps Ia to IV, can be used to find the net formation rate of species α per unit volume ($\dot{S}_{\alpha}$ in Kg

³The *steady-state* approximation requires that the *net* formation rate of a species, as expressed in equation (2.19), to be zero. The *partial equilibrium* assumption requires the forward and reverse rates of a particular reaction step to be equal in magnitude (i.e. a dynamic equilibrium)

⁴A large proportion of this data enables the calculation of the forward rate constant, $k_{f,r}$. For those reaction steps where there is no information provided to evaluate $k_{b,r}$, appendix H shows how this may be obtained provided the equilibrium constant, $K_c(T)$, is known.

$\text{m}^{-3}\text{s}^{-1}$),

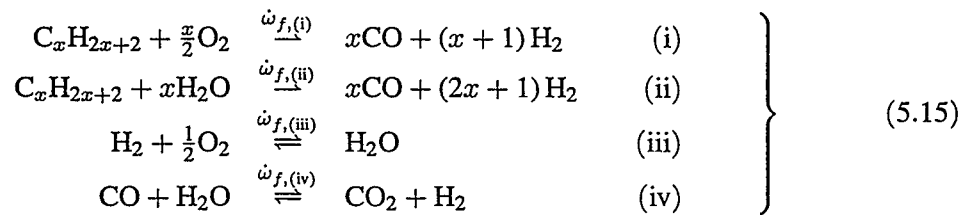
$$\left. \begin{aligned} \dot{S}_{\text{CH}_4} &= -W_{\text{CH}_4}\dot{\omega}_{\text{Ia}} \\ \dot{S}_{\text{CH}_3} &= W_{\text{CH}_3}(\dot{\omega}_{\text{Ia}} - \dot{\omega}_{\text{Ib}}) \\ \dot{S}_{\text{CO}} &= W_{\text{CO}}(\dot{\omega}_{\text{Ib}} - \dot{\omega}_{\text{II}}) \\ \dot{S}_{\text{H}} &= W_{\text{H}}(-\dot{\omega}_{\text{Ia}} - \dot{\omega}_{\text{Ib}} - 2\dot{\omega}_{\text{III}} + 2\dot{\omega}_{\text{IV}}) \\ \dot{S}_{\text{H}_2} &= W_{\text{H}_2}(\dot{\omega}_{\text{Ia}} - 3\dot{\omega}_{\text{Ib}} + \dot{\omega}_{\text{II}} + \dot{\omega}_{\text{III}} - 3\dot{\omega}_{\text{IV}}) \\ \dot{S}_{\text{H}_2\text{O}} &= W_{\text{H}_2\text{O}}(-\dot{\omega}_{\text{Ib}} - \dot{\omega}_{\text{II}} + 2\dot{\omega}_{\text{IV}}) \\ \dot{S}_{\text{O}_2} &= -W_{\text{O}_2}\dot{\omega}_{\text{IV}} \\ \dot{S}_{\text{CO}_2} &= W_{\text{CO}_2}\dot{\omega}_{\text{II}} \end{aligned} \right\} \quad (5.14)$$

Note that although the expressions written down in equation (5.14) have units of $\text{Kg m}^{-3}\text{s}^{-1}$, the quantities which are actually solved are in terms of the specific mole number, $n_\alpha (= \frac{Y_\alpha}{W_\alpha})$. This is a choice which is made out of convenience and the corresponding rates, $\dot{r}_\alpha (= \frac{\dot{S}_\alpha}{W_\alpha})$, have units of $\text{Kmol Kg}^{-1}\text{s}^{-1}$. Equation (5.12) indicates that the overall rates depend on the elementary rates which in turn require knowledge of the steady-state concentrations. These concentrations are obtained using explicit algebraic expressions. Their forms are not presented here but can be found in Chelliah et al. [10]. The investigators carried out predictions of an axisymmetric counterflow diffusion flame using the (parent) detailed scheme and the five-step reduced mechanism for a wide range of strain rates. Their results indicated that the discrepancies between the two were very small for the non-radical species ($\leq 3\%$), but larger for the radicals ($\leq 20\%$). It is not clear what effect the less accurate representation of the radical species field may have on the turbulent flame calculations.

5.3.1.2 The Global Alkane Hydrocarbon Mechanism of Jones and Lindstedt

While the systematic reduction of detailed hydrocarbon schemes lead to reduced mechanisms that reproduce many (laminar) flame characteristics accurately, the number of species to be considered is quite large. This makes their application to turbulent flames more difficult because of the short radical time scales incorporated into the description. These problems are particularly relevant to the transported pdf approach where the need to maintain the number of independent scalars to a minimum is essential. Furthermore, for many fuels *global* schemes currently provide a relatively simple and computationally efficient way of describing the laminar flame structures.

The task of constructing a global reaction scheme for hydrocarbon combustion may be simplified if the flame structure is viewed as comprising two reaction zones: a primary zone where the fuel is converted to CO and H_2 and a secondary zone where oxidation to H_2O and CO_2 occurs. The scheme proposed by Jones and Lindstedt [46] takes the following form,



The scheme is designed to work for alkanes up to butane in premixed and non-premixed laminar flames. The most significant feature is that radical species do not appear.

Reaction steps (iii) and (iv) appearing in equation (5.15) were derived from a linear combination of certain bimolecular chain reactions which were considered to be important to the flame structure⁵ [46]; however, the choice of which reactions to presume in partial equilibrium is not unique and leads to different proposals for $\dot{\omega}_{f,(iii)}$ and $\dot{\omega}_{f,(iv)}$. For example, it was observed that if $H + O_2 \rightleftharpoons OH + O$ was assumed to be in partial equilibrium, then the resulting rate expressions failed to accurately reproduce the H_2 concentrations in rich premixed flames and the fuel-rich region of diffusion flames. Relaxing the partial equilibrium assumption for this reaction led to acceptable results for the H_2 and CO concentrations in the fuel-rich regions, but their validity in the lean regions became doubtful. The actual forms proposed by Jones and Lindstedt are:

$$\dot{\omega}_{f,(iii)} = k_{f,(iii)} [H_2]^{\frac{1}{2}} [O_2]^{\frac{9}{4}} [H_2O]^{-1} \quad (5.16)$$

$$\dot{\omega}_{f,(iv)} = k_{f,(iv)} [CO] [H_2O] \quad (5.17)$$

Generally speaking, the construction of a rate expression should allow for acceptable numerical convergence properties. Consequently it is desirable to use only positive numbers as concentration exponents, and it is advantageous to use exponents greater than unity so that second-order methods (such as a Newton-Raphson technique) can be used to integrate the rate expressions. The rate expression presented in equation (5.16) does not satisfy these requirements in the concentration exponents of H_2 and H_2O ; as $[H_2O] \rightarrow 0$, $\dot{\omega}_{f,(iii)} \rightarrow \infty$ and as $[H_2] \rightarrow 0$, $\frac{\partial \dot{\omega}_{f,(iii)}}{\partial [H_2]} \rightarrow \infty$. To be able to use equation (5.16), artificial clipping of the rate is required for the concentrations of H_2 and/or H_2O below an arbitrarily chosen 'small' value. The manner in which this is done, together with the details of how the reverse rates are calculated are described in appendix H. Because of the above-mentioned (undesirable) features associated with equation (5.16), Jones and Lindstedt [46] proposed an alternative expression,

$$\dot{\omega}_{f,(iii)}^* = k_{f,(iii)}^* [H_2]^{\frac{1}{4}} [O_2]^{\frac{3}{2}} \quad (5.18)$$

although it is claimed that its use would result in reduced accuracy in fuel-lean regions. Equation (5.18) still does not overcome the problem of $\frac{\partial \dot{\omega}_{f,(iii)}}{\partial [H_2]} \rightarrow \infty$ as $[H_2] \rightarrow 0$.

The initial estimates for activation energies were obtained from the partial equilibrium analysis used to construct the rate expressions, however, further optimisation was required so that the scheme performed as robustly as possible, i.e. that it was capable of reproducing acceptable H_2 and CO concentrations as well as burning velocities in various laminar flames. Specifically, detailed measurements of the scalar field in a freely propagating plane premixed flame and a confined laminar jet diffusion flame were employed to evaluate the reduced scheme. The values of the constants required to construct $k_{f,(i)} \rightarrow k_{f,(iv)}$ are presented in table 5.1.

The two competing fuel breakdown reactions (steps (i) and (ii)) broadly determine the shape of the primary reaction zone. It was observed [46] that recourse to detailed hydrocarbon mechanisms and the use of systematic reduction were not useful, primarily because of the requirement to exclude radical species from the reduced scheme; studies in chemical kinetics suggest that the initial attack on the fuel is made predominantly by OH and H radicals. Jones and Lindstedt [46] point out that whilst earlier investigations specified reaction step (i), in equation (5.15), step (ii) should also be used in order to reproduce measurements in both premixed and diffusion flames. The rate constants (table 5.1) and rate expressions were specified so that the measurements in the

⁵The importance of a particular reaction step is determined by examining the effects on the flame structure of including and excluding it from the detailed hydrocarbon scheme.

Reaction step	Rate constant	x	B	a	E
(iii)	$k_{f(iii)}$	1	2.5×10^{16}	-1	40,000
		3	3.0×10^{16}	-1	40,000
	$k_{f(iii)}^*$	1	0.68×10^{16}	-1	40,000
		3	0.85×10^{16}	-1	40,000
(iv)	$k_{f(iv)}$	$1 \rightarrow 4$	2.75×10^9	0	20,000
(ii)	$k_{f(ii)}$	$1 \rightarrow 4$	0.3×10^9	0	30,000
(i)	$k_{f(i)}$	1	0.44×10^{12}	0	30,000
		3	0.40×10^{12}	0	30,000

Table 5.1: Data required to construct the forward rate constants for the global reaction scheme of Jones and Lindstedt [46]. x is the stoichiometric coefficient of the alkane hydrocarbon, C_xH_{2x+2} . B , a and E are defined through equation (2.18). Units employed: Kg, m, s, Kmol, Kcal, K.

previously described experimental configurations were accurately reproduced. The form of the rate expressions are:

$$\dot{\omega}_{f,(i)} = k_{f(i)} [C_xH_{2x+2}]^{\frac{1}{2}} [O_2]^{\frac{5}{4}} \quad (5.19)$$

$$\dot{\omega}_{f,(ii)} = k_{f(ii)} [C_xH_{2x+2}] [H_2O] \quad (5.20)$$

The rate equations describing the evolution of the individual species can then be obtained in the same manner as for equation (5.14).

5.3.1.3 Tabulation of the Thermochemistry

The reduced chemistry schemes described in the previous section involve the solution of a stiff and coupled set of equations. By employing an approximate factorisation technique, the chemical reaction can be treated separately through a set of stiff ODE's. The ODE's were solved using LSODE⁶, a suite of programs designed to solve initial value problems involving stiff and non-stiff systems of first-order ODE's [38]. This widely-used (public domain) package was employed to solve the ODE's arising out of the chemical kinetics. Two basic multi-step methods are available in this solver; an implicit Adams method and one based on backward differentiation formulae. Any further description of the solver would be beyond the scope of this work and the interested reader should consult the reference cited above.

In principle all particles are subjected to chemical reaction; however, in practice only those particles having a mixture fraction lying between (typically) $0 \leq \xi \leq 0.3$ are likely to show any significant change in their properties due to reaction. It is important to reduce the dimensionality of the problem as much as possible. For this purpose two conditions can be employed:

- Equal diffusivity of the scalars⁷. The *element* mass fractions, and hence their element

⁶An abbreviation for: Livermore solver for ordinary differential equations

⁷It is important to bear in mind that equal diffusivity is *not* an assumption that is invoked simply to reduce the dimensionality of the scalar pdf. This assumption has a far more fundamental justification, relating entirely to the modelling of the unclosed terms discussed in the earlier chapters.

specific mole numbers remain constant during chemical reaction, so that the carbon-to-hydrogen ($\frac{n_C}{n_H}$) and the oxygen-to-nitrogen ($\frac{n_O}{n_N}$) ratios do not change; this constitutes a physical requirement. The assumption is that these ratios (also) do not change under the influence of transport. More specifically, for a two-stream mixing process with fuel (C_xH_y) and air initially separate, $\frac{n_C}{n_H} = \frac{x}{y}$ (where $y = 2x + 2$ for alkanes). On a volumetric basis, air typically consists of 21% O_2 and 79% N_2 , so that $\frac{n_O}{n_N} \equiv \psi \approx 0.265^\dagger$. The implication of these relations is that any *two* scalars participating in the reaction can be eliminated (i.e. they can be expressed in terms of the remaining scalars).

- Mass conservation can also be invoked ($\sum_\alpha Y_\alpha = \sum_\alpha n_\alpha W_\alpha = 1$) so that an additional scalar can be eliminated from the pool of independent scalars characterising the pdf.

Based on the reduced mechanisms that are employed, the participating species are C_xH_y , O_2 , CO_2 , H_2O , CO , H_2 and N_2 , and for the systematically reduced mechanism of Chelliah et al. [10] (for CH_4 only) the radicals H and CH_3 are also present. For the global hydrocarbon scheme this implies that four out of the seven species present will have to be specified independently. For the systematically reduced CH_4 mechanism, six out of nine species will have to be specified. The choice of the independent scalars is not unique, nor is it arbitrary. The mixture fraction is clearly a sensible choice since it is a fundamental quantity related to the degree of mixing that has taken place. Knowing ξ fixes n_{N_2} (as shown in appendix F) and so the latter can no longer be treated as an independent scalar. The analysis of appendix F also demonstrates that if $n_{C_xH_y}$ and n_{CO} are chosen as independent scalars, then within the constraints of the problem, n_{CO_2} is no longer an independent scalar since it is known in terms of ξ , $n_{C_xH_y}$ and n_{CO} . The following choice of independent scalars is made:

- Global hydrocarbon scheme: ξ , $n_{C_xH_y}$, n_{CO} and n_{H_2} .
- Systematically reduced mechanism: ξ , n_{CH_4} , n_{CH_3} , n_{CO} , n_H , n_{H_2}

The concentrations of CO_2 , H_2O and O_2 can be evaluated once the values of the independent scalars have been specified. In equation (5.14), the rate equations have been written for *all* the reactive species (i.e. including the rates of CO_2 , H_2O and O_2 , which could be calculated from the independent *reactive* species rates). This measure is not necessary in so far as the mathematical treatment of the problem is concerned. However, numerically, LSODE calculates the Jacobian of the reaction rate matrix in order to integrate the stiff ODE's, and it is felt that by calculating for all the species, the rate expressions would have a less complex *analytical* form. Consider for example the global hydrocarbon scheme,

$$\dot{r}_{C_xH_{2x+2}} = -\frac{1}{\theta} \left(\dot{\omega}_{f,(i)} + \dot{\omega}_{f,(ii)} \right) \quad (5.21)$$

Equations (5.19) and (5.20) indicate that to evaluate $\dot{\omega}_{f,(i)}$ and $\dot{\omega}_{f,(ii)}$, $[O_2](= \varrho n_{O_2})$ and $[H_2O]$ are required. These concentrations could be replaced by the appropriate expressions which are functions of ξ , $n_{C_xH_y}$, n_{CO} and n_{H_2} . However, it is clear that in doing this the form of equation (5.21) is analytically more complicated, and so the calculation of the Jacobian could be more sensitive to small perturbations arising out of the numerical treatment. If the Jacobian was

[†] A volume fraction can be interpreted as a mole fraction (X_α), and the latter is related to the specific mole number by, $X_\alpha = n_\alpha / \sum_\beta n_\beta$. Thus the ratio of the volume fractions of O_2 and N_2 is equal to $\frac{n_O}{n_N}$.

calculated exactly there would be no difference in solving for six rate equations or three. In practice it was found that the two different approaches brought about no observable effect in the homogeneous turbulence calculations and the laminar flames that were investigated.

The description of the thermochemistry is now complete and can be summarised as follows:

- After the application of the transport and mixing operations in the Monte Carlo simulation, the operation of chemical reaction is applied to the particle properties.
- Under adiabatic and constant pressure conditions, the enthalpy per unit mass of mixture is unchanged during chemical reaction. Knowledge of ξ permits the calculation of the *unburnt* composition at 298K; hence h can be calculated from equation (2.8).
- Given p , h and n_α , the temperature of the reacting mixture can be calculated, the details of which can be found in appendix H. The equation of state for an ideal gas can subsequently be employed to obtain the density.
- The temperature of the mixture is then used to find the equilibrium constant $K_c(T)$, and together with ρ and n_α , the forward and reverse rates are constructed. The reaction rates of the reactive species can then be evaluated and subsequently integrated over a fixed time interval.
- The change in the value of n_α as a result of the integration advances the particle properties to $x + \delta x$. The ensemble of particle properties then approximate $\tilde{P}(\underline{\psi}; x + \delta x, \omega)$.
- The mean density is obtained from the ensemble average of the particle densities at a given point⁸. This is then used in the velocity field finite-difference procedure to advance $\tilde{u}$, k and ε , and then the whole operation is repeated.

The process of chemical reaction is by far the most time-consuming part of the calculation procedure; it can take orders of magnitude longer than the transport and mixing operations. Therefore the thermochemistry needs to be tabulated in order to avoid integrating the large number of stiff ODE's. The steps previously described for updating the scalar properties, *due to the sole effect of chemical reaction*, are performed on an 'once and for all' basis, and the results are stored in a 'look-up' table. In order to use such a table the chemical rate equations have to be solved for all the possible composition states. To characterise these composition states, the *bounds* for the independent scalars need to be determined. The details of how the bounds are determined for the four and six scalar reduced chemistry are discussed in appendix F. The manner in which the look-up table is generated through the hierarchy of the bounds (i.e. how ξ fixes the bounds on $C_x H_y$, and how ξ and $C_x H_y$ fix the bounds on CO, etc...) serves to locate the *initial* composition state of any notional particle in the multi-dimensional composition space. This means that at any scalar location in the look-up table, only the *change* in the scalar properties over pre-defined time steps (typically 10^{-5} s and 10^{-4} s) relative to the *initial* state need to be stored. The only exception to this is the density; for a given composition state, the density of the mixture *prior* to the effect of chemical reaction is stored. The way in which the bounds are linearly affected by one another indicates that the scalar hypervolume, which constitutes the look-up table, has a complicated shape. For any particular composition state associated with a given particle, its

⁸The method by which the scalar statistics of the flowfield are obtained from the particle properties is described in appendix E.

nearest neighbours in the look-up table act as nodes with which a multi-linear interpolation[‡] can be performed to advance the particle state over a time step $\delta t_i = \frac{\Delta x}{u_i}$ §.

In order to construct the look-up table the scalar bounds are discretised into an array of dimension (25, 20, 9, 9) for the global scheme, and (15, 11⁵) for the systematically reduced mechanism. The grid point distributions in the array are uniform except in the ξ -direction which has more grids placed locally around the stoichiometric point. In order to ensure a good resolution of the chemical changes, the tabulation is performed for $0 \leq \xi \leq 0.3$; the density must be tabulated over the entire mixture fraction range so that the density can be evaluated at all points in the flowfield, not just those near the stoichiometric condition. Consequently *two* tables are constructed; one which tabulates the chemistry and the other for the density. These are referred to as the reaction (interpolation) table and the density table. The reaction table contains the changes of the independent *reactive* scalars. For the global scheme the changes in the values of three independent reactive scalars are stored over time intervals of 10^{-5} s and 10^{-4} s. During the Monte Carlo simulations, if $\delta t_i \geq 10^{-4}$ s, the scalar changes associated with the latter time scale are employed for the multi-linear interpolations, until the time elapsed exceeds $(\delta t_i - 10^{-4})$ s, in which case the data stored for the shorter time scale is used to advance the particle properties. The entries in the table are stored as unformatted (binary) data and a 1.3Mb table was formed for a domain of dimension (25, 20, 9, 9). The systematically reduced mechanism involves more independent reactive scalars; for the reaction table memory limitations constrain the changes in the independent reactive scalar properties to be stored for only one time level, namely 10^{-5} s. Even then, the size of this table is nearly 68Mb, and because of this, the density table is dimensioned to a somewhat smaller size (15, 8⁵) resulting in a 6Mb table. Thus, in total, 74Mb is reserved for the interpolation tables which is quite a significant memory requirement.

Finally, the following point related to the construction of the reaction tables requires consideration. Whilst discussing equation (5.21) it was pointed out that *all* the reaction rates of the participating species are calculated instead of just those for the independent reactive scalars. This implies that after performing the integration of the ODE's, updated values for all the scalars are available; this raises the following questions:

- Does the change brought about by chemical reaction ensure boundedness[¶] of the particle properties ?
- How do the values of n_{CO_2} , n_{O_2} and $n_{\text{H}_2\text{O}}$ (after the integration) compare with their counterparts calculated in terms of the independent scalars?

The chemical schemes ensure that mass is conserved (i.e. $\sum_{\alpha} \dot{S}_{\alpha} = 0$). Furthermore, the chemistry cannot affect the ratios of elemental ($\frac{n_{\text{C}}}{n_{\text{H}}}$) and ($\frac{n_{\text{O}}}{n_{\text{N}}}$). Since the initial amounts of ($\frac{n_{\text{C}}}{n_{\text{H}}}$) and ($\frac{n_{\text{O}}}{n_{\text{N}}}$) are set by the fuel and air, this means that the evolution of the scalars according to the chemical mechanism satisfy the same conditions used for the construction of the bounds.

[‡]The algorithm for this interpolation procedure is described in detail in the work of Chen et al. [15]; the method is essentially an efficient way of performing linear interpolation for a point located in multi-dimensional space. Because an interpolation technique is used, the look-up table is also referred to as an interpolation table.

[§]The time step δt_i is the physical time scale set by the thermofluid dynamics; however in the look-up table, the changes in the scalar properties have only been stored for pre-defined, arbitrary time steps $\approx 10^{-5}$ s and 10^{-4} s. Therefore, in order to perform the chemical reaction operation over a time step δt_i may well require several interpolations. For each new interpolation, the nearest neighbours to the evolving composition state need to be re-assigned.

[¶]By 'boundedness' it is meant that the scalar concentrations lie within the bounds specified in appendix F.

This demonstrates that the reaction scheme ensures boundedness, and the results for n_{CO_2} , n_{O_2} and $n_{\text{H}_2\text{O}}$ are the same regardless of whether LSODE or the concentrations of the independent scalars are employed. However, the ODE's arising from the chemical kinetics cannot be integrated exactly, simply because a numerical scheme is employed for this purpose. Consequently, if the boundedness of the results *after* integration is not verified, the effect of cumulative round-off could perturb the solution; this perturbation can be important in systems of stiff ODE's. To avoid such an occurrence, the magnitudes of the independent scalars should be verified to ensure that boundedness is satisfied *before* storing the relevant information in the look-up table. Extensive tests have shown that whilst such corrections to the scalar concentrations are necessary, the magnitude of these changes is typically about 0.01% mole fraction in the major species and intermediates (CO and H₂).

Chapter 6

Results and Discussion

6.1 Introduction

The main objective of this thesis is the application of the transported pdf approach to combusting, inhomogeneous turbulent flows. However, results from the calculations of homogeneous, isotropic turbulence and laminar jet diffusion flames are also presented because they highlight important features of the transported pdf closure. The reduced chemical schemes discussed in section 5.3.1 have been previously tested by their originators in laminar flame calculations; in developing these mechanisms the molecular transport of the scalar field was treated without making recourse to assumptions of equal diffusivity. The present contribution does not try to re-perform those calculations; instead a laminar diffusion flame, which was also considered for the evaluation of the scheme of Jones and Lindstedt [46] (the global hydrocarbon scheme), was investigated subject to the assumption of equal diffusivities. The effect of this assumption on the predictions of the laminar flame is compared with the results of Jones and Lindstedt.

It was mentioned in the section 5.3.1.3 that the chemistry needs to be tabulated in order to ensure manageable computer run-times; the excessive cpu requirements are related to the time required for the integration of the stiff ODE's associated with the properties of the *whole ensemble* of particles *at many node points*. Laminar flame calculations constitute a very practical way of testing the accuracy of the interpolation technique since both the 'look-up' table and the direct integration (using LSODE) can be used. Another situation where the tabulation of the chemistry is not necessarily required is in the calculation of homogeneous, isotropic and reacting turbulence. Unlike the laminar flame calculations, a transported pdf equation is solved, but because of the assumed statistical homogeneity[†] only one node (in physical space) is considered, whereas in the inhomogeneous flow calculations particles have to be placed at all nodes covering the region of interest. Whilst the results are only of qualitative value, the investigation of homogeneous, isotropic turbulence is important because the behaviour of the mixing models can be evaluated. The evolution of the scalar statistics demonstrates interesting properties when subjected to different initial conditions and with chemical reaction present, the coupling of mixing and reaction can be examined.

The transported pdf approach was tested in a turbulent jet diffusion flame configuration, and evaluated using detailed measurements of the velocity and scalar field from four experimental flames. More complicated flows have not been investigated. The transported pdf technique was

[†]In this thesis, statistical homogeneity implies that the scalar pdf does not change with position.

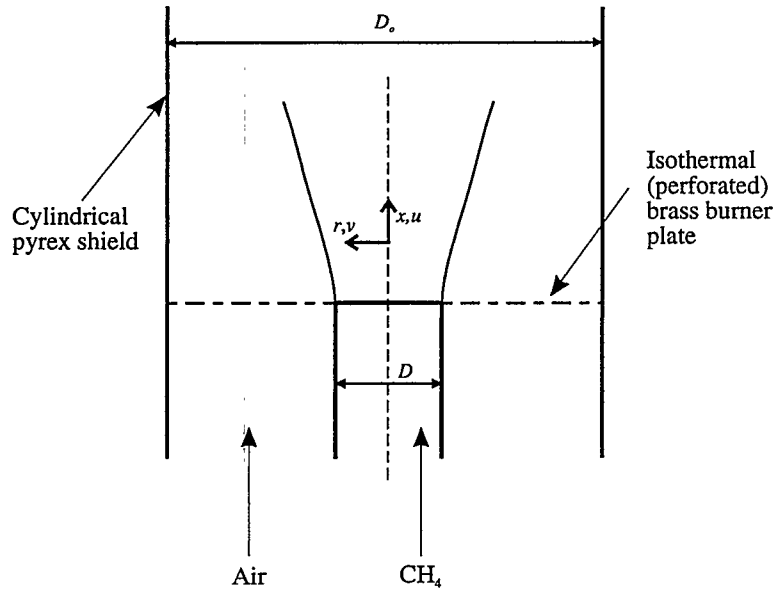


Figure 6.1: Schematic of the confined, axisymmetric, laminar CH_4 -air jet diffusion flame investigated by Mitchell et al. [69]. $D = 1.27\text{cm}$ and $D_o = 5.08\text{cm}$. Air and fuel are admitted at 187.7 cc/s and 5.7 cc/s respectively.

applied using two different mixing models, namely LMSE and a coalescence-dispersion closure; with the latter model, the calculations were performed with $\mathcal{A}(x) = 1^\dagger$ and referred to as modified Curl.

6.2 Predictions of a Confined Laminar Jet Diffusion Flame

The configuration under investigation is illustrated in figure 6.1 and was experimentally investigated by Mitchell et al. [69]

A vertical, cylindrical burner was used to produce a confined, steady and atmospheric diffusion flame. The burner consisted of two concentric tubes of radii 0.635cm and 2.54cm , connected to a perforated solid brass disc which served as the burner plate. Fuel issued through the inner tube and air through the outer. A Pyrex glass cylinder shielded the flame. Sampling probes leading to gas analysers were employed for CO , CO_2 , CH_4 , H_2 , N_2 and O_2 . Water analyses were made by determining the dew point[§] of the sample gas. Mitchell et al. did not discuss the accuracy of this particular measuring technique. Temperature measurements were made using fine-wire thermocouples and velocities using LDV. The laminar diffusion flame was supplied with methane at $5.7\text{ cm}^3\text{ s}^{-1}$ and air at $187.7\text{ cm}^3\text{ s}^{-1}$. The luminous edge of the steady flame closed at the burner axis 5.8 cm above the burner plate. At each sample location, three samples were taken; generally, the agreement between the three sets of measurements of the concentration field was reported to be within 5%.

The flame was simulated by solving the species transport equations (2.16) for the independent scalar variables pertaining to the reduced mechanisms. For the global hydrocarbon scheme these

[†]The significance of $\mathcal{A}(x)$ is described in section 4.7.2.

[§]The dew point of a gas-vapour mixture is the temperature at which the vapour condenses (or solidifies) when it is cooled at constant pressure.

are ξ , n_{CH_4} , n_{CO} and n_{H_2} ; for the systematically reduced scheme, n_{CH_3} and n_{H} are also included. The finite-difference procedure described in appendix D was employed for solving the species transport equation, *with the source terms for reaction excluded*. The effect of reaction was then 'added' to the scalar properties through the use of the look-up tables or by direct integration. This approach is consistent with the operator splitting technique which is also used in the Monte Carlo simulation of the pdf transport equation. The laminar flame formulation used here is different from that employed by Jones and Lindstedt [46]. In their work a fully implicit method was employed and the species transport equation (including the source term for chemical reaction) was solved using an iterative Newton-Raphson technique. In the present study iterative methods are not used, and to ensure accuracy of solution, fine meshes (up to 500 grid nodes across the jet) were used so that the reaction zone would be well resolved. The forward step size was controlled through the definition of a diffusion number, similar in principle to the Courant number (which is a measure of how much convection occurs in a given direction over a time step). The diffusion number, $\mathcal{F}$, is defined as:

$$\mathcal{F} \equiv \left(\frac{\nu_i}{Sc} \frac{1}{\delta y_i} \right) \left(\frac{\delta x}{u_i} \right) \frac{1}{\delta y_i} \quad (6.1)$$

where i identifies a grid node ($i = 1, \dots, N+1$), $Sc \approx 0.7$ and $\nu_i = \frac{\mu_i}{\rho_i}$. The mixture viscosity was estimated using Wilke's semi-empirical formula (taken from Bird et al. [7]):

$$\mu_i = \sum_{\alpha=1}^N \frac{X_{\alpha} \mu_{\alpha}}{\sum_{\beta=1}^N X_{\beta} \Theta_{\alpha\beta}} \quad (6.2)$$

where $\mu_{\alpha} = \mu_{\alpha}(T, p)$ was obtained from Svehla's compilation [90]. Linear interpolation was employed to find μ_{α} at the temperatures that were not specifically tabulated. X_{α} is the mole fraction of species α ; $X_{\alpha} = n_{\alpha} / (\sum_{\gamma=1}^N n_{\gamma})$ and,

$$\Theta_{\alpha\beta} = \frac{1}{\sqrt{8}} \left(1 + \frac{W_{\alpha}}{W_{\beta}} \right)^{-\frac{1}{2}} \left[1 + \left(\frac{\mu_{\alpha}}{\mu_{\beta}} \right)^{\frac{1}{2}} \left(\frac{W_{\beta}}{W_{\alpha}} \right)^{\frac{1}{4}} \right]^2 \quad (6.3)$$

$\mathcal{F}$ is proportional to the ratio of the distance a *fluid* particle will diffuse in one 'time' step to the cross-stream mesh spacing. Typical maximum values of $\mathcal{F}$ employed were 1, 0.1 and 0.01. The largest differences in the species concentrations brought about by these various specifications amounted to less than about 2% - i.e. the results were largely unaffected by this parameter provided $\mathcal{F} \leq 1$.

The treatment of the initial conditions in the calculation procedure deserves further mention. Jones and Lindstedt [46] have pointed out that there is uncertainty in the precise boundary conditions to be applied at the burner inlet, such that if conditions similar to those found in the computations of Mitchell et al. [69] are used, erroneous velocity and mixture fraction profiles appear at the first measuring station ($x = 12\text{mm}$). It was also observed that this result was independent of the type of chemistry, mesh refinement or numerical technique (i.e. parabolic or elliptic formulation) that was employed. Consequently, the procedure adopted by Jones and Lindstedt was employed; the computation was started at the first measuring station using the measurements as the initial condition. Figures 6.4 and 6.5 demonstrate how the initial condition

was specified. The mixture fraction was calculated using the measurements of the carbon-containing species such that,

$$\xi \equiv \frac{n_C}{n_{C,1}} = (n_{CO_2} + n_{CO} + n_{CH_4}) W_{CH_4} \quad (6.4)$$

The advantage of using this specification is that it avoids the use of the water concentration, since the accuracy of its measurement is questionable. The solid and dashed lines shown in figures 6.4 and 6.5 are mere curve fits. With the thermochemistry employed in this thesis, the initial specification of CO_2 , H_2O , N_2 and O_2 depends entirely on ξ , n_{CH_4} , n_{CO} and n_{H_2} (in addition to n_{CH_3} and n_H for the mechanism of Chelliah et al. [10]). Consequently it is not possible to simply curve fit all the data that is available; such a specification would not be consistent with the assumption of constant C/H and O/N ratios. Initially the main objective was to fit the measurements of the independent scalars as closely as possible (hence the dashed lines in figures 6.4 and 6.5). However, this led to a poor representation of the CO_2 , O_2 and temperature fields near the centre-line.

The 'overall optimized curve-fit'¹ (solid line) indicates a much improved initial specification of the CO_2 and O_2 field and at the (slight) detriment of the CH_4 , CO and H_2 profiles. The improved specifications were obtained by reducing the CH_4 concentration near the centre-line. In doing this, a pool of carbon and hydrogen atoms are made available. The carbon atoms are (mainly) taken up by the oxygen atoms to form CO and CO_2 ; in the case of the global hydrocarbon scheme the carbon atoms cannot combine with anything else. This procedure goes some way towards removing the high O_2 concentration near the centre-line. By limiting the initial amount of CO , more CO_2 can be formed so as to counteract the drop in CO_2 which is registered by the dashed line in figure 6.4. The pool of hydrogen atoms is taken up by H_2 and H_2O ; this is confirmed in figure 6.5. Since it is the peak levels of H_2 and CO that are of interest, their concentrations are not allowed to change substantially. This is why the increase in the levels of H_2 and CO is less manifest than that of H_2O and CO_2 . As a consequence of the increase in H_2O and CO_2 near the centre-line, the temperature of the mixture locally increases. Whilst the use of these two different specifications led to small differences in the subsequent evolution of the jet flame, the *peak* levels of the species concentrations were hardly affected, and it is this feature of the predictions that is of interest to this discussion. To reproduce laminar flame measurements as well as possible was *not* the aim of the current calculations; this was performed when the reduced schemes were being developed. In this study the aim was to investigate the sensitivity of the predicted peak scalar concentrations to the assumption of equal diffusivity of the scalars.

In principle the extra carbon and hydrogen atoms made available by the reduction in the CH_4 concentration could be used to specify initial values of hydrogen and methyl radicals (H and CH_3 respectively) to be used in conjunction with the systematically reduced mechanism. This constitutes another source of uncertainty in the representation of the initial conditions. Measurements of H and CH_3 were not made in the work of Mitchell et al. [69], and in fact such data is rare in most of the combustion literature. The following (arbitrary) specification was therefore adopted: the initial concentrations of H and CH_3 were taken either as zero, or the largest possible values were employed (subject to the constraints imposed by the bounds that are derived in appendix F). Figure 6.6 shows the resulting initial conditions and this clearly demonstrates that

¹The optimization was performed by inspection, i.e. selecting the profiles which appeared to fit the initial conditions as well as possible.

in specifying non-zero initial values of H and CH₃, fewer carbon and hydrogen atoms are available for combining with the oxygen atoms; the consequences of this are that the initial O₂ concentration near the centre-line increases and that of H₂O correspondingly drops. Figure 6.7 shows profiles of N₂, CH₃ and H at the two measuring stations downstream of $x = 12\text{mm}$. Clearly the effect of the two different initial specifications for the radical species appears to be small; the effect on the remainder of the species concentrations (figures not presented) is hardly distinguishable. This is a potentially fortunate result in that the precise details of the initial radical levels (which are not known) do not affect the solution further downstream. However, this does not mean that the levels of H and CH₃ are unimportant in the evolution of the flow. The results could well imply that the levels of the other species play an important role in determining the concentration of the radicals so that after a few steps of the calculation the chemistry ensures that the radical species concentrations are much the same regardless of the initial conditions.

Figures 6.8-6.11 show the results obtained with the two reduced mechanisms, and without the use of the interpolation tables. The most prominent feature of the results is the very substantial overprediction of the intermediate species, CO and H₂. This is necessarily coupled to an underprediction of H₂O and CO₂. However, in contrast to the H₂O and CO₂ predictions, the *peak* levels of CO and H₂ are very poorly reproduced. The most significant difference between the results of the calculations reported here and those of Jones and Lindstedt [46] relates to the treatment of the diffusivity. Clearly the effect of the assumption is quite considerable; such effects have also been observed in relation to the H₂ concentration when using detailed kinetic mechanisms, Lindstedt [62]. In this situation the effect on the CO field is less pronounced, presumably due to the additional paths available for CO consumption. These results could therefore suggest that reduced schemes may show a greater sensitivity to the assumption of equal diffusivity than the detailed mechanisms. The fact that the peak levels of CO and H₂ are lower for the systematically reduced CH₄-air mechanism than for the global hydrocarbon mechanism appears to be related to the less accurate representation of the fuel burn-up using the former mechanism. The reason why the consumption of CH₄ is predicted differently by the two mechanisms is not entirely clear. With the global scheme, the hydrocarbon is consumed by O₂ and H₂O to form CO and H₂ - the path involving O₂ is a particularly direct and robust way in which the break-down of CH₄ can be ensured. However, realistically CH₄ is initially attacked by radical species, such as H; it seems plausible to suggest that given the very high diffusivity of H, the consumption of CH₄ is more sensitive with the use of the systematically reduced mechanism than with the global scheme (when invoking the equal diffusivity assumption).

The temperature profiles (figure 6.11) show that the peak values are quite well reproduced but at $x = 24\text{mm}$ the predicted reaction zone appears to be to the left of the measurements; this is consistent with the CH₄, O₂, H₂O and CO₂ profiles. Mitchell et al. [69] point out that in relation to *their* predictions (which included only major species, no radicals and no CO or H₂), the flame temperature was overpredicted partly due to the uncertainty in the radiation corrections applied to the thermocouple whenever carbon particles settled on the bead, but also due to the neglect of energy-absorbing pyrolysis reactions. These reactions manifested themselves as a luminous core situated on the rich side of the primary reaction zone. The luminosity of the flame was associated with the light emitted from the incandescent carbon particles formed by pyrolysis of the CH₄ in the fuel-rich regions of the flame. The reaction zone, which according to experimental observation was typically 2mm thick, had a blue hue, the intensity of which decreased with increasing height above the burner plate. The blue hue of the reaction zone was associated with the burnout of CO.

In figure 6.8 the measurements indicate that the CO level drops from $x = 24\text{mm}$ to $x = 50\text{mm}$ (hence the observed decrease in the intensity of the blue hue). However, the predictions follow the opposite trend, because the CH_4 has not been completely consumed. Despite the differences near the centre-line between the temperature profiles (and hence in the density field) using the two reduced mechanisms, the effect on the mixture fraction evolution can hardly be detected.

Another interesting feature of the results (figure 6.10) is the relative peak levels of CH_3 and H radicals. The work of Chelliah et al. [10] indicates that the peak levels of CH_3 are typically an order of magnitude greater than those of H, whilst the predictions show the converse effect. Despite the substantial quantitative difference, the shapes of the predicted profiles compare favourably with the results of Chelliah et al.

Figure 6.9 indicates that finite levels of O_2 are measured near the centre-line. Mitchell et al. [69] claimed that this can be attributed to the O_2 which penetrates the flame surface near the base, since in this region it is argued that the temperature and reaction intensity are too low to consume all the oxygen in the primary reaction zone before it can diffuse towards the centre-line. The predictions also indicate finite O_2 levels near the centre-line partly because some O_2 is already specified to exist in that region through the initial condition (figure 6.5).

Comparison of the velocity field predictions (figure 6.11) with those of Mitchell et al. [69] (where an elliptic formulation was employed) and Jones and Lindstedt [46] shows that the differences are quite small, indicating that the current predictions are not in error due to a poor representation of the velocity field. The measurements of velocity at $x = 50\text{mm}$ imply that at $r = 1.5\text{cm}$ the velocity is zero; the wall boundary appears at $r = 2.54\text{cm}$ (refer to schematic, figure 6.1). None of the predictions from the various sources cited suggest that the velocity should be zero in this region. Mitchell et al. point out that the gases along the axis of the flow are accelerated by the prevailing buoyancy forces which give rise to a large recirculation cell. However, the results of their work suggest that this recirculation cell occurs in regions outside the areas where the measurements are made.

Figures 6.12-6.15 show the same calculations discussed above, only this time the interpolation is employed. The agreement between the interpolated and integrated result for the global hydrocarbon scheme, figure 6.14, is very encouraging. The predictions for the velocity and temperature profiles are only shown since they indicate the largest differences. Generally speaking, the similarity of the results using the interpolation table and LSODE for the mechanism of Chelliah et al. is also encouraging, except in the representation of the CH_3 and H profiles (figure 6.13) - this may explain to a certain extent the shortcomings of the interpolated results for the other properties using this mechanism. It is difficult to determine whether the discrepancy between the 'exact' result (represented by the use of LSODE) and the interpolated one is due to the coarseness of the table. This is because in order to obtain a substantially improved result would require a considerable refinement in the size of the current table; currently the look-up table for chemical reaction is about 68Mb which is already a substantial amount of memory. Another, potentially more plausible, reason for the discrepancy may be the manner in which the table is constructed. In the way the six-dimensional table is currently constructed the bounds for CH_3 and H require arbitrary clipping. This is done so that radical species with unrealistically large concentrations are not tabulated, since they will only serve to use up space in the composition space which could be replaced by physically more plausible (and tangible) entries. Furthermore, the evolution of the composition states with very high radical concentrations subject to equation (5.14) is another area of uncertainty. The steady-state approximations which are frequently invoked to construct reduced

schemes are valid in the high temperature reaction zone - the applicability of the reduced scheme to composition states where the temperature and/or mixture strength are substantially different from those found in the reaction zone of a laminar flame is therefore questionable.

6.3 The Calculations of Statistically Homogeneous, Isotropic Turbulence

6.3.1 Non-Reacting Test Cases

Figures 6.16 and 6.17 show the result of tests using modified Curl and LMSE for a single non-reactive, bounded scalar (ξ , the mixture fraction). Results are shown using two different turbulent time scales; one where $\tau (\equiv \frac{k}{\varepsilon})$ is held fixed at an arbitrarily selected value, τ_o , and another where the modelled ε -equation together with the k -equation are employed to characterise the evolution of τ (this latter procedure is described in appendix E). The choice of τ_o is arbitrary since the results are only of qualitative significance. In this respect the evolution of the moments with constant τ are *qualitatively* the same as those with a variable τ with the time axis appropriately shifted.

The mean of the scalar is unaffected, and through the appropriate choice of the constants appearing in the mixing models, the variance decay rate consistent with equation (4.6) is obtained. For τ constant, equation (4.6) can be integrated (using straightforward separation of variables) subject to the condition that at $t = 0$, $\langle \phi'^2 \rangle = \langle \phi'^2 \rangle_o$. The solution is,

$$\langle \phi'^2 \rangle = \langle \phi'^2 \rangle_o \exp \left(-\frac{C_D}{\tau_o} t \right) \quad (6.5)$$

When $\tau = \tau_o + (C_{\varepsilon_2} - 1) t$, integration of equation (4.6) yields:

$$\langle \phi'^2 \rangle = \langle \phi'^2 \rangle_o \left[1 + \frac{(C_{\varepsilon_2} - 1)}{\tau_o} t \right]^{-\frac{C_D}{(C_{\varepsilon_2} - 1)}} \quad (6.6)$$

Equations (6.5) and (6.6) are represented by the crosses, labelled as 'Analytical expression' in figures 6.16 and 6.17. Equation (4.7) defines the higher order moments also presented in the same figures. The skewness remains *constant* - this result can be demonstrated analytically for coalescence-dispersion and LMSE closures regardless of the initial form of the pdf [23, 13]. The reason why the skewness is *zero* in figures 6.16 and 6.17 is because the initial pdf's are symmetric about the mean, so that $\langle \phi'^3 \rangle = 0$, hence from equation (4.7), $\Omega_3 = 0$. If the initial distribution is not symmetric, $\langle \phi'^3 \rangle$ will be finite and at large times ($t \rightarrow \infty$), $\langle \phi'^2 \rangle^{\frac{3}{2}}$ and $\langle \phi'^3 \rangle$ will tend to zero *at the same rate*, so that Ω_3 remains constant (though not necessarily zero).

Figure 6.16 exemplifies the behaviour referred to in section 4.7.2; as the mixing progresses and the variance decays, the flatness and superskewness rise monotonically, so that as $t \rightarrow \infty$ then $\Omega_3, \Omega_4 \rightarrow \infty$. In contrast, LMSE predicts uniform values for these quantities as a function of time. The reason for this difference in behaviour, and for the presence of 'wiggles' which appear most prominently in the test case where $\tau = \tau_o$, is elucidated in figures 6.18-6.21. Here the terms in the denominator and numerator of equation (4.7) for Ω_3, Ω_4 and Ω_6 are presented. It is clear that the uniformity of Ω_4 using LMSE (figure 6.19) is due to the fact that μ_4 and σ^4 (the fourth central moment and the fourth power of the mixture fraction rms, respectively) decay at the same rate, whereas with modified Curl, μ_4 decays less rapidly than σ^4 . The reason for the

appearance of the 'wiggles' is attributed to the effect of finite particle sample size (resulting in statistical 'noise') coupled with machine roundoff; such effects are inevitable in these numerical calculations. The reason why the appearance of the 'noise' is so prominent towards the final stages of mixing is because quantities such as μ_6 and σ^6 are then sufficiently small in magnitude to be comparable with machine roundoff. In the case of modified Curl (figure 6.20) since σ^6 decays considerably more rapidly than μ_6 , small fluctuations in μ_6 will result in large fluctuations in Ω_6 . The reason why the wiggles appear in the situation where $\tau = \tau_o$ and not for $\tau = \tau_o + (C_{\varepsilon_2} - 1)t$ is simply because in the former case the rms of ξ has dropped to a sufficiently small value so that these effects are visible. If the latter time scale is calculated long enough, the same behaviour will be observed. The effect of 'noise' is more prominent in quantities involving large exponents; therefore the presence of 'noise' is less apparent for σ (and hence σ^6) than it is for μ_6 . This explains why in figure 6.21, σ^6 decays more smoothly than μ_6 .

The reason why the behaviour of Ω_4 and Ω_6 are so different using the two mixing models has been highlighted in section 4.7.3. Basically the evolution of the higher order moments in the latter stages of mixing is strongly affected by the presence of particle properties near the bounds of the scalar sample space ($0 \leq \hat{\xi} \leq 1$)[†]. The pdf profiles shown in figures 6.22 and 6.23 encompass the statistics presented in figures 6.16 and 6.17. On the pdf plots, a Gaussian distribution has been superimposed having the same mean and variance as the pdf arising from the mixing model. It is clear from these figures that with modified Curl the presence of the persistent peaks at the bounds of the scalar domain and more generally the long tails of the pdf, are the reasons why the higher order moments remain larger than for LMSE. With LMSE the pdf evolution is completely unphysical, but because *all* the particles have to mix, all the particle properties will migrate towards the mean.

The simulations with modified Curl employed 10^5 particles to represent the pdf, although the same qualitative features of the turbulence could be captured using as few as 10^3 particles. For LMSE the combined effect of the initial conditions and the subsequent evolution of the pdf as two δ -functions approaching each other means that the simulation can be performed using only two particles - one for each δ -function. This has the advantage of reducing the run times drastically and removing a substantial amount of round-off when it comes to evaluating the moments of the pdf, particularly the higher order moments. It is important to note that such a sample size can only be used because the initial pdf is symmetric, discontinuous and it evolves (pathologically) as a double δ -function.

6.3.2 Reacting Test Cases

The calculations described in this section make use of the reduced chemistry presented in section 5. Consequently a *multi*-scalar pdf is evaluated at each time step and only the mean of ξ and n_{N_2} (the strictly conserved scalars) remain constant. The use of the 'look-up' (interpolation) table is not essential provided the number of particles representing the pdf is not too large ($\sim 10^3$); this means that the test case of homogeneous, isotropic turbulence can also be used to determine the accuracy of the interpolation technique, although the time required for these calculations can be typically an order of magnitude longer than for the laminar flame calculations. CH_4 is used as fuel so that comparisons can be made between the predictions of the two reduced mechanisms. The manner in which the particles are initially distributed characterises the initial pdf. Out of a total

[†] $\hat{\xi}$ denotes the sample space of the random variable ξ .

of $\mathcal{N}_p$ particles, $\mathcal{N}_{\xi_0}$ particles are placed at $\hat{\xi} = 0$ (corresponding to air) and hence $(\mathcal{N}_p - \mathcal{N}_{\xi_0})$ particles are placed at $\hat{\xi} = 1$ (corresponding to CH_4). The requirement is,

$$\tilde{\xi} = \frac{1}{\mathcal{N}_p} [\mathcal{N}_{\xi_0} \cdot 0 + (\mathcal{N}_p - \mathcal{N}_{\xi_0}) \cdot 1] = \xi_{st} \quad (6.7)$$

Equation (6.7) yields $\mathcal{N}_{\xi_0} = \mathcal{N}_p (1 - \xi_{st})$. It can be shown (appendix G) that,

$$\xi_{st} = \frac{W_{\text{C}_x\text{H}_y}}{\left(W_{\text{O}_2} + \frac{W_{\text{N}_2}}{\mathcal{U}}\right) \left(x + \frac{y}{4}\right) + W_{\text{C}_x\text{H}_y}} \quad (6.8)$$

For $\mathcal{U} \approx 0.265$, $x = 1$ and $y = 4$, equation (6.8) yields $\xi_{st, \text{CH}_4} \approx 0.055^\ddagger$. So if $\mathcal{N}_p = 1000$ then, $\mathcal{N}_{\xi_0} = 945$ and so $\mathcal{N}_{\xi_1} = 55$. In the case of LMSE significant cpu gains can be made by mixing and reacting only one of the particles in the $\hat{\xi} = 0$ group and another one from $\hat{\xi} = 1$; the remainder of the particles evolve in exactly the same way as their respective δ -function group members. This approach for LMSE can only be adopted for discontinuous initial conditions; for continuous initial conditions, all the particles have to mix and react.

6.3.2.1 Results using the Global Hydrocarbon Mechanism

Figure 6.24 shows the comparison between the predictions of modified Curl and LMSE. In these calculations an air temperature of 800K was employed so as to ensure that the reaction rates would be sufficiently large to allow for combustion to proceed effectively. However, based on the mechanism of Jones and Lindstedt [46] (and provided the conditions are adiabatic), a stoichiometric mixture of hydrocarbon fuel and air will ignite even if the air temperature is as low as 298K. It would obviously take a (pathologically) long period of time before the temperature is high enough for ignition to take place, but nevertheless, due to the irreversible fuel breakdown step using oxygen, the rate of consumption of fuel will be finite.

Figure 6.24 demonstrates that the behaviour of the two models with reaction appears to be significantly different for the CO and H_2 , however for the remainder of the species the same trends are predicted. It is clear that the behaviour of LMSE is somewhat erratic in comparison to modified Curl, and in order to understand the solution of LMSE in more detail figure 6.25 is considered. This figure describes the evolution of the individual δ -functions which when averaged produce the result in figure 6.24. Since the number of particles initially placed at $\hat{\xi} = 0$ is far greater than at $\hat{\xi} = 1$ (in order to ensure $\tilde{\xi} = \xi_{st, \text{CH}_4}$) the average result is weighted considerably more towards the evolution of the δ -function on the lean side. On the lean side figure 6.25 demonstrates that as soon as CH_4 is produced *through the effect of the mixing model*[§], it is consumed by the fuel breakup such that there appears to be no net change in the CH_4 profile. Furthermore, any H_2 that is produced is immediately combined with O_2 to yield H_2O ; some of this is used in the water-shift reaction to form CO_2 and H_2 from which the H_2 is then fed back into the H_2 - O_2 step. This explains the sharp rise in the CO_2 and H_2O concentrations and hence the rise in the temperature. In contrast, on the fuel rich side ($\hat{\xi} \geq \xi_{st, \text{CH}_4}$) the stoichiometry limits the reaction rate so that the temperature

[‡]In this section on homogeneous, isotropic turbulence ξ_{st} and ξ_{st, CH_4} will be used interchangeably

[§]The significance of this statement needs to be elaborated. It is clear that molecular mixing cannot create or consume a specie; this is a physical requirement. However, the initial condition is such that the δ -function on the lean side contains no CH_4 , yet on the rich side it does. Therefore there is an initial mean composition of fuel, and using LMSE the δ -function on the lean side will migrate towards this mean (which is itself evolving), thereby 'producing' CH_4 . This observation really indicates the extent to which LMSE lacks realism when applied to multi-scalar reactive problems.

risks nearly linearly over a relatively long period of time. During this mixing-dominated period the CH_4 concentration drops as the CH_4 δ -function approaches the stoichiometric (mean) value; this is matched by a rise in O_2 . When the temperature reaches about 1000K at $\frac{t}{\tau_o} \approx 2.5$, the O_2 virtually disappears in a very short period of time leading to the steep changes in the other profiles - most notably the rise in CO and H_2 . In this fuel-rich regime the formation of CO and H_2 is considerable whilst on the lean side any CO and H_2 that is formed is immediately converted to CO_2 and H_2O . The behaviour of the modified Curl closure appears to be more plausible in the sense that in the initial stages of reaction CO and H_2 are formed, but subsequently the final products (CO_2 and H_2O) are formed at the expense of the former intermediate species.

Figure 6.26 shows the solution for the test problem in figure 6.24 described above, only this time the interpolation tables are employed. It is clear that the interpolation technique works well; any noticeable discrepancies may well be associated with the lack of resolution of the table. Pdf shapes using modified Curl are presented in figures 6.27 and 6.28. The process of homogenisation brought about by the molecular mixing model is evident from figure 6.27. There is a persistent peak at $\hat{\xi} = 0$ due to the large number of particles originally placed there to ensure that the mixture is overall stoichiometric. The temperature pdf shows the large contribution at 800K corresponding to the δ -function associated with the air. As the reaction proceeds this spike diminishes in size and the temperature pdf broadens with a bias towards the higher temperatures.

The differences between LMSE and modified Curl have been noted in the context of the previous calculations. However, in all of these calculations the initial conditions were discontinuous. In order to perform a calculation with a 'continuous' initial distribution, the pdf from figure 6.27 at $\frac{t}{\tau_o} = 5$ was employed as a starting condition. An *unburnt* fuel/air mixture was specified in terms of this pdf and the mixture allowed to react. The result is shown in figure 6.29 - there is hardly any detectable difference between the use of these two mixing models. The implication of this is that the lack of realism in LMSE is only significant when discontinuous initial conditions are employed. If the discontinuities are somehow smoothened out, the behaviour of modified Curl and LMSE are quite similar.

6.3.2.2 Results using the Systematically Reduced Mechanism

The reactions reported using the global hydrocarbon scheme cannot be compared directly to those employing the systematically reduced scheme because with the latter mechanism a mixture consisting of CH_4 - O_2 - N_2 will not react - there is no path for the reaction to proceed. The test case amounts to a non-isothermal mixing problem. The procedure that is adopted to ensure combustion does take place is to employ an initial *triple* δ -function specification; one at each of the following locations: $\hat{\xi} = 0, \xi_{st}$ and 1. The δ -function at $\tilde{\xi} = \xi_{st}$ provides high temperature reactive species such as CO_2 and H_2O which help the overall reaction to proceed. The number of particles placed in each of these locations has to satisfy:

$$\mathcal{N}_{\xi_o} + \mathcal{N}_{\xi_{st}} + \mathcal{N}_{\xi_1} = \mathcal{N}_p \quad (6.9)$$

and,

$$\tilde{\xi} = \frac{1}{\mathcal{N}_p} (\mathcal{N}_{\xi_o} \cdot 0 + \mathcal{N}_{\xi_{st}} \cdot \xi_{st} + \mathcal{N}_{\xi_1} \cdot 1) \quad (6.10)$$

with the requirement that $\tilde{\xi} = \xi_{st}$. These equations are still not closed unless, for example, the ratio $\frac{\mathcal{N}_{\xi_{st}}}{\mathcal{N}_{\xi_1}} (\equiv \mathcal{R})$ is known. This is arbitrarily specified; typical values employed as part of the

sensitivity analysis were $\Re = 0.5, 1, 1.5$. Although these values do affect the evolution of the species concentrations, it was found that qualitatively the trends were much the same and for this reason the results that are presented employ $\Re = 1$. With the definition of $\Re$, the values of $\mathcal{N}_{\xi_o}$, $\mathcal{N}_{\xi_{st}}$ and $\mathcal{N}_{\xi_l}$ can be expressed as:

$$\frac{\mathcal{N}_{\xi_o}}{\mathcal{N}_p} = 1 - \xi_{st} \left(\frac{1 + \Re}{1 + \Re \xi_{st}} \right); \quad \frac{\mathcal{N}_{\xi_{st}}}{\mathcal{N}_p} = \Re \left(\frac{\xi_{st}}{1 + \Re \xi_{st}} \right); \quad \frac{\mathcal{N}_{\xi_l}}{\mathcal{N}_p} = \left(\frac{\xi_{st}}{1 + \Re \xi_{st}} \right) \quad (6.11)$$

For $\Re = 0$ the results stemming from equation (6.7) are obtained. When a continuous initial distribution is specified, the pdf presented in figure 6.27 ($\frac{t}{\tau_o} = 5$) is once again employed. In this situation the region of hot products is arbitrarily confined to a narrow band about $\hat{\xi} = \xi_{st}$ defined through: $\xi_l \leq \xi_{st} \leq \xi_r$ where l and r denote 'lean' and 'rich' respectively. $\xi_l = 0.052$ and $\xi_r = 0.058$, bearing in mind that $\xi_{st} = 0.055$. The manner in which the hot products were specified (as a function of ξ only) for the triple δ -function specification was also the subject of sensitivity analysis. The following were employed:

- A fully burnt mixture where $n_{CH_4} = n_{O_2} = n_{CO} = n_H = n_{CH_3} = n_{H_2} = 0$ and only H_2O , CO_2 and N_2 were present. The reaction steps (Ia-IV) shown in equation (5.10) would suggest that even such a specification would not allow a direct path for the consumption of CH_4 due to the absence of H and CH_3 . In fact it does and the reason is that the rate expressions for this reduced scheme are not directly coupled to the stoichiometry shown in equation (5.10), but rather to the elementary rates, expressed in equation (5.12).
- An equilibrium calculation using the method of Gordon and McBride [32] was employed to relate the levels of the reactive species to ξ_{st} . In this approach finite levels of CO , H_2 and H appear.
- The values of the species concentration at $\xi = \xi_{st}$ arising from the laminar flame calculations of Chelliah et al. [10] were employed. These provide slightly more H , CO and H_2 (in addition to some CH_3) than the equilibrium specification.

Qualitatively these various specifications had little effect on the results, and for this reason predictions are only shown for the specification involving a fully-burnt mixture.

Figures 6.30 and 6.31 show the results of the calculation using modified Curl and LMSE respectively. As with the global hydrocarbon scheme, the behaviour of LMSE appears to be very unrealistic. Modified Curl demonstrates a more plausible evolution of the scalar properties. In analogy with figure 6.29 where LMSE and modified Curl gave exactly the same result when subjected to a continuous initial distribution, it is surprising to note in figure 6.32 that in the context of the systematically reduced scheme this is far from being the case. The behaviour of LMSE approaches that of modified Curl to a small extent, however, significant discrepancies still exist. The reasons for this are not entirely clear, although the robustness of the systematically reduced mechanism does appear to be a factor in characterising the results. Errors in the implementation of this scheme are ruled out on the grounds that the results of the laminar flame calculations appear to agree (qualitatively) with those using the global hydrocarbon scheme.

Another area of concern is the ability of the interpolation table to reproduce the results obtained when the stiff ODE's are integrated directly. This comparison is made in figures 6.33 and 6.34. CH_4 , O_2 , CO_2 and temperature are rather well reproduced; however, the H_2 , H and CO fields

Mass Flow Rate		Exit Velocity U_o [m s ⁻¹]	Burner Exit Reynolds number Re
C_3H_8 [Kg s ⁻¹]	H_2 [Kg s ⁻¹]		
9.14×10^{-4}	0.055×10^{-4}	17.9	24000
11.99×10^{-4}	0.073×10^{-4}	23.6	31500
16.25×10^{-4}	0.287×10^{-4}	31.9	42700

Table 6.1: Experimental conditions for the propane-air jet diffusion flame of Godoy [31]. Burner diameter 6mm.

are quite poorly reproduced. In the context of the laminar flames the discrepancies between the 'exact' and the interpolated results were found to be marginal, yet in this situation they are quite significant. No further reasons for the differences, in addition to those proposed for the case of the laminar flame, are known at this stage.

6.4 Predictions of Propane-Fuelled Turbulent Jet Diffusion Flames

6.4.1 The Flame of Godoy

Godoy [31] investigated a vertical, axisymmetric, turbulent jet diffusion flame, where the fuel jet (propane) issues into stagnant air surroundings. A detailed discussion of the experimental procedure that was employed to obtain the measurements can be found in the work of Godoy, and in this section only a brief summary is provided. Propane is often taken as being representative of many practical fuels and a round jet is a simple geometry that allows easy access to every zone in the flow. For the Reynolds numbers used in the experimental study ($Re \geq 2 \times 10^4$) it was necessary to stabilise the flame; to do so an annular pilot hydrogen flame was employed. The mass flow rate of the pilot flame was very low compared with the mass flow rate of the main fuel. It was argued that the pilot affected only the flame root and that in the downstream region ($x/D \geq 20$, where D is the initial diameter of the fuel jet) where most of the measurements were made, the species composition was not affected. The burner itself was a stainless steel tube of 6mm inside diameter. The thickness of the annulus conveying the pilot gases (~ 0.2 mm) could be varied through a tapered arrangement in order to stabilise the flame more effectively. Typically the turbulent flame was about 200 burner diameters long.

To minimize buoyancy effects (and to ensure fully developed turbulent conditions), the predominance of inertial forces was achieved by using as high Re as possible. Three turbulent flames with $Re = 24000$, 31500 and 42700 (defined at the nozzle) were studied (see table 6.1). The flow conditions near the burner exit were dominated by the exit momentum flux; further downstream the inertial effects were reduced by the lower velocities such that in these regions ($x/D \geq 100$) the effect of buoyancy became more important. In the turbulence models employed in the current computational study, no account of buoyancy is included with the exception of the treatment of the gravitational term in the momentum equation. This, however, is far from sufficient in addressing the effect of buoyancy on the evolution of the jet. With the Reynolds stress closure, the modelling of the fluctuating pressure terms (section 2.5.2.3) could be extended to include buoyancy effects, but the aim of the work described in this thesis is on the evaluation of the scalar pdf formalism

rather than turbulence modelling.

The measurements of CO and CO₂ were made using non-dispersive infrared analysers, N₂ and O₂ using gas chromatography and the unburnt hydrocarbon (UHC) with a flame ionization detector. For analysis using the infrared analysers and the gas chromatogram, the samples need to be free of water. The UHC was expressed as equivalent propane. Velocity measurements were made by Wilhelmi (cited in [31]) for $Re = 24000$ using an LDV system. The propane flow was seeded with TiO₂ particles, but at the edge regions of the flame and for $x/D \geq 80$, sufficient particles were not detected in the measuring volume in order to yield a reliable measurement of velocity. Consequently these measurements do not extend as far downstream as the scalar properties.

The mixture fraction (calculated from the measurements) employed the carbon element mass fraction.

$$\xi \equiv \frac{n_C}{n_{C,1}} = (3n_{C_3H_8} + n_{CO} + n_{CO_2}) \left(\frac{W_{C_3H_8}}{3} \right) \quad (6.12)$$

where $n_{C,1}$ is the specific mole number of carbon *atoms* in the fuel stream (denoted by '1'). The quantities appearing in equation (6.12) need to be concentrations on a *wet* basis and whilst the measurements provide the wet concentration of UHC, the remainder of the scalars are obtained on a dry basis; all the measured concentrations are expressed as mole fractions. The wet concentrations (X_α) are related to the dry (X_α^d) through the following relation:

$$X_\alpha^d = \frac{X_\alpha}{1 - X_{H_2O}} \quad (6.13)$$

where X_α denotes the mole fraction of species α ($\alpha = 1, \dots, N$). The mole fraction is related to the specific mole number through,

$$X_\alpha = W_m n_\alpha, \quad \text{where} \quad W_m \equiv \sum_{\beta=1}^N W_\beta X_\beta = \left(\sum_{\beta=1}^N n_\beta \right)^{-1} \quad (6.14)$$

W_m is referred to as the *mixture* molar mass. In order to evaluate ξ the values of X_{H_2} and X_{H_2O} need to be calculated. The latter can be obtained by assuming a constant oxygen-to-nitrogen ratio,

$$\frac{n_O}{n_N} = \mathcal{U} = \frac{n_{H_2O} + n_{CO} + 2n_{CO_2} + 2n_{O_2}}{2n_{N_2}} = \frac{X_{H_2O} + X_{CO} + 2X_{CO_2} + 2X_{O_2}}{2X_{N_2}} \quad (6.15)$$

Since X_{CO}^d , $X_{CO_2}^d$, $X_{O_2}^d$ and $X_{N_2}^d$ are known from the measurements, using equation (6.13) they can be substituted into equation (6.15) to obtain X_{H_2O} (with $\mathcal{U} \approx 0.265$). Once X_{H_2O} is known, all the known wet concentrations (including the measured $X_{C_3H_8}$) can be substituted into the mass conservation equation, $\sum_\alpha X_\alpha = 1$, to yield X_{H_2} . When all the mole fractions are known, equation (6.14) can be used to obtain the mixture molar mass (W_m) and then the species specific mole numbers (n_α). The appropriate n_α can then be used in equation (6.12) to calculate the mixture fraction.

Before any calculations of Godoy's flame are presented, it is essential to establish that the Monte Carlo simulation that is being employed provides a very good approximation to the finite-difference solution of the governing equations. Obviously this comparison cannot be performed for the case of the reacting flow since the finite-difference solution of the multi-scalar pdf equation is prohibitively time consuming. Instead, the conditions in Godoy's burner are employed (e.g. the same initial fuel jet diameter, mean inlet velocity and stagnant surrounds) but with the C₃H₈ issuing

from the burner replaced by air so that a constant density, inert flow calculation can be performed. In this case the only scalar independent variable is the mixture fraction which acts as passive scalar. The transport equation for the mean and variance of the mixture fraction (equations (3.8) and (3.9)) are solved and compared with the solution of a Monte Carlo simulation for $P(\hat{\xi}; x, \omega)$; the results are shown in figures 6.35 and 6.36. With gradient modelling used for approximating the turbulent transport, and a mixing model which predicts a variance decay rate in accordance with equation (4.6), equation (4.3) can be multiplied by ψ or $(\psi - \langle \phi \rangle)^2$ (for the case of a single scalar pdf) and subsequently integrated over the scalar space to yield equations (3.8) or (3.9), respectively. Figures 6.35 and 6.36 demonstrate that based on a sample size of 50,000 particles the Monte Carlo simulation represents the evolution of the statistics very well. The largest differences can be found in the profiles of the variance. The higher order statistics, such as the variance, are certainly more sensitive to errors arising from the use of a finite sample size than the mean and this explains why the variance predicted from the Monte Carlo simulation exhibits more statistical 'noise'. The variance profiles compare favourably with the result of the finite-difference equation in the sense that over the region where the gradients are significant the solutions are very similar. The discrepancies towards the edge of the solution domain are not affected by increases in the number of particles used to represent the pdf. As the edge of the solution domain is reached $\tilde{u} \rightarrow 0$, and from equation (D.30), this implies that $\delta y \rightarrow \infty$. Consequently there are resolution limitations in this region, such that a small difference in the predicted entrainment rate can show up as a large difference in the radial profile.

Predictions of Godoy's measurements are presented for the flames where the initial pipe Reynolds numbers are 24000 and 42700; these are the test cases where the flow experiences the largest and smallest residence times, respectively. Except for the treatment of the piloted methane-air jet diffusion flames (discussed in section 6.5.2) the remaining experimental jet flames that are calculated in this thesis are all *artificially 'ignited'*. This is achieved by allowing the jet to evolve as a *non-reacting* flow up to an arbitrarily located downstream position (typically one to four burner diameters); at this 'ignition' plane, those particles along the radial axis with a mixture fraction lying between the flammability limits of the fuel are assigned the composition of fully-burned products consistent with the local mixture fraction. This mixture comprises CO_2 , H_2O , N_2 and *surplus* fuel or oxygen depending on the local stoichiometry. If the mixture is 'ignited' too close to the burner exit, the rich mixture hinders further reaction to take place, so that the mean temperature rapidly drops; the ignited jet soon reverts to a non-isothermal mixing problem [15]. Specifying a wider range for the mixture fraction (than that corresponding to the flammability limits) has little effect on the subsequent downstream scalar evolution since those particles which are either too fuel-rich or lean do not have a sufficiently high temperature to allow for appreciable chemical reaction over the time scales of the flow.

Figures 6.37 to 6.48 are the results for the test case $Re = 24000$ using a $k-\epsilon$ formulation, modified Curl and LMSE, fifty cross-stream nodes (1000 particles per node) and a forward step size of 1% of the half jet width. The simulations using modified Curl required about three hours on a Silicon Graphics (Iris Indigo) workstation using global optimisation for compilation purposes. For LMSE, the run time was about five hours. There are two reasons why LMSE should require more time to perform the calculation. Firstly with LMSE all the particles are subjected to the mixing process, whereas with modified Curl only those particles which are randomly selected undergo mixing. Secondly, with LMSE the run times are strongly affected by the choice of Λ'_m , equation (E.26), and for the calculations reported in this thesis $\Lambda'_m = 0.2$ was employed to ensure

that the mixing time step is not too large; however, it is felt that a larger Λ'_m could have been used without a significant effect on the results. The initial conditions were characterised by top hat velocity and turbulence kinetic energy profiles. The level of k was initially set to allow for turbulence intensities of about 5% of the mean velocity of the fuel issuing from the burner. The initial turbulence length scale at the burner exit was taken as a ramp function which was uniform in value over most of the cross-section and dropped linearly to zero at $r = D$. The dissipation rate of k was then estimated using $\varepsilon = \frac{k^{3/2}}{l}$. It is clear that only the global hydrocarbon scheme [46] can be employed for the propane flames since the systematically reduced mechanism [10] is designed to work for methane-air combustion only.

Figure 6.37 demonstrates that the mean mixture fraction is predicted very well in the region $x/D \leq 80$. The similarity of the predictions of LMSE and modified Curl is an interesting result; this ties in well with the results of homogeneous reacting turbulence (section 6.3.2.1) where the statistics of the scalar field with the two mixing models is virtually the same provided the initial condition is a continuous pdf. In this situation the initial conditions associated with the properties in the burner and the free stream imply a discontinuous pdf; however, the process of transport in physical space smoothens the initial discontinuities. Interest is mainly confined to the results for $x/D \leq 80$, since it is in these regions where the influence of buoyancy is not appreciable. The predictions of the nitrogen evolution (figure 6.38) follows much the same trends (in terms of accuracy) as the mixture fraction field. Since there is no source of nitrogen, it is transported towards the centre-line along the mean concentration gradient, and at $x/D \approx 200$, the radial profile is nearly uniform. Figure 6.39 presents the comparison of the velocity predictions using the k - ε model and the measurements. The turbulence model reproduces the measurements very well; such a good level of agreement is to be expected since the k - ε model has been subjected to a considerable amount of testing in flows where the thin shear layer approximation is valid. This is why the jet configuration was chosen for the current application of the transported pdf formalism so that the shortcomings of the turbulence model in reproducing the flow behaviour are reduced; without an accurate representation of $\tilde{\xi}$ and $\tilde{u}$, it is not worthwhile trying to proceed with the evaluation of the reactive scalar field.

Figure 6.40 shows the predictions of C_3H_8 and the measurements of UHC (taken to be propane). The consumption and transport of fuel appear to be well reproduced; in particular at $x/D = 200$ the pdf formalism is in agreement with the measurements in that virtually complete fuel burn-up has taken place - this indicates that the global chemical mechanism reproduces the gross flame features quite well. The corresponding profiles for the oxygen concentration is shown in figure 6.41. Godoy's measurements indicate that for $x/D \leq 40$ there is no O_2 detected on the axis of the jet. This is an interesting observation because it is at variance with measurements in methane-air turbulent jet diffusion flames reported by Hassan et al. [35] (described in section 6.5.1). The reasons for this are far from certain and may well depend on how the flame was initiated. Effects which could explain finite (centre-line) levels of O_2 include: local partial extinction due to very rapid mixing near the burner, finite intermittency near the centre-line or oxygen penetration between ring vortices shed from the burner. However, Hassan et al. [35] argued that these explanations were unlikely; local extinction effects were ruled out due to the presence of the hydrogen pilot flame in the near burner region, vortex shedding was simply not observed and the centre-line pdf's of temperature did not indicate the presence of any intermittency. The only other plausible explanation put forward by Hassan et al. involved the basic intermittency phenomenon at the 'edge' of the jet, whereby fresh fuel and air may be brought into contact without the

presence of hot products, leading to a partial premixing. The *predictions* in figure 6.41 indicate finite centre-line concentrations of O_2 in regions where the measurements suggest there should be none. Furthermore, as more fuel is consumed the measurements correctly indicate that the centre-line O_2 concentrations should rise; at least in the region $80 \leq x/D \leq 100$ the predictions do not reproduce this effect. The reason why the predictions indicate O_2 near the centre-line at $x/D = 20$ is probably related to the method by which the mixture is 'ignited' for the purpose of the calculation procedure.

The predictions of the products of chemical reaction (CO_2 , CO , H_2 and H_2O) are shown in figures 6.42 to 6.45. The most striking observation is the persistent overprediction of CO throughout the entire region over which the flame was experimentally investigated. Measurements of the H_2 concentration are not available, however figure 6.44 also indicates that the peak levels of H_2 are much greater than the typical values found in laminar or turbulent flames. The overpredictions in CO need to be matched by underpredictions of the other carbon-containing species in the reactive mixture. More will be said about the conservation of elemental species further on; for the time being it is argued that given the largely disparate scales between the concentrations of C_3H_8 and those of CO , CO_2 , a small underprediction in C_3H_8 is sufficient to make available a large pool of carbon atoms which would have to be taken up by CO and CO_2 . Generally speaking the CO_2 predictions can be considered to be satisfactory to the extent that the peak values are reasonably accurate. Qualitatively, the results are consistent in that as the fuel is (nearly) entirely consumed by $x/D = 200$, the CO and H_2 levels start to drop, leading to the formation of H_2O and CO_2 . The lack of discrepancy in the results using modified Curl and LMSE is an important outcome, suggesting that both are equally as 'good' at representing the 'molecular mixing' behaviour.

The results discussed above are reminiscent of the predictions of the confined laminar flame, section 6.2. In fact, the calculation of Godoy's flame was originally attempted prior to the simulation of the laminar flame of Mitchell et al. [69]. Upon examination of the results for the turbulent propane jet flame it was felt that the overprediction in the CO field was difficult to understand given that the global scheme yielded very good CO and H_2 levels in laminar flames (with a realistic treatment of diffusion). This was the motivation behind the calculation of laminar flames with equal diffusivity being assumed, and it is felt that the results in the laminar flame may have some bearing on the behaviour in the turbulent flame.

The mixing models that were employed for the turbulent flame calculations employ $\frac{k}{\epsilon}$ as the characteristic turbulent time scale. Therefore it is of interest to compare the profiles of $\frac{k}{\epsilon}$ from the results in this flame. Figure 6.46 shows that over a large section of the jet $\frac{k}{\epsilon}$ is predicted in much the same way by both models. The discrepancies which do arise near the edge of the jet can be explained as follows: $\frac{k}{\epsilon}$ is a quotient whose value is obviously sensitive to the magnitude of the numerator and the denominator, particularly near the edge of the jet where k and ϵ both tend to zero. Figures 6.47 and 6.48 indicate that the behaviour of k and ϵ (respectively) is much the same regardless of the mixing model and that the differences appearing in figure 6.46 are of little physical consequence.

The predictions shown in figures 6.37 to 6.48 employed 50,000 particles; typically the pdf at each point along the cross-section of the jet was represented by 1000 particles. The effect of the sample size on the accuracy of the results needs to be established if conclusions can be confidently made concerning the strengths and the weaknesses of the scalar pdf technique. Figure 6.49 is a graph which helps to indicate how well the current Monte Carlo/pdf approach conserves mass. With the calculation procedure that is being used, mass continuity is in principle automatically

satisfied because of the transformation of the governing equations to dimensionless stream function coordinates. Despite the fact that the reactive species concentrations are changing as the jet evolves downstream of the burner, at any particular cross-section the flow rate of *element* mass fraction should be constant. Due to the equal diffusivity assumption, the measure of this is given by,

$$\mathcal{I}(x) \equiv \int_0^{R(x)} \bar{\rho} \tilde{u} r \tilde{\xi} dr \quad (6.16)$$

Substituting from equations (5.3) and (5.4), (6.16) becomes:

$$\mathcal{I}(x) = (\Psi_E - \Psi_I) \int_0^1 \tilde{\xi} d\omega \quad (6.17)$$

For mass conservation to be observed $\mathcal{I}(x)$ should remain invariant as x increases. Figure 6.49 shows how the sample size affects this result; with 10^6 particles (i.e. 2×10^4 particles per node) $\mathcal{I}(x)$ varies by very small amounts ($\leq 5\%$) whilst with 50,000 particles there is about 12% variation by about $x/D = 200$, relative to the starting value at $x/D = 0$. Figure 6.49 demonstrates that in the limit of the sample size tending to infinity, $\mathcal{I}(x)$ will become a horizontal line. The large deviations in $\mathcal{I}(x)$ with 50,000 particles may suggest that the mean profiles shown in figures 6.37 to 6.48 may be inaccurate. For this reason a calculation was performed using 10^6 particles (all other conditions were kept the same). The results (figures 6.50 and 6.51) using modified Curl are only shown for $\tilde{\xi}$ and $\tilde{X}_{\text{CO}}$; the level of agreement was found to be as good for the other scalars (which are not shown). It is clear that the substantial increase in sample size has very little effect on the profiles, implying that the results previously discussed (based on a sample size of 50,000 particles) are quite satisfactory.

Calculations were also carried out to investigate the performance of the Reynolds stress closure. The reason why the application of this turbulence model was attempted is because it provides another (potentially more accurate) method of calculating the k and ε fields and since these feed into the molecular mixing models, it would be of interest to find out what effect this would have on the predicted scalar properties. In terms of the velocity field, figure 6.52 indicates that there is little difference between the two models; this is to be expected. The results (using modified Curl) for the mixture fraction and the CO field are shown in figures 6.53 and 6.54. The Reynolds stress closure does not appear to affect the results significantly; in fact the differences in the predicted quantities using the two turbulence models appears to be of the same order as the difference when using the two mixing models. The peak levels of CO (and H_2 , not shown) for $x/D \leq 80$ are hardly affected. As far as the predictions of the k and ε fields are concerned, the differences arising from the use of the two turbulence models is typically less than about 8%.

The final set of results relating to the Godoy flame are the predictions of the measurements at $Re = 42700$. So far very little has been said about buoyancy. Nevertheless, in reality, the increase in Reynolds number does influence the role of buoyancy. It is to be expected that decreasing the Reynolds number increases the role of buoyancy. The effect of buoyancy is to increase the mixing rates such that more air from the surroundings is entrained; Godoy [31] points out that this occurs at a roughly constant spreading rate. This is plausible provided the overall velocity at a section also increases under the influence of buoyancy in conjunction with the increased entrainment. Because more air is entrained, the fuel burns up more quickly and so the flame shortens. These

observations are borne out by the measurements of Godoy. The important feature emerging from the predictions and measurements (figure 6.55 to 6.57) is the apparent insensitivity of the peak levels of the reactive scalar field to the Reynolds number; doubling the Reynolds number halves the mean residence time of the flow. This implies that in this particular flame the residence times of the flow are such that there are no significant kinetic effects (e.g. local extinction) operative. More will be said about such effects when discussing the piloted diffusion flame of Masri et al. [66] in section 6.5.2.

Jones and Kollmann [44] had also investigated the flame of Godoy [31] but using a different, and less realistic, chemical scheme. A reduced scheme for the oxidation of propane was employed; the thermochemical state (including composition, density and temperature) was obtained via a partial equilibrium assumption in which the free energy was minimized subject to the constraints that the mixture fraction, total mole number and mass fraction of C_3H_8 (three-scalar chemistry) take predetermined values. Such a constrained equilibrium approach led to underpredictions of CO and the agreement of the CO_2 predictions was poorer than displayed in figure 6.42. More recently Chen and Kollmann [12] extended the work by incorporating a four scalar description of the chemistry. The major difference between the three and four scalar description lied in the assumption of partial equilibrium for the CO oxidation process. In [44] the CO/ CO_2 ratio was determined entirely by the H/OH ratio and the equilibrium constant, whilst in [12] CO was treated as an independent scalar. The effect was a significant improvement in CO and CO_2 predictions (although they were still under and overpredicted, respectively) with little effect on the mixture fraction profiles. Clearly, the treatment of the chemistry has little effect on the conserved scalars, but a greater effect on the reactive scalars.

6.4.2 The Flame of Heitor, Moreira and Pires

The propane-air turbulent jet diffusion flame investigated by Heitor et al. [37]² has a very similar configuration to that of Godoy [31]. The differences are that the fuel pipe diameter is 5mm (instead of 6mm) and the mean fuel inlet velocity is 33ms^{-1} (in place of the values shown in table 6.1); the Reynolds number is about 37000. This flame is investigated primarily because of its similarity to the configuration of Godoy, but in addition measurements of temperature and velocity are reported. The measurement techniques employed for sampling the CO, CO_2 and velocity fields were the same as those described in section 6.4.1; measurements of temperature were made with thermocouples ($40\mu\text{m}$ diameter).

The results are presented in figures 6.58 to 6.61. Figure 6.58 shows that the predictions of CO and CO_2 are consistent with the levels calculated for the Godoy flame. In contrast the measurements of Heitor et al. indicate that the peak CO level is at most 4% (mole fraction) whereas in the flame discussed in the previous section, the level of CO is typically 6%. Figure 6.59 demonstrates that since CO and CO_2 are overpredicted, the levels of O_2 are underpredicted and consequently higher temperatures are calculated. Another interesting observation concerning the measurements is the absence of O_2 near the centre-line, in agreement with the measurements of Godoy. The mean velocity (figure 6.60) is very well reproduced at the first two measuring

²In this experimental study two different burners were employed. One with an internal contraction prior to exit into the stagnant surrounds and the other with a pipe of constant cross-section. The latter burner is discussed in this work, since the former cannot be treated by the current calculation procedure which is specifically set up to solve boundary layer flows.

stations. However, by $x/D = 30$ the predictions suggest a faster spreading rate of the jet. The superiority of the Reynolds stress model in predicting $(\widetilde{v''^2})^{1/2}$ is evident, although the differences are not significant, particularly in relation to $\widetilde{u''v''}$. Figure 6.61 presents the predictions of $(\widetilde{u''^2})^{1/2}$ and $(\widetilde{T'^2})^{1/2}$; for $x/D \geq 10$ the Reynolds stress model appears to reproduce the measurements of $(\widetilde{u''^2})^{1/2}$ quite well. However, despite the simplicity of the $k-\epsilon$ model (in comparison to the Reynolds stress model) it is clear that it also performs satisfactorily and the effect of these differences on the reactive scalar field appears to be quite small.

The calculation procedure appears to reproduce the *fluctuating* velocity field quite well. This implies that the predictions of k (and therefore those of ϵ) are at least of the right order of magnitude and qualitatively correct. This, in turn, means that the apparent overpredictions in CO (for example) is not related to the inaccuracy of $\frac{k}{\epsilon}$ which serves as an input to the mixing model; it appears that it has more to do with the lack of physics underlying the derivation of the mixing models.

6.5 Predictions of Methane-Fuelled Turbulent Jet Diffusion Flames

6.5.1 The Flame of Hassan, Lockwood and Moneib

In the experimental study of Hassan et al. [35] three types of flames were investigated: burner stabilised flames with and without N_2 dilution of the fuel stream and lifted flames. Given the manner in which the flow is artificially ignited in the current calculation procedure, it is clear that the predictions need to be restricted to the burner stabilised flames. The fuel employed was North Sea gas (94 % methane), chosen on the grounds that it avoids the significant formation of soot. The burner pipe was thin-walled, vertically aligned and 7.74mm in diameter. It was long enough to ensure fully-developed conditions at its exit. Flame stabilisation was achieved by admitting H_2 from a 0.25mm wide annulus between the burner and a surrounding pipe. The flow rate of H_2 was 1% by mass of that of the fuel. In contrast to the previous experimental investigations, Hassan et al. chose to employ a quartz microprobe relying on sonic expansion for sample quenching, in preference to the bulkier alternative of a water-cooled stainless steel probe. The justification for its use was that quartz probes provide minimal thermal disturbance and possess diminished catalytic activity.

Each of the flames studied were operated at a pipe exit Reynolds number in the range $10^4 \leq Re \leq 3 \times 10^4$ (transition $Re \approx 500$ for free jets). The initial conditions corresponding to the flames investigated by Hassan et al. are summarised in tables 6.2 and 6.3. Since it assumed that the fuel is methane, the systematically reduced scheme of Chelliah et al. [10] can also be employed for the calculations. Measurements of UHC, H_2O and N_2 were not made and so it is not possible to calculate a mixture fraction from the measurements. Detailed measurements of the radial profiles were only reported for run SA2. Figure 6.62 shows the results for the CO and temperature fields using the global hydrocarbon mechanism. As with the Godoy flame, the effects of buoyancy become important in the downstream solution ($x/D \geq 80$). The mean temperature profiles at $x/D = 36.2$ and 75 appear to be very well predicted with little difference between the mixing models. By $x/D = 204$, the temperature profile is overpredicted by a significant margin, particularly near the centre-line; this may suggest that the measured flow is cooling down (through the entrainment of 'cold' air from the surroundings) more rapidly. Since the buoyancy

Run	Mass Flow Rate		Burner Velocity U_o [m s ⁻¹]	Burner Exit Reynolds number
	Fuel [Kg/hr]	H ₂ [Kg/hr]		
SA1	3.283	0.0227	29.5	13362
SA2	4.924	0.1545	44.3	20065
SA3	8.207	0.3070	73.9	33454
SA4	9.848	0.3347	88.6	40130

Table 6.2: Stabilised natural gas diffusion flames, Hassan et al. [35]. $D = 7.74$ mm. Burner velocity chosen to conform with the fuel mass flow rate, $\rho_{CH_4} = 0.656$ Kg m⁻³ and $\mu_{CH_4} = 1.121 \times 10^{-5}$ Kg m⁻¹s⁻¹ at 298K.

Run	Mass Flow Rate			Density $\bar{\rho}$ [Kg m ⁻³]	Burner Velocity U_o [m s ⁻¹]
	Fuel [Kg/hr]	N ₂ [Kg/hr]	H ₂ [Kg/hr]		
NA1	3.06	1.865	0.169	0.78	37.3
NA2	2.185	2.74	0.1545	0.86	33.8
NA3	1.311	3.613	0.1545	0.95	30.6
NA4	0.874	4.05	0.144	1.01	28.8

Table 6.3: Stabilised natural gas/N₂-diluted diffusion flames, Hassan et al. [35]. $D = 7.74$ mm. $Re \approx 16500$

forces influence the entrainment, it is clear that to reproduce the measurements in the region of $x/D = 204$ requires modelling of the buoyancy effects. The overprediction of the CO has been a major characteristic of the solution formalism that has been adopted, even though the qualitative trends appear to be correctly represented. The maximum levels of CO predicted are slightly less than for the Godoy flame. One possible reason for this is that a different hydrocarbon is being used, with fewer carbon atoms per molecule. Figure 6.63 presents the predictions of O₂ and CO₂ in relation to the measurements. As with the Godoy flame, the peak levels of CO₂ are well reproduced for $x/D \leq 80$. In accordance with the overprediction of temperature at $x/D = 204$, the mean O₂ and CO₂ concentrations are under- and overpredicted respectively. Figure 6.64 indicates that the overpredictions of CO are matched by very high H₂ concentrations, particularly at $x/D = 75$. However, by $x/D = 204$ all the fuel has been consumed and most of the CO and H₂ have been oxidised to CO₂ and H₂O. Figure 6.65 shows the axial (centre-line) evolution for the scalar properties. The most accurately predicted quantity appears to be the mean temperature for $x/D \leq 100$. Qualitatively the rms of temperature is acceptable for $x/D \leq 100$, whilst the subsequent (measured) peak may well be due to more 'cold' air particles amongst the hot burnt pockets so that the overall mean temperature decreases whilst the fluctuations increase. Whilst the measurements indicate a finite level of O₂ along the centre-line, the calculation predicts that by about 50 diameters this has virtually disappeared. The behaviour of CO₂ is quite satisfactory; however the reasons for the subsequent (predicted) plateau in the CO₂ level are not clear. The strong rise in the CO concentration coupled with virtually no centre-line O₂ is clearly a factor, since the CO₂ profile starts to rise again as the centre-line CO levels drops and that of O₂ rises.

Hassan et al. [35] point out that the most noteworthy feature of the scalar field measurements is the finite O₂ concentration for $x/D \leq 80$. In this respect, the effect of N₂ dilution is to

reduce, relative to the undiluted flame, the centre-line oxygen concentration for $x/D \leq 80$. It is argued that the effect of N_2 dilution is to increase the stoichiometric mixture fraction, and as a consequence it "suppresses the retardation of reaction and so the oxygen concentration". With N_2 dilution, ξ_{st} is given by:

$$\xi_{st} = \frac{W_{C_xH_y}}{\left(W_{O_2} + \frac{W_{N_2}}{U}\right) \left(x + \frac{y}{4}\right) \left(1 - \frac{\dot{m}_{N_2,1}}{\dot{m}_{C_xH_y,1} + \dot{m}_{N_2,1}}\right) + W_{C_xH_y}} \quad (6.18)$$

where $\dot{m}_{N_2,1}$ and $\dot{m}_{C_xH_y,1}$ are given in table 6.3. The derivation of this expression is given in appendix G. Equation (6.18) shows that for $\dot{m}_{N_2,1} \geq 0$ the magnitude of ξ_{st} is greater than if $\dot{m}_{N_2,1} = 0$. Hassan et al. noted that as a consequence of the increase in ξ_{st} the flame is shortened in inverse proportion. The addition of N_2 affects the bounds for the construction of the interpolation table. These details are discussed in appendix F.

Figure 6.66 shows the results for the oxygen field comparing the measurements and predictions of the burner stabilised flames, with and without N_2 dilution. The predictions of the NA1 and NA2 flames do not demonstrate a reduction in the peak levels of O_2 , for $x/D \leq 50$, relative to SA1 and SA2. For NA3 and NA4 there is a significant reduction in the centre-line O_2 concentration relative to SA3 and SA4. Furthermore, the general level of agreement between the predictions for the nitrogen diluted flames is better than for the non-diluted flames. As $\dot{m}_{N_2,1}$ is increased the centre-line temperature profiles (not shown) indicate that the peak temperatures, albeit of the same magnitude, fall away more quickly than if $\dot{m}_{N_2,1} = 0$. Thus, as more N_2 is added to the fuel stream, at the expense of the fuel itself, the amount of reaction taking place is less pronounced; this may explain why the level of agreement between measurement and prediction improves for these test cases. The improvement in the prediction of CO_2 and CO profiles (figures 6.67 and 6.68) is also evident. In the case of CO_2 , it can be noticed that the peak levels move closer towards $x/D = 0$ as the degree of N_2 dilution increases; this is consistent with the observation that the mean flame length shortens. The effect of N_2 dilution on the CO concentration results in overpredictions for NA1 and NA2 (consistent with the undiluted behaviour), but for NA3 the centre-line evolution is very well reproduced. For NA4 the levels CO are underpredicted; these results are in stark contrast SA1-SA4 where CO is consistently overpredicted, and the peak levels are hardly unaffected by the flow residence times (as shown by the measurements and the predictions).

The discussion now centres around the predictions of the non-diluted flame (SA2) using the systematically reduced mechanism of Chelliah et al. [10]. The results initially presented for this test case were performed with the mixture being artificially ignited at 15 diameters downstream of the burner. This is an unrealistically large distance and the reason for igniting the mixture at such a location is because with ignition at $x/D = 10$, the mean flame temperature drops monotonically thereafter; the flow reverts to a mixing problem. This clearly indicates that the reactive mixture at the ignition plane cannot sustain itself downstream. This result raises questions with regards to the suitability of the systematically reduced mechanism with the transported pdf formalism. For the time being the results obtained with ignition of the mixture at $x/D = 15$ will be discussed.

Figure 6.69 shows the results for the mixture fraction, temperature and mean density. The first significant observation is the difference in the predictions of LMSE and modified Curl at $x/D = 75$, whilst at $x/D = 36.2$ the results are quite similar. LMSE is manifesting a tendency towards extinction and the reasons for this are not entirely clear. The calculations of homogeneous, isotropic and reacting turbulence indicated that with the global hydrocarbon scheme employing a continuous initial distribution, the scalar field evolved in the same manner with the two mixing

models. However, this was not the case with the systematically reduced mechanism, and the current result could be interpreted as a manifestation of this in an inhomogeneous test case. With modified Curl, the temperature in figure 6.69 is predicted just as well as in figure 6.62. Whilst the difference in $\bar{T}$ using modified Curl and LMSE is significant at $x/D = 75$, the effect on the mixture fraction profiles appears to be very small. Figure 6.70 shows some more features of the reactive scalars. The peak CO levels are not overpredicted. However it would be unwise to assume that this result has been achieved because the chemistry is more realistic; this is because to achieve these results the mixture had to be ignited at $x/D = 15$, which is considered too far downstream. From the oxygen profiles it is clear that the centre-line concentrations are overpredicted at $x/D = 36.2$ and $x/D = 75$; this partly explains why the CO levels appear to be more sensible. The differences in the CO and H₂ levels arising from the use of the two reaction schemes are significant; upon close examination of the CH₄ profiles (figures 6.64 and 6.71), the mechanism of Chelliah et al. [10] suggests that there is more fuel present, particularly at the larger radii (which occupy a greater volume). With modified Curl the profiles of CO₂ and H₂O do not differ substantially for the two schemes. This is to be expected since the temperature profiles do not differ significantly as pointed out earlier on. It is difficult to comment on the accuracy of the radical species (figure 6.71) since there are no measurements available. The peak values of CH₃ appear to be of the same order of magnitude relative to the peak values in the laminar flame calculated by Chelliah et al.; this is in contrast to the H-radical, where the concentrations are an order of magnitude higher. The fact that LMSE is tending towards extinction appears to be coupled to the rapid depletion of the radical pool at $x/D = 75$ relative to the solution employing modified Curl.

Given the significant effect that the use of a more elaborate scheme appears to have on the evolution of the reacting jet, figures 6.72-6.74 are presented in order to compare the two solutions associated with the ignition of the mixture at 10 and 15 diameters downstream of the burner. Figure 6.72 demonstrates that the peak mean temperature of the flow is steadily dropping for the calculation with ignition at $x/D = 10$. The large difference in temperature affects the density which in turn influences the mixture fraction; the jet with a *lower* overall density spreads faster. Figure 6.73 indicates that O₂ is higher for the flame that 'goes out', which is to be expected; the area under the curves for CH₄ appear to be much the same, however bearing in mind that the density is much higher further downstream for the flame initially ignited at $x/D = 10$, this indicates that less fuel is being consumed for that test case. The reason why the flame temperature drops monotonically when ignited at $x/D = 10$ can be explained using figure 6.74. This plot shows that whilst the peak values of CH₃ and H are the same at $x/D = 10$ or 15 (depending on the location of ignition), the subsequent evolution of the flame which cannot sustain itself is due to the depletion of the radical pool; by $x/D = 30$ there are hardly any radical species available. The only difference between the ignition of the mixture at either of the two specified locations is in the degree of interdiffusion between the fuel stream and the surrounding air stream.

The results in homogeneous, isotropic and reacting turbulence indicated that with the use of the systematically reduced mechanism the agreement between the predictions using the interpolation table and LSODE were not entirely satisfactory for all the scalar properties. For those calculations, as well as the laminar flame simulations, the direct integration of the stiff ODE's (using LSODE) did not lead to prohibitively long cpu times. For an inhomogeneous flow, the number of particles present and the number of steps required to perform the full calculation make the use of LSODE quite impractical. However, with this mechanism, ignition of the mixture at $x/D = 10$ leads to a progressive drop in the mean temperature. Since those 'notional' particles with a temperature

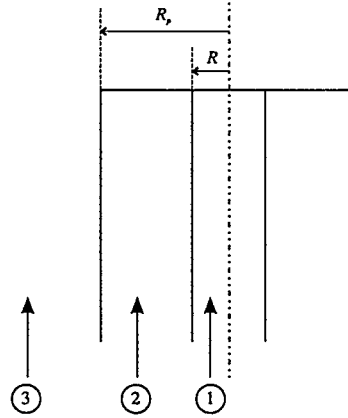


Figure 6.2: Schematic of the piloted methane-air turbulent jet diffusion flame of Masri et al. [66]. The streams labelled 1, 2 and 3 contain the following: 1. CH_4 at 298K, $Re \approx 20000$, $u_{in} = 41\text{ms}^{-1}$ (flame L) and $u_{in} = 48\text{ms}^{-1}$ (flame B); 2. 'Hot' (fully) burnt products from a stoichiometric mixture of C_2H_2 , H_2 and air, 2600K, $Re \approx 604$, $u_{in} = 24\text{ms}^{-1}$; 3. Coflowing air at 298K, $Re \approx 10^6$, $u_{in} = 15\text{ms}^{-1}$.

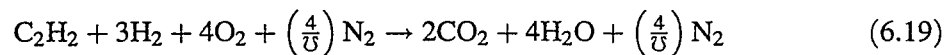
greater than 700K are only subjected to chemical reaction, this means that a comparison between the performance of a calculation using the interpolation table and the direct integration can be made. Such a test is clearly necessary given the implications of the above results which raise questions on the suitability of the reduced scheme in the calculation procedure. A calculation using LSODE for flame SA2 was performed up to $x/D = 30$ and even with full optimisation this required over three days of cpu time. With ignition at $x/D = 15$, the run time required for this calculation could easily be an order of magnitude higher. Figures 6.75 and 6.76 compare the solutions of the interpolation table and LSODE. The agreement is quite satisfactory in the sense that the correct qualitative trends are being predicted. Therefore the results presented earlier on are definitely not misleading, and for the gross flame characteristics (such as CH_4 , CO_2 and O_2 levels, not shown) the agreement is very good.

Further calculations using the scheme of Chelliah et al. [10] are not described in the context of the flame of Hassan et al. since it is felt that the uncertainties which have arisen through its use imply that it is not very well suited to the current formalism.

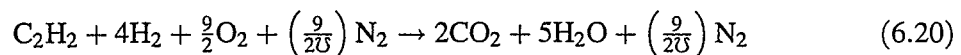
6.5.2 The Flame of Masri, Bilger and Dibble

The work of Masri et al. [66, 67, 68] provides extensive experimental data on the compositional structure and temperature for turbulent methane-air jet diffusion flames in which the chemical kinetic effects (e.g. local extinction) can be significant. A schematic of the basic experimental configuration is illustrated in figure 6.2.

The burner which produces the flame consists of a central jet of fuel surrounded by a large annulus (the pilot stream) of coflowing premixed combustion products (originating from a unburnt mixture of C_2H_2 , H_2 and air) with the carbon-to-hydrogen ratio adjusted to be the same as that of methane. This specification is not unique, for example the following one step irreversible reaction,



implies a carbon-to-hydrogen ratio of 1:4, whilst



ensures a carbon-to-hydrogen ratio 1:5. Therefore the amount of H_2 determines the carbon-to-hydrogen ratio. For the fully burnt mixture of acetylene and hydrogen the stoichiometric mixture fraction is given by:

$$\xi_{st} = \frac{W_{C_2H_2} + 3W_{H_2}}{2(W_{CO_2} + 4W_{H_2O} + \frac{2}{3}W_{N_2})} \approx 0.049 \quad (6.21)$$

An equilibrium calculation using the method of Gordon and McBride [32] for the annulus mixture yielded a temperature of about 2491K. When radical species are excluded from the equilibrium composition the 'fully-burnt' temperature rises to 2637K. Given that a real mixture contains radicals, it is not clear why Masri et al. estimate that the pilot stream is at 2600K.

The burner and pilot annulus are centred in a much larger coflowing stream of air. Masri et al. designed this arrangement in order to study turbulence-chemistry interactions under conditions approaching extinction, in a flow which is fluid dynamically 'simple'. By increasing the fuel jet velocity, the flame approached extinction in a region $15 \leq x/D \leq 35$, where D refers to the fuel jet diameter. At blow-off the heat release from the pilot annulus maintained combustion of the fuel and outside air over the first 15 diameters. It is pointed out [67] that in the vicinity of $x/D \approx 20$, where the stabilising effect of the pilot stream has virtually disappeared, the mixing is intense. If the fuel jet velocity is sufficiently high (prior to blow-off), the peak temperatures can drop to about 820K, implying local extinction; however further downstream ($x/D \approx 30$) the 'intensity' of mixing is reduced, the flame re-lights and the temperature increases again. It is quite clear that such a test case manifests appreciable finite-rate effects; it was experimentally observed that regardless of the mean jet velocity (and therefore how close the flame was to extinction), $(\xi''^2)^{\frac{1}{2}}$ remained constant at a given axial location. These conditions pose a severe test on any turbulence and thermochemical model employed to predict a turbulent combustions flows. Four different flame conditions were experimentally investigated by Masri et al. at various mean fuel inlet velocities. Flames 'L' and 'B', characterised by mean fuel inlet velocities of 41ms^{-1} and 48ms^{-1} respectively, are considered in this work. These flames are chosen partly because they encompass the range of behaviour previously discussed. In addition these flames have also been investigated by Chen et al. [15] and Vervisch [99, 100], and so a comparison of the predictions can be made.

Masri et al. [66] made simultaneous space and time resolved measurements of CH_4 , O_2 , N_2 , H_2O , H_2 , CO and CO_2 using pulsed spontaneous Raman scattering in the blue (visibly soot free) regions of the flames and temperature was measured from the Rayleigh scattered signal. A "fluorescence" interference which contaminated, in varying degrees, the Rayleigh and Raman lines was observed and its effect accounted for using correction coefficients generated from measurements in laminar flames of known composition. This problem seems to occur only in hydrocarbon fuels, particularly in the rich ($0.06 \leq \xi \leq 0.25$) regions of the flame for samples where the temperature is greater than 1273K. At each laser shot the Rayleigh scattering as well as the Raman signals from CH_4 , O_2 , N_2 , H_2O , H_2 , CO and CO_2 were collected simultaneously. The manner in which the statistical moments of these scalars were obtained, together with the influence of fluorescence is described in [66]. The species concentrations were expressed as density-weighted averages and the temperature and density as conventional, unweighted averages. The accuracy of the measurements was largely dependent on the severity of the fluorescence interference. The investigators claimed that the concentration and temperature measurements were affected by shot noise and calibration errors by less than 5% of the maximum values. In the pdf plots

typically 1250 sampled data points at a particular location were used to construct the histogram, including the ones with 'intense' fluorescence³. This leads to values of the pdf's of all the scalars being slightly biased towards higher values in the regions of intense fluorescence. The mixture fraction reported in [66] was calculated from an hydrogen-atom balance; the definition accounted for hydrogen atoms present in the air stream due to the presence of water vapour (quantified in terms of the specific humidity, Kg of H₂O/Kg dry air).

The pilot stream is significant in the role it plays in this flame configuration; the very low Reynolds number and high temperature of the flow issuing from the annulus indicates that it acts as a source of flame stabilisation. In addition, the shear size of the pilot stream sets it apart from the jet flames considered up to now, where the effect of the pilot stream was ignored in the calculation procedure. In the present context, the mixture is no longer artificially ignited at some arbitrarily located plane downstream of the burner. Instead the initial conditions associated with the pilot stream are introduced into the calculation procedure and it is expected that this procedure will be sufficient in ensuring subsequent chemical reaction further downstream.⁴ This procedure is undoubtedly more realistic than the artificial ignition. It should be borne in mind that if the pilot stream is not sufficiently large in comparison to the fuel stream (as in the previous jet flames) then such a practice may not be sufficient to ensure combustion downstream; effectively, the localised high temperature pilot gas would be quenched by the colder surroundings before sufficient mixing of fuel and air has taken place. This effect is a feature of the solution formalism and has nothing to do with what actually happens in reality. Regardless of the method of ignition that is employed, if the hot burnt products are represented in terms of a fully burnt mixture of CH₄/O₂/N₂ then the resulting peak temperature will be about 2400K, as opposed to 2600K arising from the complete burnout of a mixture of C₂H₂/H₂/O₂/N₂. For the initial set of predictions to be reported, the former specification is employed.

All the predictions presented for the flames of Masri et al. [66] employ the scheme of Jones and Lindstedt [46] (the global hydrocarbon scheme). Attempts at using the scheme of Chelliah et al. resulted in the same effects observed for the predictions of the flame of Hassan et al. [35] when ignited at $x/D = 10$. Igniting the mixture at a potentially large downstream position in order to ensure that the scheme of Chelliah et al. does work is not a potentially good idea in view of the fact that the measurements made by Masri et al. start quite close to the burner ($x/D = 10$).

The comparison of measurements and predictions for flame L will be considered first, since this particular flame does not exhibit the observed effects of local extinction and subsequent downstream relighting. Figure 6.77 compares the density-weighted CH₄ mass fraction and mixture fraction radial profiles. The behaviour of the predictions are consistent with what has been observed in the previous flames; namely that the mixing field and fuel breakdown are well represented, and that the differences arising out of the use of modified Curl and LMSE are quite small. The spreading rate of the predicted jet could be increased somewhat in order to conform more closely with the experimental measurements. Increased spreading rates could be achieved by increasing the characteristic turbulent length scales at inlet to the flow domain; this would in turn reduce ε which results in a larger eddy-viscosity (since $\mu_t \propto \frac{k}{\varepsilon}$). A larger eddy-viscosity implies increased turbulent mixing and hence spreading rate of the jet.

³An 'intense' signal is arbitrarily defined [67] as one where the maximum and minimum fluorescence corrections deviate by more than 25% from the mean fluorescence corrected value.

⁴The former method of specifying burnt particles at some arbitrary downstream location was adopted by Chen et al. [15], whilst Vervisch [99, 100] used the procedure which is also employed in this work.

The accuracy of the prediction of $\tilde{Y}_{N_2}$ (figure 6.78) is related to that of $\tilde{\xi}$ to the extent that both are conserved scalars and are affected by the reaction through the mean density. The mean density is very well predicted over a significant section of the jet. Towards the edges the overpredictions are presumably related to the fact that the jet is not spreading as fast as the measurements suggest. Figure 6.79 presents an interesting feature which has been discussed in the context of the flames of Godoy [31], Heitor et al. [37] and Hassan et al. [35] (sections 6.4.1, 6.4.2 and 6.5.1, respectively) - the appearance of O_2 at the centre-line. As with the previous flames, in the initial stages of jet evolution, the predictions do not indicate the existence of any O_2 near the centre-line. The profiles for CO_2 appear to be in good agreement at $x/D = 10$, but this deteriorates further downstream, although the peak levels are not too far off from those of the measurements. The fact that the accuracy of the predictions appear to be better at $x/D = 10$ than at the other measuring stations is a feature which is also observed in flame B; this aspect of the predictions will be discussed further after consideration of flame B.

Whilst the CO levels (figure 6.80) are underpredicted at $x/D = 10$, this is definitely not the case by $x/D = 30$. Furthermore the H_2 concentration (figure 6.81) is substantially overpredicted for $x/D \geq 20$. This final point is important since the measurements (and predictions) indicate that the peak levels of H_2 are an order of magnitude below those of CO (and both appear in relatively small amounts). This highlights the difficulty in reproducing the behaviour of these intermediate species which are strongly affected by how accurately the reaction zone is represented. The measurements of H_2O appear to be reproduced quite well. If the present calculation is extended to $x/D = 70$ it is noticed that by $x/D = 50$, $\tilde{Y}_{CO} \approx 0.11$ and $\tilde{Y}_{H_2} \approx 0.015$; by $x/D = 70$ $\tilde{Y}_{CO} \approx 0.13$ and $\tilde{Y}_{H_2} \approx 0.017$, with $\tilde{T}_{max} \approx 1600K$. This demonstrates that up to $x/D = 30$ only a brief 'history' of the flame is being observed and that as the flame evolves further downstream, the intermediate species' concentrations increase even further.

Figure 6.82 presents the prediction of the root-mean-square (rms) of CH_4 and ξ , denoted as $(\tilde{Y}_{CH_4}''^2)^{1/2}$ and $(\tilde{\xi}''^2)^{1/2}$ respectively. The correct trends are reproduced and the qualitative agreement is quite good, although there is scope for improvement. With reference to figure 6.77 it is noticed that the overpredictions in the rms values observed in the region $0.5 \geq r/R \leq 2$ ($R = D/2$) occur at the same location as the largest mean scalar gradients. Whether this is an indication that there is too much production of scalar fluctuations, or the mixing model does not dissipate the scalar fluctuations enough or even because it is a consequence of the spreading rate not being correctly predicted is an open question. Figure 6.83 indicates that at $x/D = 10$ the maximum and minimum (turning points) for $(\tilde{\rho}'^2)^{1/2}$ have been very well picked up by the predictions. This is quite an achievement given that the initial density field is quite complicated due to the presence of the hot pilot gas ($\rho_{pilot} \approx 0.145Kg/m^3$) located between the fuel ($\rho_{CH_4} \approx 0.656Kg/m^3$) and air ($\rho_{air} \approx 1.18Kg/m^3$) streams. Figure 6.84 demonstrates that both $(\tilde{Y}_{O_2}''^2)^{1/2}$ and $(\tilde{Y}_{CO_2}''^2)^{1/2}$ are quite well predicted; in the case of CO_2 this is interesting since the corresponding mean profiles (figure 6.79) are not too well predicted. Furthermore, despite the fact that $\tilde{Y}_{O_2}$ indicates that there is no predicted centre-line oxygen, small (yet finite) fluctuation levels are detected at $x/D = 20$. Unlike the behaviour of the mean and rms of CO_2 , figures 6.85 and 6.86 show that the overpredictions in $\tilde{Y}_{CO}$ and $\tilde{Y}_{H_2}$ lead to overpredictions in the corresponding rms values; this effect is particularly acute in the case of $(\tilde{Y}_{H_2}''^2)^{1/2}$. The predictions of $(\tilde{T}'^2)^{1/2}$ at $x/D = 10$, figure 6.85, indicate a peak at $r/R \approx 1$ whereas the measurements suggest a monotonic rise. The reason for this is not entirely clear; the prediction of $(\tilde{Y}_{H_2O}''^2)^{1/2}$ (figure 6.86) at $x/D = 10$ also indicates a peak in this region, and

it is possible that the calculation procedure registers this feature as a residual effect of the pilot stream since this peak appears to disappear by about $x/D = 20$.

Since the current method of solution allows the pdf to be treated as an evolving quantity, it seems natural to present some pdf's and compare them against the measurements compiled by Masri et al. [67]. Figure 6.87 presents the *marginal* pdf $\tilde{P}(\hat{\xi}; x, \omega)$ at $x/D = 20$ for three different radial positions. Before discussing the level of agreement which exists between the plots, the reason why the measurements and predictions are compared at $r/R = 1.97$ and $r/R = 1.98$ (as an example) needs to be elaborated. With the calculation procedure being used the grid position is outside the control of the analyst, and is evaluated in terms of the transformation to dimensionless stream function coordinates. These details are discussed in section 5 and appendix D. However, provided the mesh is sufficiently fine in the ω -direction, upon transformation to x - r coordinates, a grid node in physical space close to the location of the measurements can usually be found, but a one-to-one correspondence cannot be guaranteed.

Despite the differences in the fine details of the pdf profiles in figure 6.87, the predicted and measured pdf's are located in the same regions of the $\hat{\xi}$ space; near the centre-line the mixture is rich and the pdf's are closer to $\hat{\xi} = 1$, whilst at $r/R = 3.11$ the pdf's are biased towards $\hat{\xi} = 0$. Given that $\tilde{\xi}$ has been qualitatively well-predicted, the observations concerning the location of the pdf's is to be expected. It was mentioned earlier on that the predicted spreading rate of the jet is less than the actual spreading rate experimentally measured. Consequently at $r/R = 0$ the predicted pdf is closer to $\hat{\xi} = 1$ than the measured pdf. Similarly at $r/R = 3.11$ the predicted pdf shows a much stronger spike at $\hat{\xi} = 0$ than the measured pdf. At $x/D = 20$ and $r/R = 0$, figure 6.82 indicates that the predicted and measured values of $(\tilde{\xi}''^2)^{1/2}$ are virtually the same, although figure 6.77 demonstrates that at the same location the values of $\tilde{\xi}$ are different. However, the corresponding pdf profiles in figure 6.87 have different shapes - if only $\tilde{\xi}$ was different they would look identical but situated at different locations along the $\hat{\xi}$ axis. This suggests that the higher-order moments may also be different in order to account for the differences in the shapes of the pdf. This particular feature is further exemplified in figure 6.88 which presents $\tilde{P}(\hat{\xi}; x, \omega)$ due to the LMSE model; at $r/R = 0$ the differences between the predicted pdf's in figures 6.87 and 6.88 are noticeable, yet the mean and rms show little difference between the two mixing models. Figure 6.89 compares the *unweighted* pdf $P(\hat{T}; x, \omega)$. Qualitatively the agreement appears to be quite good and this is reflected to a certain extent by the mean and rms profiles (figures 6.80 and 6.85 at $x/D = 20$). At $x/D = 20$ and $r/R \approx 2$, $\bar{T} \approx 1150\text{K}$ and $(\bar{T}'^2)^{1/2} \approx 200\text{--}400\text{K}$ (measurement and prediction, respectively); it is interesting to note that both the measured and predicted pdf's register signals up to 2000K, albeit with much lower intensity. The importance of this result has been highlighted by Masri et al. [67], who point out that whilst the frequency of encountering 'hot' burnt pockets of gas decreases sharply as the flame approaches extinction, their very existence may be responsible for keeping the flame alight - this argument is particularly relevant to flame B where local extinction is followed by relighting of the flame further downstream. Figure 6.90 shows $\tilde{P}(\hat{Y}_{\text{CO}}; x, \omega)$; the results are consistent with the profiles of $\hat{Y}_{\text{CO}}$ (figure 6.80) since at $x/D = 20$ and $r/R = 0$ the mean CO levels are underpredicted and at $r/R \approx 2$ they are overpredicted.

The predictions for flame B are now discussed. The only difference between flame L and flame B is the initial mean fuel jet velocity; a difference of 7ms^{-1} . Figure 6.91 shows the radial profiles for $\tilde{\xi}$ and $\tilde{Y}_{\text{CH}_4}$. It is clear that up to about $x/D = 30$ both mixing models provide a very good representation of $\tilde{\xi}$, particularly at $x/D = 20$ where the mixing is claimed [66] to be intense and

the conditions for combustion are critical. By $x/D = 50$ differences emerge between LMSE and modified Curl, but they do not appear to be significant. Figure 6.92 demonstrates that despite the small effect on the conserved scalar field ($\tilde{\xi}$ and $\tilde{Y}_{N_2}$), there are substantial differences in the mean density based on the two mixing models. Using LMSE the density is consistently rising, implying that the temperature is dropping. This is the first instance in the calculation of jet flames using the scheme of Jones and Lindstedt [46] that the solutions employing modified Curl and LMSE differ so drastically. Figures 6.93–6.95 show how the remainder of the reactive scalars behave. The most informative plot is figure 6.94, which demonstrates the temperature evolution. The measurements indicate that the peak temperature at $x/D = 20$ is about 900K; this is clearly the region in which the flame is locally experiencing extinction, yet the calculation procedure has not picked up this feature at all. In this region both LMSE and modified Curl yield very similar results and it is only for $x/D \geq 20$ that differences emerge. LMSE is clearly indicating a continual drop in the peak temperature, consistent with the density profiles discussed earlier. In conjunction with this drop in temperature, $\tilde{Y}_{CO}$, $\tilde{Y}_{H_2}$, $\tilde{Y}_{H_2O}$ and $\tilde{Y}_{CO_2}$ all drop substantially (relative to the solution using modified Curl); this is matched by a rise in the level of $\tilde{Y}_{O_2}$ across the jet. The behaviour of $\tilde{Y}_{CH_4}$ (figure 6.91) at $x/D = 50$ indicates that there is slightly *less* fuel present using LMSE than with modified Curl. As with the flame of Hassan et al. [35] discussed earlier, this paradoxical observation is explained by the difference in the density fields which demonstrates that the actual mass of fuel present at $x/D = 50$ is greater with LMSE.

It is interesting to note that the large differences observed between the results of the application of the two mixing models is also apparent in the calculations of Chen et al. [15] with the use of modified Curl and two different reaction schemes. In their work, a systematically reduced five-scalar, four-step mechanism was employed (involving a single H-radical) as well as a four-scalar constrained equilibrium scheme (developed on heuristic arguments). Their mean temperature profiles indicate that with both chemical schemes (and for flames L and B) the peak temperatures drop as x/D increases; for the constrained equilibrium scheme this occurs more rapidly, such that with flame B at $x/D = 50$, $\bar{T}_{max} = 850K$. The effect of local extinction (observed with flame B) is not registered at all, with peak temperatures of about 1600K being predicted at $x/D = 20$ (with both schemes) whilst in figure 6.94, $\bar{T}_{max} = 1400K$ (which is slightly closer to the measured $\bar{T}_{max} \approx 900K$). In going from $x/D = 30$ to $x/D = 50$ the predictions of Chen et al. indicate a *drop* in peak temperature from about 1300K (for the five-scalar reduced scheme) to 1200K, whereas in figure 6.94 a slight *rise* in temperature is predicted with modified Curl, consistent with the experimental trend. Unfortunately Chen et al. [15] do not present their predictions of CO and H₂; however Vervisch [99] does report large overpredictions of CO and H₂ (comparable with those reported in this thesis) in flame B using the scheme of Jones and Lindstedt [46]. The calculations employed LMSE and the results did not indicate the drop in temperature observed in figure 6.94. The reasons for this discrepancy are not clear.

Another interesting area for comparison is related to the predictions of H₂O. Chen et al. [15] argue that as the jet velocity is increased, $\tilde{Y}_{H_2O}$ should be lower due to reduced flow residence times. This effect was not observed in the jet flames discussed in the previous sections. However, in the flame of Masri et al. [66], where an increase in the central fuel jet velocity can lead to local extinction, it is clear that the Reynolds number will influence the flow properties quite significantly. The measurements of Masri et al. confirm the reduction in the levels of H₂O as the fuel jet velocity increases. The predictions of Chen et al. indicate the opposite trend, i.e. an increase in $\tilde{Y}_{H_2O}$ in going from flame L to B. It is suggested [15] that this tendency for approaching equilibrium

with increasing strain rate highlights the deficient behaviour of modified Curl in modelling the molecular mixing effects. More specifically, the shortcomings of modified Curl are attributed to its discontinuous representation of the mixing process - hence the motivation for reaction zone conditioning (described in section 4.7.4). However the results in figures 6.81 and 6.95 indicate that with modified Curl the levels of H_2O predicted in flame B are either the same or *less* than those in flame L. This result is at variance with the conclusions of Chen et al. [15], and since the difference between these two sets of results lies in the chemical mechanism employed, this suggests that both the mixing model and the chemistry may limit the scalar pdf approach. Due to the strong coupling which naturally exists between mixing and reaction, it is expected that sub-models used to represent either of these processes will, in general, influence the overall results.

For completeness, some of the predictions of the fluctuating field of flame B are also presented. The predictions of the pdf's are not included since it is difficult to extract information from them which could elucidate the reasons behind the current trends in the predictions. Figure 6.96 shows the predictions and measurements of $(\xi''^2)^{\frac{1}{2}}$ and $(Y_{\text{CH}_4}'^2)^{\frac{1}{2}}$. It is encouraging to note that the (experimentally observed) insensitivity of $(\xi''^2)^{\frac{1}{2}}$ to the initial mean jet velocity (referred to earlier on) has also been reproduced by the predictions, as shown by figures 6.82 and 6.96. In the radial profile of $(\xi''^2)^{\frac{1}{2}}$ Chen et al. [15] predict a second peak at $x/D = 10$ (typically 10% of the maximum value) when $r/R = 4$ and attribute this to the residual effects of the simulated pilot flame. However, figure 6.96 and the results of Vervisch [99] do not indicate such a peak. This may be due to the fact that in the latter examples the pilot flame is simulated directly in terms of the initial conditions. Figure 6.97 demonstrates that with modified Curl $(\rho'^2)^{\frac{1}{2}}$ is predicted very well. The level of agreement between prediction and measurements is better than in [15], where broader profiles are obtained; for example at $x/D = 10$, the peak value of $(\rho'^2)^{\frac{1}{2}}$ is predicted to occur at $r/R \approx 5$ in [15]. With LMSE the peak value of $(\rho'^2)^{\frac{1}{2}}$ drops as the mean density rises. The peak also moves towards the centre-line as more O_2 diffuses into the jet and the flame progressively goes out. The remainder of the scalar fluctuation predictions are not shown; however, for modified Curl the same qualitative features as predicted in flame L are observed. In particular the significant overprediction of $(Y_{\text{CO}}''^2)^{\frac{1}{2}}$ and $(Y_{\text{H}_2}''^2)^{\frac{1}{2}}$. In contrast LMSE predicts a decaying fluctuating field as a function of x/D .

In the remainder of this section the test case of flame B will be analysed subject to various conditions. The results will demonstrate several interesting aspects of the predictions. The first test is a calculation which cannot be realised in a laboratory and involves the simulation of flame B with chemical reaction switched off. The same initial conditions are employed, but no CO and H_2 can be produced and the only H_2O and CO_2 originate from the pilot stream. Figures 6.98-6.99 show the results using modified Curl. The solid lines have already been discussed in the paragraphs above. The most striking observation is that switching off the reaction has little effect on the evolution of CH_4 , and for the temperature the discrepancy is very small at $x/D = 10$. Therefore up to $x/D = 10$ the effect of the pilot stream is very significant and almost dominates the evolution of the flow. This goes some way towards explaining why the predictions appear to be so accurate at $x/D = 10$ in the results of flames L and B.

It was mentioned earlier on when discussing the representation of the pilot stream in terms of a fully-burnt stoichiometric mixture of CH_4/air that the equilibrium temperature is typically 2400K, which is about 200K less than the actual temperature of the pilot stream which contains a fully-burnt stoichiometric mixture of $\text{C}_2\text{H}_2/\text{H}_2/\text{air}$. Consequently the pilot flame should be

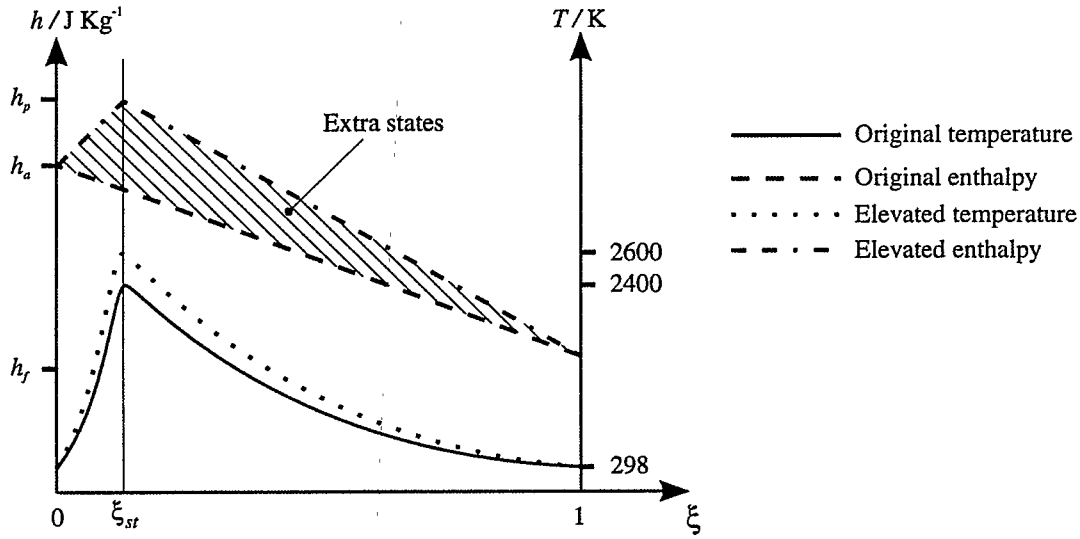


Figure 6.3: This figure illustrates the effect of introducing the enthalpy as an extra scalar in the manner proposed by Sion and Chen [86]. The 'original' temperature allows for a peak (stoichiometric) value of 2400K, whilst the addition of the 'elevated' enthalpy raises this to 2600K. Extra compositional states are also introduced which need to be accounted for when constructing the bounds for the interpolation table.

considered as a third stream, the other two being the central fuel jet and the coflowing air. In order to avoid the description of a three-stream mixing process, the enthalpy (h) can be incorporated as an extra independent scalar (in addition to ξ , n_{CH_4} , n_{CO} and n_{H_2}) in the manner described by Sion and Chen [86]. Briefly, the pilot stream is still considered as combustion products arising from a CH_4/air mixture, only now the *initial* air and fuel temperatures leading to the formation of the fully-burnt products are changed from 298K to 678K - this ensures that the equilibrium temperature of the combustion products increases from 2400K to 2600K. Henceforth, the 'elevated' enthalpy is referred to as h_p (where $\xi = \xi_{st}$), and the enthalpy of the air and fuel streams are designated as h_a ($\xi = 0$) and h_f ($\xi = 1$) respectively. For a two-stream mixing process h is linearly related to ξ . Up to now the enthalpy (symbolised by h_o) has been specified through ξ by,

$$h_o = h_f \xi + (1 - \xi) h_a \quad (6.22)$$

If $0 \leq \xi \leq \xi_{st}$, the linearity requirements imply that,

$$h_l = \frac{\xi}{\xi_{st}} h_p + \left(1 - \frac{\xi}{\xi_{st}}\right) h_a \quad (6.23)$$

and if $\xi_{st} \leq \xi \leq 1$,

$$h_r = \left(\frac{1 - \xi}{1 - \xi_{st}}\right) h_p + \left(\frac{\xi - \xi_{st}}{1 - \xi_{st}}\right) h_f \quad (6.24)$$

Equations (6.23) and (6.24) are obtained by straightforward linear interpolation between the states $[(h_a, 0), (h_p, \xi_{st})]$ and $[(h_p, \xi_{st}), (h_f, 1)]$ respectively. Consequently the bounds for the enthalpy become, $h \in \{h_l, h_o\}$ for lean mixtures and $h \in \{h_r, h_o\}$ for rich mixtures. Evaluation of h_p , h_f and h_a using JANAF polynomials indicate that $h_a, h_f \leq 0$ and $h_p \geq 0$ so that $h_o \leq h_l$ and $h_o \leq h_r$. The effect of introducing enthalpy as an extra scalar is illustrated schematically in figure 6.3.

The effect of this thermochemical representation on the predictions of flame L are shown in figure 6.100. The effect of the higher initial temperature can be clearly detected in figure 6.100 and this leads to slightly higher values of CO (as well as H₂O and H₂, not shown). The modification leading to the introduction of enthalpy as an extra scalar does not appear to yield better predictions, at least at this level of closure. Flame B, figure 6.101, shows little change in comparison to figure 6.94 as a result of these modifications - it was originally thought that with the pilot stream at 2600K, the results using LMSE would no longer predict extinction over the region $30 \leq x/D \leq 50$.

Another interesting test is to calculate flame B using a Reynolds stress model. Based on the results of the Godoy flame, there should be little effect on the results. Figure 6.102 shows the results for $\tilde{Y}_{\text{CH}_4}$ and $\tilde{\xi}$. Comparing this with figure 6.91 (where the $k-\epsilon$ model is employed) indicates that with the Reynolds stress model the jet is spreading more slowly. Figure 6.103 indicates that the effect on the predictions of LMSE are quite significant. Basically by spreading less rapidly, less oxygen is entrained and so it appears that this avoids the quenching of the flame zone. With flame L, the application of the Reynolds stress model yields much the same result as the $k-\epsilon$ model. It is argued that because of the appreciable finite-rate effects which are observed in flame B, the predictions appear to be quite sensitive to features such as the spreading rate, which would not affect the other flames discussed thus far. Fairweather et al. [27] have indicated that they obtained improved predictions for the spreading rate by adjusting the value of C_{ϵ_2} from 1.9 (the value given in table 2.1) to 1.8. In the case of the $k-\epsilon$ model it was pointed out in section 2.5.1 that for a round jet issuing into *stagnant* surrounds improved spreading rates can be obtained with $C_{\epsilon_2} = 1.85$, although the 'standard' value is usually quoted to be 1.92. With the current Reynolds stress model a similar situation seems to exist. Furthermore, the predictions of the Godoy flame (figures 6.52-6.54) and the results presented in figures 6.102 and 6.103 employed $C_{\epsilon_2} = 1.8$. However if C_{ϵ_2} were to be increased to 1.9, this would lead to more 'destruction' of ϵ based on the modelled form of the ϵ -equation (2.54). This means that locally ϵ would be smaller in magnitude, and so the turbulent mixing rates would be enhanced, thereby increasing the spreading rate. Figure 6.104 demonstrates the improvement in the spreading rate of the jet as C_{ϵ_2} is increased from 1.8 to 1.9. However, figure 6.105 shows that the improvement in the $\tilde{\xi}$ and $\tilde{Y}_{\text{CH}_4}$ fields lead to the same trends for $\tilde{Y}_{\text{CO}}$ and $\bar{T}$ observed with the use of $k-\epsilon$. Changing C_{ϵ_2} by about 5% leads to significantly different results for LMSE, and this raises questions with regards to the suitability of the current level of closure in reproducing finite-rate effects such as extinction and re-ignition.

The final test to be considered is the influence of the constant C_D appearing in the LMSE model. Calculations were performed with $C_D = 2$ and $C_D = 0.5$, i.e. doubled and halved relative to the original value of $C_D = 1$. The effect of this change is quite profound as shown in figures 6.106 for the case of flame B; increasing C_D appears to result in a faster approach to equilibrium. In essence, increasing C_D implies that the mixture has homogenised more effectively so that combustion can proceed more efficiently. The improved profiles of $\tilde{Y}_{\text{CO}}$ with $C_D = 0.5$ is offset by the very poor prediction of $\bar{T}$. These results are consistent with those of Chen et al. [15] who increased the relevant 'constant' in modified Curl by 10% and also observed a faster approach to equilibrium. This effect is not just confined to flame B; figure 6.107 shows the effect of doubling and halving C_D for flame L. The differences are clearly noticeable and by $x/D = 30$ the mean temperature for the calculation employing $C_D = 0.5$ indicates that the flame is going out.

Chapter 7

Closure

7.1 Summary

In this thesis the modelling approaches for predicting single phase, gaseous, low Mach number and high Reynolds number turbulent combustion have been discussed for non-premixed configurations.

Scalar pdf modelling at the single-point and single-time level constitutes the simplest form of closure for the statistics of the scalar field. The primary appeal of such an approach is that realistic chemistry can be explicitly incorporated without introducing assumptions about the instantaneous flame structure (as in the conserved scalar formalism). More elaborate proposals have been suggested to augment the pdf closure, however, they (necessarily) introduce further unknowns and their evaluation would require substantially greater cpu capabilities. It appears, at least for the time being, that a combination of a phenomenological turbulence model coupled to a modelled scalar pdf evolution equation is the most practical level of closure since the merits of the respective approaches in calculating the velocity and the scalar field can be exploited.

In scalar pdf modelling, closure for the molecular mixing term is required. The molecular mixing term is central to the turbulence-chemistry interactions primarily because it involves molecular diffusivities and compositional gradients, which scale with the dissipative motions in a turbulent flow, and this is precisely where combustion occurs. The most widely used class of mixing models are those based on binary interaction of fluid parcels (coalescence-dispersion closures), and linear mean square estimation (LMSE). In addition to their poor representation of pdf evolution in homogeneous, isotropic and inert turbulence, these mixing models have a serious drawback, ultimately related to the fact that they employ k/ϵ as the (only) characteristic turbulent time scale; a time scale representative of the large scale energy-containing motions. Furthermore the diffusivities of the reacting scalars are taken to be effectively uniform, whilst it is well-known that species such as H_2 and H diffuse considerably faster than others by virtue of their significantly lower molar masses. Consequently these models do not address the problem of mixing in the presence of a flame front where information concerning local scalar gradients and molecular diffusivities can be important.

The main objective of the work described in this thesis was to apply the transported pdf approach to calculate the evolution of hydrocarbon-air turbulent jet diffusion flames. In particular, the study focused on the prediction of the levels of CO and in assessing how well the current formalism can reproduce finite-rate effects, such as local extinction. Calculations of turbulent jet diffusion flames were performed. The velocity field was approximated using either the $k-\epsilon$

or a full Reynolds stress closure and solved using a finite-difference procedure. The scalar field was represented through the modelled evolution equation for the scalar pdf and solved using a Monte Carlo simulation. The pdf equation employed gradient transport modelling to represent the turbulent diffusion, and the molecular mixing term was modelled in terms of the LMSE and coalescence-dispersion closures. The source terms for chemical reaction were represented via a global and systematically reduced scheme. The thermochemistry was tabulated on an once and for all basis and the results stored in a look-up table; multi-linear interpolation was employed in order to extract the necessary information from the tables. In order to assess the transported pdf technique for the inhomogeneous flow calculations, investigations of homogeneous, isotropic turbulence and a laminar jet diffusion flame were also undertaken. In the next section the conclusions relating to the results of these calculations are discussed.

7.2 Conclusions

7.2.1 Laminar Flame Calculations

The simulation of a confined, laminar, CH_4/air jet diffusion flame proved to be a very useful way of testing the interpolation technique used in conjunction with the tabulation of the thermochemistry. The interpolation (look-up) tables for the global mechanism and the systematically reduced scheme demonstrated that they were capable of representing the chemistry accurately and efficiently. More specifically for the global scheme the largest observable discrepancy between the interpolated result and the solution using LSODE was about 2 to 3%. With the systematically reduced scheme the same was true for the major species (CH_4 , O_2 , CO_2 and H_2O), but the margin of difference was slightly greater for CO and H_2 (5%) and for the radicals it was (at worst) about 20%. The reason why the look-up table for the systematically reduced scheme yielded a poorer result can be attributed partly to the fact that with this chemical scheme more scalars are involved and so the table is substantially larger; the coarseness of the scalar grids may well be a factor, particularly with regard to the radical species.

The laminar flame calculation is of theoretical importance since it demonstrates how sensitive the results are to the treatment of the molecular diffusion. This was demonstrated by the substantial overprediction of CO and H_2 when equal diffusivity of the scalars was assumed; an effect which is not observed when differential diffusion effects are incorporated in the theoretical description. Whilst the global scheme predicted levels of CO and H_2 to be around 10 and 13% mole fraction (respectively), the systematically reduced scheme predicted 7 and 10%. The reason why the *latter* scheme predicts smaller levels of CO and H_2 is related to the poorer representation of the fuel consumption. Moreover, the fuel consumption step in this reduced scheme occurs through the attack of the H-radical and since this specie has a diffusivity substantially higher than the rest, it is not surprising that the results are affected so dramatically when the diffusivities all take the same value.

7.2.2 Homogeneous, Isotropic Turbulence

The most notable feature of the homogeneous, isotropic turbulence calculations was associated with the reacting test cases. In such flows the coupling of mixing and reaction can be investigated in some detail, and certain important aspects of this coupling were identified. Calculations were

performed using two sets of initial conditions. These were an initial pdf corresponding to a double δ -function in mixture fraction space (suitably weighted so that the mean was at stoichiometric), and a continuous initial distribution; the latter pdf distribution was obtained from the evaluation of the former after a period of five eddy turn-over times.

In the case of the global scheme the results using LMSE and a discontinuous initial condition clearly demonstrated a pathological evolution, whilst modified Curl yielded physically more plausible results. When subject to a continuous initial condition both models yielded results where the differences were hardly noticeable. This suggests that the pathological behaviour associated with the pdf evolution using LMSE is only significant when discontinuous distributions prevail. Comparisons of calculations with LSODE and the interpolation table were very encouraging, indicating that a high degree of confidence could be placed in tests which use the interpolation technique.

With the systematically reduced scheme, ignition could not be achieved unless hot stoichiometric products were also present. A comparison of LSODE and the interpolation technique indicated acceptable agreement for the major species and temperature, but for CO, H₂ and the radicals there were large discrepancies, and sometimes different qualitative trends were observed. This may suggest that, in this situation, regions of the interpolation table are being accessed which are different to those encountered in the laminar flame calculations (which yielded acceptable results with regards to the interpolation) - the effect of the mixing obviously has a strong effect on where the composition state will be located. To construct the bounds for the allowable scalar domain, artificial clipping of the maximum levels of CH₃ and H were imposed in order to avoid tabulating pathological states which would involve unrealistically high levels of radical concentrations. The conditions in a real flame would never reach such composition states, and so their tabulation would be a wasteful use of the table. It is highly unlikely that such a crude form of clipping avoids all the possible pathological states. More importantly, the general applicability of this reduced scheme hinges on steady-state approximations used as part of the systematic reduction from a detailed mechanism. Therefore the relevance of this scheme comes under question when applied to situations where this approximation may not be valid.

7.2.3 Turbulent Jet Diffusion Flames

Initially the discussion will focus on the results using the global scheme, and for those flames in which extinction effects were not experimentally observed. The similarity of the predictions using LMSE and modified Curl was consistent with the observations in homogeneous, isotropic and reacting turbulence. Generally speaking overpredictions of CO and H₂ were obtained (particularly in the peak values). Their levels were of the same order as those predicted in the laminar flames with the assumption of equal diffusivity. This partly implies that to answer why the CO levels (for example) are overpredicted in the turbulent flame, it is necessary to know why they are overpredicted in the laminar flame. The overprediction of CO is not a universally observed characteristic of the application of the pdf technique to hydrocarbon-air jet diffusion flames. Results from other investigators [44, 12] show that the use of constrained equilibrium chemistry with modified Curl leads to underpredictions of CO; however, constrained equilibrium schemes do not appear to work well for laminar flames and so it is questionable how useful they can be in turbulent flames.

Despite the shortcomings of the current level of closure in reproducing the behaviour of quanti-

ties which are sensitive to the details of the reaction zone (i.e. CO and H₂), the formalism is capable of reproducing many of the gross flame characteristics and achieving this by treating the chemistry explicitly. A considerable amount of statistical information relating to the scalar field has been obtained through the solution of one equation where the variable is $\tilde{P}(\hat{\xi}, \hat{n}_{C_xH_y}, \hat{n}_{CO}, \hat{n}_{H_2}; x, \omega)$. This is a remarkable achievement given that with the moment closure approach each of the statistical variables would have to be obtained through the solution of its own modelled transport equation.

The predictions indicated that the velocity field was also well reproduced (based on the measurements in [31, 37]). This is an important result since it goes some way towards eliminating some of the uncertainties concerning the accuracy of the calculation procedure. For example, since the fluctuating velocity field was well represented it is possible to conclude that k and ε are also relatively well represented in so far as the input to the mixing model via $\frac{k}{\varepsilon}$ is concerned.

The shortcomings of the systematically reduced scheme in the transported pdf formalism come to the fore in the jet flame calculations. The plane of 'ignition' needed to be placed at about fifteen burner diameters from the burner exit in order to ensure subsequent downstream combustion - this is an unacceptably large distance. Even with ignition at ten diameters the flame temperature monotonically decreased, implying global extinction. This clearly raises questions concerning the suitability of this reaction scheme for turbulent flame calculations where the chemistry is treated explicitly.

Calculations of flames subjected to local extinction and subsequent downstream relighting, [66] (flame B), were analysed using only the global scheme. This was the first instance where significant differences between the results of LMSE and modified Curl were observed in the jet flame configuration. Such differences were also apparent in the work of Chen et al. [15] when using modified Curl and two different reaction schemes. This implies that both the chemistry and the mixing seem to limit the transported scalar pdf approach when appreciable finite-rate effects are present. Since the mixing and reaction are intricately coupled, a substantial improvement in the representation of only one of these processes may not be sufficient in bringing about a significant improvement in the accuracy of the predictions. The sensitivity of the results to the value of C_{ε_2} (appearing in equation (2.54)) was particularly pronounced in flame B when using LMSE; an increase of 5% in C_{ε_2} led to increases in the spreading rate such that the colder surrounding air helped to quench the flame zone. The suitability of the current level of closure in addressing the problem of turbulent flame extinction does come under question; but this has to be balanced against the fact that flames near extinction are very sensitive to small changes in certain parameters, such as the spreading rate, and this result may simply be a manifestation of such an effect.

The effect of C_D using the LMSE model for flames L and B also had an important effect - a two-fold increase in its value increased the degree of homogenization and thereby allowed combustion to proceed more effectively. This result is interesting since variations in C_D can be loosely interpreted as an indirect way of incorporating differential diffusion effects into the mixing model. It is not being suggested that the current mixing models (LMSE and modified Curl) should be 'improved' by allowing their 'constants to be variable'. However, it underpins the need to improve mixing models so that some degree of preferential mixing can be incorporated.

7.3 Recommendations for Future Work

The results presented in this thesis suggest that the accurate representation of the details of the fine scales of turbulent motion may well be necessary in order to predict effects such as CO emissions, flame extinction and ignition. This may seem to be an obvious conclusion, however, the number of theories (or models) which have been (and are being) proposed do not appear to address this issue, or at least make it their modelling priority.

The results presented in this thesis essentially relate to one type of flow - turbulent jet diffusion flames. Consequently the investigation of transported pdf techniques in other geometries would be useful to ascertain the generality of the observations made in the present contribution. This is a relatively short term recommendation; the ultimate objective would have to be the improvement of the description of the turbulence-chemistry interactions. Recently several studies on 'improved' mixing models (e.g. binomial sampling, mapping closures) have appeared which ensure that an arbitrary initial passive scalar pdf relaxes to a Gaussian pdf in a homogeneous turbulence field. However, the results presented here suggest that relaxation to a Gaussian pdf, although desirable, may not be the primary factor in accurately predicting turbulent flame properties where the reactive scalar pdf's can be far from Gaussian. If this is the case then these newer mixing models may not yield improved CO and H₂ predictions unless a more detailed description of the fine scales is incorporated.

Suggestions are increasingly made that the use of the joint pdf of the scalar and its gradient represents a promising approach and alleviates the problem of evaluating turbulent scalar time scales. However, it must be borne in mind that even when the hurdle of approximating the non-closed terms in this equation is crossed, the dimensionality becomes another consideration. For the four-scalar chemistry and two-dimensional axisymmetric flows considered in this thesis, $(4+2 \times 4 =)$ 12 independent scalars would be required. Tabulation of the chemistry would be out of the question, and parallelization would be essential. However, if substantially improved predictions can be obtained then the intensive computational requirements could be justified.

Bibliography

- [1] R. Aris. *Vectors, Tensors and the Basic Equations of Fluid Mechanics*. Prentice-Hall, 1962. Prentice-Hall International Series in the Physical and Chemical Engineering Sciences.
- [2] R. Armstrong, M.I. Koszykowski, F. Dai, J.-Y. Chen, N.J. Brown, and J. Macfarlane. Full chemistry in turbulent combustion dynamics. In *Abstracts from Fifth International Conference on Numerical Combustion*, page 11, Garmisch-Partenkirchen, Germany, September 29 - October 1 1993.
- [3] R.W. Bilger. The structure of diffusion flames. *Combustion Science and Technology*, 13:155–170, 1976.
- [4] R.W. Bilger. Turbulent jet diffusion flames. *Progress in Energy and Combustion Science*, 1:87–109, 1976.
- [5] R.W. Bilger. Reaction rates in diffusion flames. *Combustion and Flame*, 30:277–284, 1977.
- [6] R.W. Bilger. Turbulent flows with non-premixed reactants. In P.A. Libby and F.A. Williams, editors, *Turbulent Reacting Flows*, pages 65–113. Springer-Verlag, 1980. Appeared under 'Topics in Applied Physics', vol.44.
- [7] R.B. Bird, W.E. Stewart, and E.N. Lightfoot. *Transport Phenomena*. John Wiley & Sons, Inc., 1960.
- [8] P. Bradshaw. *An Introduction to Turbulence and its Measurement*. Pergamon, 1971.
- [9] S.P. Burke and T.E.W. Schumann. Diffusion flames. *Industrial and Engineering Chemistry*, 20(10):998–1004, 1928.
- [10] H.K. Chelliah, K. Seshadri, and C.K. Law. Reduced kinetic mechanisms for counterflow methane-air diffusion flames. In N. Peters and B. Rogg, editors, *Reduced kinetic mechanisms for applications in combustion systems*, chapter 13, pages 224–240. Springer-Verlag, 1993. Lecture notes in Physics m15.
- [11] H. Chen, S. Chen, and R.H. Kraichnan. Probability distribution of a stochastically advected scalar field. *Physical Review Letters*, 63(24):2657–2660, December 1989.
- [12] J.-Y. Chen and W. Kollmann. Pdf modelling of chemical non-equilibrium effects in turbulent non-premixed hydrocarbon flames. In *22nd Symposium (International) on Combustion / The Combustion Institute*, pages 645–653, 1988.

- [13] J.-Y. Chen and W. Kollmann. Analysis of mixing models. In *Eighth Symposium on Turbulent Shear Flows*, Technical University of Munich, September 9-11 1991.
- [14] J.-Y. Chen and W. Kollmann. Mixing models for turbulent flows with exothermic reactions. In *Turbulent Shear Flows 7*, pages 277–292. Springer-Verlag, 1991.
- [15] J.-Y. Chen, W. Kollmann, and R.W. Dibble. Pdf modelling of turbulent non-premixed methane jet flames. *Combustion Science and Technology*, 64:315–346, 1989.
- [16] P.Y. Chou. On velocity correlation and the solution of the equation of turbulent fluctuation. *Quarterly of Applied Mathematics*, 3:38, 1945.
- [17] R.L. Curl. Dispersed phase mixing : 1. Theory and effects in simple reactors. *A.I.Ch.E. Journal*, 9(2):175–181, 1963.
- [18] B.J. Daly and F.H. Harlow. Transport equations in turbulence. *Physics of Fluids*, 13:2634, 1970.
- [19] C. Dopazo. *Non-isothermal turbulent reactive flows : stochastic approaches*. PhD thesis, State University of New York at Stony Brook, 1973.
- [20] C. Dopazo. Probability density function approach for a turbulent axisymmetric heated jet. Centreline evolution. *Physics of Fluids*, 18(4):397–404, 1975.
- [21] C. Dopazo. Relaxation of initial probability density functions in the turbulent convection of scalar fields. *Physics of Fluids*, 22(1):20–30, 1979.
- [22] C. Dopazo and E.E. O'Brien. An approach to the autoignition of a turbulent mixture. *Acta Astronautica*, 1:1239–1266, 1974.
- [23] C. Dopazo and E.E. O'Brien. Statistical treatment of non-isothermal chemical reactions in turbulence. *Combustion Science and Technology*, 13:99–122, 1976.
- [24] H.E. Eickhoff and K. Grethe. A flame-zone model for turbulent hydrocarbon diffusion flames. *Combustion and Flame*, 35:267–275, 1979.
- [25] V. Eswaran and S.B. Pope. Direct numerical simulations of the turbulent mixing of a passive scalar. *Physics of Fluids A*, 31(3):506–520, 1988.
- [26] M.H. Everdell. *Introduction to Chemical Thermodynamics*. The English Universities Press, Ltd., 1965.
- [27] M. Fairweather, Jones W.P., H.S. Ledin, and R.P. Lindstedt. Predictions of soot formation in turbulent non-premixed propane flames. In *24th Symposium (International) on Combustion / The Combustion Institute*, pages 1067–1074, 1992.
- [28] R.C. Flagan and J.P. Appleton. A stochastic model of turbulent mixing with chemical reaction: nitric oxide formation in a plug-flow burner. *Combustion and Flame*, 23:249–267, 1974.
- [29] F. Gao. An analytical solution for the scalar probability density function in homogeneous turbulence. *Physics of Fluids A*, 3(4):511–513, April 1991.

- [30] F. Gao. Mapping closure and non-Gaussianity of the scalar probability density function in isotropic turbulence. *Physics of Fluids A*, 3(10):2438–2444, October 1991.
- [31] S. Godoy. *Turbulent Diffusion Flames*. PhD thesis, University of London, 1982.
- [32] S. Gordon and B.J. McBride. Computer program for calculation of complex equilibrium composition, rocket performance, incident and reflected shocks and Chapman-Jouget detonations. SP - 273, NASA, 1971.
- [33] D.C. Handscomb and J.M. Hammersley. *Monte Carlo Methods*. Methuen, London, 1965.
- [34] K. Hanjalić and B.E. Launder. A Reynolds stress model of turbulence and its application to thin shear flows. *Journal of Fluid Mechanics*, 52(4):609–638, 1972.
- [35] M.M.A. Hassan, F.C. Lockwood, and H.A. Moneib. Fluctuating temperature and mean concentration measurements in a vertical turbulent free jet diffusion flame. Paper presented at the "Italian Flame Days", Milano Marittima, June 1980.
- [36] W.R. Hawthorne, D.S. Weddell, and H.C. Hottel. Mixing and combustion in turbulent gas jets. *Third Symposium on Combustion, Flame and Explosion Phenomena*, pages 266–288, 1949.
- [37] M.V. Heitor, A.L.N. Moreira, and A.A.C. Pires. PDF/CFD based methods: Development and validation for low emissions combustor technology. Technical report, Instituto Superior Técnico, Lisbon, March 1994. First annual report, BRITE-EURAM-AERO CT 920035.
- [38] A.C. Hindmarsh. LSODE and LSODI, Two new initial value ordinary differential equation solvers. *ACM SIGNUM Newsletter*, 15(4), 1980.
- [39] J. Janicka, W. Kolbe, and W. Kollmann. The solution of a pdf transport equation for turbulent diffusion flames. In *Proceedings of the Heat Transfer and Fluid Mechanics Institute*, pages 296–312. Stanford University Press, 1978.
- [40] J. Janicka, W. Kolbe, and W. Kollmann. Closure of the equations for the probability density function of turbulent scalar fields. *Journal of Non-Equilibrium Thermodynamics*, 4:47–66, 1979.
- [41] W.P. Jones. Models for turbulent flows with variable density and combustion. In W. Kollmann, editor, *Prediction Methods for Turbulent Flows*, pages 380–421. Hemisphere Publ. Corp., 1980. A collection of the notes for a lecture series on prediction methods for turbulent flows held at the VKI in January 1979.
- [42] W.P. Jones. Turbulence modelling and numerical solution methods for variable density and combustng flows. In P.A. Libby and F.A. Williams, editors, *Turbulent Reactive Flows*, chapter 6, pages 309–374. Academic Press, 1994.
- [43] W.P. Jones and M. Kakhi. Mathematical modelling of turbulent flames. In Culick et al., editor, *Unsteady Combustion*. Kluwer Academic Publishers, 1994. to appear.

- [44] W.P. Jones and W. Kollmann. Multi-scalar pdf transport equations for turbulent diffusion flames. In Durst et al., editor, *Turbulent Shear Flows 5*, pages 296–309. Springer-Verlag, 1987.
- [45] W.P. Jones and B.E. Launder. The prediction of laminarisation with a two-equation model of turbulence. *International Journal of Heat and Mass transfer*, 15:301–314, 1972. also AIAA selected reprint series XIV, p.119-132, 1973.
- [46] W.P. Jones and R.P. Lindstedt. Global reaction schemes for hydrocarbon combustion. *Combustion and Flame*, 73:233–249, 1988.
- [47] W.P. Jones and J.J. McGuirk. Mathematical modelling of gas turbine combustion chambers. *AGARD Proc.*, No. 275:4.1–4.11, 1979.
- [48] W.P. Jones and P. Musonge. Closure of the Reynolds stress and scalar flux equations. *Physics of Fluids*, 31(12):3589–3603, 1988.
- [49] W.P. Jones and J.H. Whitelaw. Calculation methods for reacting turbulent flows. *Combustion and Flame*, 48:1–26, 1982.
- [50] J.H. Kent and R.W. Bilger. The prediction of turbulent diffusion flame fields and nitric oxide formation. In *16th Symposium (International) on Combustion / The Combustion Institute*, page 1643, 1977.
- [51] E.E. Khalil, D.B. Spalding, and J.H. Whitelaw. The calculation of local flow properties in two-dimensional furnaces. *International Journal of Heat and Mass transfer*, 18:775, 1975.
- [52] W. Kollmann. Pdf transport equations for chemically reacting flows. In R. Borghi and S.N.B. Murthy, editors, *Turbulent Reactive Flows*, pages 715–730. Springer-Verlag, 1989.
- [53] W. Kollmann. The pdf approach to turbulent flow. *Theoretical and Computational Fluid Dynamics*, 1:249–285, 1990.
- [54] W. Kollmann. Pdf transport modelling. Modelling of combustion and turbulence, Von Karman Institute for fluid dynamics, lecture series, 1992.
- [55] G. Kosaly. Theoretical remarks on a phenomenological model of turbulent mixing. *Combustion Science and Technology*, 49:227–234, 1986.
- [56] G. Kosaly and P. Givi. Modelling of turbulent molecular mixing. *Combustion and Flame*, 70:101–118, 1987.
- [57] K.N. Lakshmisha, Y. Zhang, B. Rogg, and K.N.C. Bray. Modelling auto-ignition in a turbulent medium. In *24th Symposium (International) on Combustion / The Combustion Institute*, pages 421–428, 1992.
- [58] B.E. Launder. Phenomenological modelling : Present and future ? In J.L. Lumley, editor, *Whither Turbulence ? Turbulence at the Crossroads*, pages 439–485. Springer-Verlag, 1990. Proceedings of a workshop held at Cornell University, Ithaca, March 1989. Published by Springer-Verlag as Lecture Notes in Physics 357, 1990.

- [59] B.E. Launder and D.B. Spalding. *Mathematical Models of Turbulence*. Academic Press, 1972.
- [60] P.A. Libby and F.A. Williams. Fundamental aspects. In P.A. Libby and F.A. Williams, editors, *Turbulent Reacting Flows*, pages 1–43. Springer-Verlag, 1980. Appeared under 'Topics in Applied Physics', vol.44.
- [61] S.K. Liew. *Flamelet models of turbulent non-premixed combustion*. PhD thesis, University of Southampton, 1983.
- [62] R.P. Lindstedt, 1994. Private Communication.
- [63] F.C. Lockwood and A.S. Naguib. The prediction of the fluctuations in the properties of free, round-jet, turbulent diffusion flames. *Combustion and Flame*, 24:109–124, 1975.
- [64] J.L. Lumley. Prediction Methods for Turbulent Flows. Introduction. Von Karman Institute lecture series no. 76., 1975.
- [65] T.S. Lundgren. Distribution functions in the statistical theory of turbulence. *Physics of Fluids*, 10(5):969–975, 1967.
- [66] A.R. Masri, R.W. Bilger, and R.W. Dibble. Turbulent non-premixed flames of methane near extinction : Mean structure from Raman measurements. *Combustion and Flame*, 71:245–266, 1988.
- [67] A.R. Masri, R.W. Bilger, and R.W. Dibble. Turbulent non-premixed flames of methane near extinction : Probability Density Functions. *Combustion and Flame*, 73:261–285, 1988.
- [68] A.R. Masri, R.W. Bilger, and R.W. Dibble. Conditional probability density functions measured in turbulent non-premixed flames of methane near extinction. *Combustion and Flame*, 74:267–284, 1988.
- [69] R.E. Mitchell, A.F. Sarofim, and L.A. Clomburg. Experimental and numerical investigation of confined laminar diffusion flames. *Combustion and Flame*, 37:227–244, 1980.
- [70] G.R. Newman, B.E. Launder, and J.L. Lumley. Modelling the behaviour of homogeneous scalar turbulence. *Journal of Fluid Mechanics*, 111:217–232, 1981.
- [71] A.T. Norris and S.B. Pope. Turbulent mixing model based on ordered pairing. *Combustion and Flame*, 83:27–42, 1991.
- [72] E.E. O'Brien. The probability density function (pdf) approach to reacting turbulent flows. In P.A. Libby and F.A. Williams, editors, *Turbulent Reacting Flows*, pages 185–218. Springer-Verlag, 1980. Appeared under 'Topics in Applied Physics', vol.44.
- [73] A. Papoulis. *Probability, random variables and stochastic processes*. Mc Graw-Hill, 1965.
- [74] S.V. Patankar and D.B. Spalding. *Heat and mass transfer in boundary layers: A general calculation procedure*. Intertext books, 2nd edition, 1970.
- [75] N. Peters. Laminar flamelet concepts in turbulent combustion. In *21st Symposium (International) on Combustion / The Combustion Institute*, pages 1231–1250, 1986.

- [76] N. Peters and B. Rogg, editors. *Reduced kinetic mechanisms for applications in combustion systems*. Lecture notes in Physics m15. Springer-Verlag, 1993.
- [77] S.B. Pope. The relationship between the probability approach and particle models for reaction in homogeneous turbulence. *Combustion and Flame*, 35:41–45, 1979.
- [78] S.B. Pope. The statistical theory of turbulent flames. *Philosophical Transactions of the Royal Society of London Series A*, 291:529–568, 1979.
- [79] S.B. Pope. A Monte Carlo method for the pdf equations of turbulent reactive flows. *Combustion Science and Technology*, 25:159–174, 1981.
- [80] S.B. Pope. An improved mixing model. *Combustion Science and Technology*, 28:131–135, 1982.
- [81] S.B. Pope. Pdf methods for turbulent reactive flows. *Progress in Energy and Combustion Science*, 11:119–192, 1985.
- [82] S.B. Pope. Mapping closures for turbulent mixing and reaction. *Theoretical and Computational Fluid Dynamics*, 2:255–270, 1991.
- [83] W.C. Reynolds. The potential and limitations of direct and large eddy simulation. In J.L. Lumley, editor, *Whither Turbulence ? Turbulence at the Crossroads*, pages 313–343. Springer-Verlag, 1990. Proceedings of a workshop held at Cornell University, Ithaca, March 1989. Published by Springer-Verlag as Lecture Notes in Physics 357, 1990.
- [84] J.C. Rotta. Statistische theorie nichthomogener turbulenz i und ii. *Zeitschrift für Physik*, 129:547–573, 1951.
- [85] H. Schlichting. *Boundary Layer Theory*. McGraw Hill Publishing Company, seventh edition, 1987. McGraw Hill Classic Textbook Reissue.
- [86] M. Sion and J.-Y. Chen. Scalar pdf modelling of turbulent non-premixed methanol-air flames. *Combustion Science and Technology*, 88:89–114, 1992.
- [87] G.D. Smith. *Numerical Solution of Partial Differential Equations: Finite Difference Methods*. Oxford University Press, 1978. Oxford Applied Mathematics and Computing Science Series.
- [88] D.B. Spalding. Concentration fluctuations in a round turbulent free jet. *Chemical Engineering Science*, 26:95–107, 1971.
- [89] L.A. Spielman and O. Levenspiel. A Monte Carlo treatment for reacting and coalescing dispersed phase systems. *Chemical Engineering Science*, 20:247–254, 1965.
- [90] R.A. Svehla. Estimated viscosities and thermal conductivities of gases at high temperatures. TR R-132, NASA, 1962.
- [91] S. Tavoularis and S. Corrsin. Experiments in nearly homogeneous turbulent shear flow with a uniform mean temperature gradient. Part 1. *Journal of Fluid Mechanics*, 104:311–347, 1981.

- [92] H. Tsuji and I. Yamaoka. Structure analysis of counterflow diffusion flames in the forward stagnation region of a porous cylinder. In *13th Symposium (International) on Combustion / The Combustion Institute*, pages 723–781, 1971.
- [93] M.S. Uberoi. Equipartition of energy and local isotropy in turbulent flows. *Journal of Applied Physics*, 28:1165–1170, 1957.
- [94] L. Valiño and C. Dopazo. A binomial sampling model for scalar turbulent mixing. *Physics of Fluids A*, 2(7):1204–1212, 1990.
- [95] L. Valiño and C. Dopazo. A binomial Langevin model for turbulent mixing. *Physics of Fluids A*, 3(12):3034–3037, 1991.
- [96] L. Valiño and C. Dopazo. Joint statistics of scalars and their gradients in nearly homogeneous turbulence. In A.V. Johansson and P.H. Alfredsson, editors, *Advances in Turbulence 3*, pages 312–323. Springer-Verlag, 1991.
- [97] L. Valiño and F. Gao. Monte Carlo implementation of a single-scalar mapping closure for diffusion in the presence of chemical reaction. *Physics of Fluids A*, 4(9):2062–2069, September 1992.
- [98] L. Valiño, J. Ros, and C. Dopazo. Monte Carlo implementation and analytic solution of an inert-scalar turbulent mixing test problem using a mapping closure. *Physics of Fluids A*, 3(9):2195–2198, September 1991.
- [99] L. Vervisch. *Prise en compte d'effets de cinétique chimique dans les flammes de diffusion turbulentes par l'approche fonction densité de probabilité*. PhD thesis, University of Rouen, 1991.
- [100] L. Vervisch. Applications of pdf turbulent combustion models to real non-premixed flame calculations. Modelling of combustion and turbulence Von Karman Institute lecture series 1992, 1992.
- [101] L. Vervisch, J.H. Chen, S. Mahalingam, and I.K. Puri. Numerical study of finite-rate chemistry effects and unequal Schmidt numbers in turbulent non-premixed combustion. In *Ninth Symposium on Turbulent Shear Flows*, Kyoto, Japan, August 16-18 1993.
- [102] F.A. Williams. Recent advances in the theoretical descriptions of turbulent diffusion flames. In S.N.B. Murthy, editor, *Turbulent Mixing in Non-reactive and Reactive Flows*. Plenum Press, 1975.

Appendix A

Derivation of the Single-Point, Single-Time Scalar Pdf Equation

The method of derivation presented in this appendix uses the concept of the fine-grained pdf, $\check{p}(\underline{\psi}, \underline{\phi}; \mathbf{x}, t)$. Equation (4.1) defines this quantity as:

$$\check{p}(\underline{\psi}, \underline{\phi}) \equiv \prod_{\alpha=1}^{N+1} \delta \{ \phi_{\alpha}(\mathbf{x}, t) - \psi_{\alpha} \}$$

Applying the chain rule,

$$\frac{\partial \check{p}}{\partial t} = - \sum_{\beta=1}^{N+1} \frac{\partial \phi_{\beta}}{\partial t} \frac{\partial \check{p}}{\partial \psi_{\beta}} \quad (\text{A.1})$$

$$\frac{\partial \check{p}}{\partial x_i} = - \sum_{\beta=1}^{N+1} \frac{\partial \phi_{\beta}}{\partial x_i} \frac{\partial \check{p}}{\partial \psi_{\beta}} \quad (\text{A.2})$$

The scalar transport equation can be written as:

$$\varrho \frac{\partial \phi_{\beta}}{\partial t} + \varrho u_j \frac{\partial \phi_{\beta}}{\partial x_j} = - \frac{\partial J_{i,\beta}}{\partial x_i} + \varrho(\underline{\phi}) \dot{S}_{\beta}^*(\underline{\phi}) \quad (\text{A.3})$$

where $\dot{S}_{\alpha}(\underline{\phi}) \equiv \varrho(\underline{\phi}) \dot{S}_{\alpha}^*(\underline{\phi})$ and $\dot{S}_{\beta}^*(\underline{\phi}) \equiv 0$ if $\phi_{\beta} \equiv h$ (for $\beta = N + 1$). Substituting for $\frac{\partial \phi_{\beta}}{\partial t}$ in equation (A.1) using (A.3) and then taking the ensemble average:

$$\left\langle \varrho \frac{\partial \check{p}}{\partial t} \right\rangle = - \left\langle \varrho u_j \frac{\partial \check{p}}{\partial x_j} \right\rangle + \sum_{\beta=1}^{N+1} \frac{\partial}{\partial \psi_{\beta}} \left\langle \frac{\partial J_{i,\beta}}{\partial x_i} \check{p} \right\rangle - \sum_{\beta=1}^{N+1} \frac{\partial}{\partial \psi_{\beta}} \left\langle \varrho \dot{S}_{\beta}^*(\underline{\phi}) \check{p} \right\rangle \quad (\text{A.4})$$

Equation (A.4) expresses the fact that terms such as $\varrho(\underline{\phi})$, $u_j(\mathbf{x}, t)$ and $\phi_{\beta}(\mathbf{x}, t)$ are unaffected by differentiation with respect to ψ_{β} . The term arising from the convective contribution makes use of the result shown in equation (A.2). Using mass continuity, equation (A.4) becomes:

$$\underbrace{\frac{\partial}{\partial t} \langle \varrho \check{p} \rangle}_{(a)} + \underbrace{\frac{\partial}{\partial x_j} \langle \varrho u_j \check{p} \rangle}_{(b)} = \sum_{\beta=1}^{N+1} \frac{\partial}{\partial \psi_{\beta}} \underbrace{\left\langle \frac{\partial J_{i,\beta}}{\partial x_i} \check{p} \right\rangle}_{(c)} - \sum_{\beta=1}^{N+1} \frac{\partial}{\partial \psi_{\beta}} \underbrace{\langle \varrho \dot{S}_{\beta}^*(\underline{\phi}) \check{p} \rangle}_{(d)} \quad (\text{A.5})$$

From probability theory [73], the expectation of the random variable $V(\zeta_1, \zeta_2, \dots, \zeta_M)$ is given by:

$$\begin{aligned}\langle V \rangle &= \int_{\hat{\zeta}_1} \cdots \int_{\hat{\zeta}_M} V(\hat{\zeta}_1, \dots, \hat{\zeta}_M) P(\hat{\zeta}_1, \dots, \hat{\zeta}_M) d\hat{\zeta}_1 \cdots d\hat{\zeta}_M \\ &= \int_{\hat{\zeta}_1} \cdots \int_{\hat{\zeta}_M} V(\hat{\zeta}_1, \dots, \hat{\zeta}_M) P(\hat{\zeta}_1 | \hat{\zeta}_2, \dots, \hat{\zeta}_M) P(\hat{\zeta}_2, \dots, \hat{\zeta}_M) d\hat{\zeta}_1 \cdots d\hat{\zeta}_M \\ &= \int_{\hat{\zeta}_2} \cdots \int_{\hat{\zeta}_M} \langle V(\zeta_1, \zeta_2, \dots, \zeta_M) | \zeta_2 = \hat{\zeta}_2, \dots, \zeta_M = \hat{\zeta}_M \rangle P(\hat{\zeta}_2, \dots, \hat{\zeta}_M) d\hat{\zeta}_2 \cdots d\hat{\zeta}_M\end{aligned}\quad (\text{A.6})$$

Now equation (A.5) is considered on a term by term basis; the expectations are evaluated using the result expressed in equation (A.6).

Term (a)

$$\langle \varrho(\underline{\phi}) \check{p}(\underline{\psi}, \underline{\phi}) \rangle = \int_{\hat{\underline{\phi}}} \langle \varrho(\underline{\phi}) \check{p}(\underline{\psi}, \underline{\phi}) | \underline{\phi} = \hat{\underline{\phi}} \rangle P(\hat{\underline{\phi}}) d\hat{\underline{\phi}} \quad (\text{A.7})$$

$\hat{\underline{\phi}}$ should not be confused with $\underline{\psi}$; they both constitute the sample space variables for $\underline{\phi}$, however for the purposes of the above integration $\hat{\underline{\phi}}$ acts as a dummy variable. The expected value of $\varrho(\underline{\phi})$ conditioned on the event $\underline{\phi} = \hat{\underline{\phi}}$ is simply $\varrho(\hat{\underline{\phi}})$. Using this idea,

$$\begin{aligned}\langle \varrho(\underline{\phi}) \check{p}(\underline{\psi}, \underline{\phi}) | \underline{\phi} = \hat{\underline{\phi}} \rangle &= \left\langle \varrho(\underline{\phi}) \prod_{\alpha=1}^{N+1} \delta \{ \phi_{\alpha}(\mathbf{x}, t) - \psi_{\alpha} \} \right|_{\underline{\phi} = \hat{\underline{\phi}}} \\ &= \varrho(\hat{\underline{\phi}}) \prod_{\alpha=1}^{N+1} \delta \{ \hat{\phi}_{\alpha} - \psi_{\alpha} \}\end{aligned}\quad (\text{A.8})$$

The delta function is a generalised function; in other words it is defined in terms of its integral properties:

$$\int_{-\infty}^{\infty} \delta(y) dy = 1; \quad \int_{-\infty}^{\infty} b \delta(y - c) g(y) dy = b g(c) \quad (\text{A.9})$$

Substituting the results of equations (A.8) and (A.9) into (A.7) leads to:

$$\langle \varrho(\underline{\phi}) \check{p}(\underline{\psi}, \underline{\phi}) \rangle = \varrho(\underline{\psi}) P(\underline{\psi}) \quad (\text{A.10})$$

Term (b)

Using the same procedure as for term (a),

$$\begin{aligned}\langle \varrho(\underline{\phi}) u_j(\mathbf{x}, t) \check{p}(\underline{\psi}, \underline{\phi}) \rangle &= \int_{\hat{\underline{\phi}}} \langle \varrho(\underline{\phi}) u_j(\mathbf{x}, t) \check{p}(\underline{\psi}, \underline{\phi}) | \underline{\phi} = \hat{\underline{\phi}} \rangle P(\hat{\underline{\phi}}) d\hat{\underline{\phi}} \\ &= \int_{\hat{\underline{\phi}}} \varrho(\hat{\underline{\phi}}) \langle u_j | \underline{\phi} = \hat{\underline{\phi}} \rangle \check{p}(\underline{\psi}, \hat{\underline{\phi}}) P(\hat{\underline{\phi}}) d\hat{\underline{\phi}} \\ &= \varrho(\underline{\psi}) \langle u_j | \underline{\phi} = \underline{\psi} \rangle P(\underline{\psi})\end{aligned}\quad (\text{A.11})$$

Terms (c) and (d)

Based on the results above terms (c) and (d) become, by inspection,

$$\left\langle \frac{\partial J_{i,\beta}}{\partial x_i} \check{p} \right\rangle = \left\langle \frac{\partial J_{i,\beta}}{\partial x_i} \right|_{\underline{\phi} = \underline{\psi}} P(\underline{\psi}) \quad (\text{A.12})$$

$$\langle \varrho \dot{S}_{\beta}^*(\underline{\phi}) \check{p} \rangle = \varrho(\underline{\psi}) \dot{S}_{\beta}^*(\underline{\phi}) P(\underline{\psi}) \quad (\text{A.13})$$

Substituting equations (A.10) to (A.13) into equation (A.5) yields:

$$\begin{aligned} \frac{\partial}{\partial t} \{ \varrho(\underline{\psi}) P(\underline{\psi}) \} + \frac{\partial}{\partial x_j} \{ \varrho(\underline{\psi}) \langle u_j | \underline{\phi} = \underline{\psi} \rangle P(\underline{\psi}) \} &= \sum_{\beta=1}^{N+1} \frac{\partial}{\partial \psi_\beta} \left\{ \left\langle \frac{\partial J_{i,\beta}}{\partial x_i} \right| \underline{\phi} = \underline{\psi} \right\rangle P(\underline{\psi}) \right\} \\ &\quad - \sum_{\beta=1}^{N+1} \frac{\partial}{\partial \psi_\beta} \{ \varrho(\underline{\psi}) \dot{S}_\beta^*(\underline{\phi}) P(\underline{\psi}) \} \end{aligned} \quad (\text{A.14})$$

Introducing the definition of the density-weighted pdf, $\bar{\varrho}(\underline{\phi}) \tilde{P}(\underline{\psi}) \equiv \varrho(\underline{\psi}) P(\underline{\psi})$, and performing a density-weighted decomposition of $u_j(\mathbf{x}, t)$ leads to,

$$\begin{aligned} \frac{\partial}{\partial t} \{ \bar{\varrho} \tilde{P}(\underline{\psi}) \} + \frac{\partial}{\partial x_j} \{ \bar{\varrho} \tilde{u}_j \tilde{P}(\underline{\psi}) + \bar{\varrho} \langle u_j'' | \underline{\phi} = \underline{\psi} \rangle \tilde{P}(\underline{\psi}) \} \\ = \sum_{\beta=1}^{N+1} \frac{\partial}{\partial \psi_\beta} \left\{ \left\langle \frac{\partial J_{i,\beta}}{\partial x_i} \right| \underline{\phi} = \underline{\psi} \right\rangle \tilde{P}(\underline{\psi}) \right\} - \sum_{\beta=1}^{N+1} \frac{\partial}{\partial \psi_\beta} \{ \bar{\varrho} \dot{S}_\beta^*(\underline{\phi}) \tilde{P}(\underline{\psi}) \} \end{aligned} \quad (\text{A.15})$$

Applying the density-weighted mass continuity equation to the terms on the lhs of (A.15) leads to terms I, II and IV in equation (4.3). Since $\bar{\varrho}$ is a function of $\underline{\phi}(\mathbf{x}, t)$, it is unaffected by differentiation with respect to ψ_β so that the source term for reaction in equation (A.15) leads to term III in equation (4.3). The mixing terms are identical in both equations.

Having discussed the derivation of equation (4.3), the result in equation (4.4) will now be considered. Assuming that $J_{i,\beta}$ in equation (A.4) can be expressed in terms of Fick's Law (for $\beta = 1, \dots, N$) and Fourier's Law (for $\beta = N + 1$), this implies:

$$\begin{aligned} \sum_{\beta=1}^{N+1} \frac{\partial}{\partial \psi_\beta} \left\langle \frac{\partial J_{i,\beta}}{\partial x_i} \tilde{p} \right\rangle &= - \sum_{\beta=1}^{N+1} \frac{\partial}{\partial \psi_\beta} \left\langle \frac{\partial}{\partial x_i} \left(\frac{\mu}{Sc} \frac{\partial \phi_\beta}{\partial x_i} \right) \tilde{p} \right\rangle \\ &= - \sum_{\beta=1}^{N+1} \left\langle \frac{\partial}{\partial x_i} \left(\frac{\mu}{Sc} \frac{\partial \phi_\beta}{\partial x_i} \right) \frac{\partial \tilde{p}}{\partial \psi_\beta} \right\rangle \\ &= \underbrace{- \sum_{\beta=1}^{N+1} \frac{\partial}{\partial x_i} \left\langle \frac{\mu}{Sc} \frac{\partial \phi_\beta}{\partial x_i} \frac{\partial \tilde{p}}{\partial \psi_\beta} \right\rangle}_{(e)} + \underbrace{\sum_{\beta=1}^{N+1} \frac{\partial}{\partial \psi_\beta} \left\langle \frac{\mu}{Sc} \frac{\partial \phi_\beta}{\partial x_i} \frac{\partial \tilde{p}}{\partial x_i} \right\rangle}_{(f)} \end{aligned} \quad (\text{A.16})$$

At the energy containing scales of turbulent motion, ϕ' can be defined as the characteristic scalar fluctuation. However at the fine scales, the controlling parameter is the scalar dissipation rate; in this case, the scalar fluctuations are of the order $(\chi_\phi \frac{\nu^2}{\nu})^{\frac{1}{2}}$. In analogy with the order of magnitude estimate for ε , energy cascade arguments would suggest that $\chi_\phi \sim \mathcal{O}(\frac{u'}{L} \phi'^2)$. Comparing the order of magnitude of terms (e) and (f) in equation (A.16), and ignoring the contributions which are common to both of them,

$$\begin{aligned} \frac{\text{Term (e)}}{\text{Term (f)}} &\sim \mathcal{O} \left\{ \left[\left(\frac{u'}{L} \phi'^2 \frac{\eta^2}{\nu} \right)^{-\frac{1}{2}} L^{-1} \right] \cdot [\phi' \eta] \right\} \\ &\sim \mathcal{O} \left(\frac{\nu}{u' L} \right)^{\frac{1}{2}}, \quad \text{where } Re \equiv \frac{\nu}{u' L} \end{aligned} \quad (\text{A.17})$$

So as $Re \rightarrow \infty$, term (e) is negligible in comparison to term (f) and its contribution can be neglected in equation (A.16).

Substituting for $\frac{\partial \tilde{p}}{\partial x_i}$ in equation (A.16) using (A.2) yields:

$$\begin{aligned}
 \sum_{\beta=1}^{N+1} \frac{\partial}{\partial \psi_\beta} \left\langle \frac{\partial J_{i,\beta}}{\partial x_i} \tilde{p} \right\rangle &= - \sum_{\beta=1}^{N+1} \frac{\partial}{\partial \psi_\beta} \left\langle \frac{\mu}{Sc} \frac{\partial \phi_\beta}{\partial x_i} \sum_{\gamma=1}^{N+1} \frac{\partial \phi_\gamma}{\partial x_i} \frac{\partial \tilde{p}}{\partial \psi_\gamma} \right\rangle \\
 &= - \sum_{\beta=1}^{N+1} \sum_{\gamma=1}^{N+1} \frac{\partial^2}{\partial \psi_\beta \partial \psi_\gamma} \left\langle \frac{\mu}{Sc} \frac{\partial \phi_\beta}{\partial x_i} \frac{\partial \phi_\gamma}{\partial x_i} \tilde{p} \right\rangle \\
 &= - \sum_{\beta=1}^{N+1} \sum_{\gamma=1}^{N+1} \frac{\partial^2}{\partial \psi_\beta \partial \psi_\gamma} \left\{ \left\langle \frac{\mu}{Sc} \frac{\partial \phi_\beta}{\partial x_i} \frac{\partial \phi_\gamma}{\partial x_i} \right| \underline{\phi} = \underline{\psi} \right\rangle P(\underline{\psi}) \right\} \quad (\text{A.18})
 \end{aligned}$$

Equation (A.18) is identical to (4.4).

Appendix B

The Basic Equations that are Solved

In this appendix the basic equations describing the evolution of the turbulent jet flows, investigated in this thesis, are written down. The equations effectively summarise the level of closure that is being adopted. It is assumed that the turbulence is statistically two-dimensional, stationary, axisymmetric and non-swirling. The thin shear layer approximation is invoked and the equations are written down in cylindrical polar coordinates. The velocity field is represented in terms of the continuity and $\tilde{u}$ -momentum equations.

B.1 Mass Continuity

This equation is written down for completeness, although it is not solved in the calculation procedure because of the transformation to a dimensionless stream function coordinate system. The stream function is defined so that it automatically satisfies the mass continuity equation in a steady 'two-dimensional' flow.

$$\frac{\partial}{\partial x}(\bar{\rho}\tilde{u}) + \frac{1}{r}\frac{\partial}{\partial r}(\bar{\rho}\tilde{u}r) = 0 \quad (\text{B.1})$$

Since r, x are independent, this can be written as:

$$\frac{\partial}{\partial x}(\bar{\rho}\tilde{u}r) + \frac{\partial}{\partial r}(\bar{\rho}\tilde{u}r) = 0 \quad (\text{B.2})$$

B.2 Momentum Equation

Due to the thin shear layer approximation, only the $\tilde{u}$ -component of the momentum equation needs to be solved.

$$\underbrace{\bar{\rho}\tilde{u}\frac{\partial\tilde{u}}{\partial x} + \bar{\rho}\tilde{v}\frac{\partial\tilde{u}}{\partial r}}_{\text{convection}} = \underbrace{\frac{1}{r}\frac{\partial}{\partial r}\left(r\mu\frac{\partial\tilde{u}}{\partial r}\right)}_{\text{diffusion}} - \underbrace{\frac{1}{r}\frac{\partial}{\partial r}\left(r\bar{\rho}\widetilde{u''v''}\right) - \frac{\partial p}{\partial x} + \bar{\rho}g_x}_{\text{source terms}} \quad (\text{B.3})$$

The Reynolds stress term in equation (B.3) requires closure¹. In this work, both an eddy viscosity and a Reynolds stress closure are employed to model this term. The constants appearing in the modelled equations are presented in sections 2.5.1 and 2.5.2 for eddy-viscosity and Reynolds stress closures, respectively. For the latter model, table 2.1 can also be consulted.

¹The Reynolds stress term in equation (B.3) only appears as a 'source' if a Reynolds stress closure is employed. In the k - ϵ model the Reynolds stress appears, in form, as a diffusion term.

B.2.1 The k - ε Model

The correlation of fluctuating velocities is given by:

$$-\bar{\rho}\widetilde{u''v''} = \mu_t \frac{\partial \tilde{u}}{\partial r}, \quad \text{where} \quad \mu_t = C_\mu \bar{\rho} \frac{k^2}{\varepsilon} \quad (\text{B.4})$$

In order to use this closure, the values of k and ε are required; these are obtained from their respective modelled equations,

$$\bar{\rho}\tilde{u} \frac{\partial k}{\partial x} + \bar{\rho}\tilde{v} \frac{\partial k}{\partial r} = \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\mu_t}{\sigma_k} \frac{\partial k}{\partial r} \right) + \mu_t \left(\frac{\partial \tilde{u}}{\partial r} \right)^2 - \bar{\rho}\varepsilon \quad (\text{B.5})$$

$$\bar{\rho}\tilde{u} \frac{\partial \varepsilon}{\partial x} + \bar{\rho}\tilde{v} \frac{\partial \varepsilon}{\partial r} = \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\mu_t}{\sigma_\varepsilon} \frac{\partial \varepsilon}{\partial r} \right) + C_{\varepsilon_1} \frac{\varepsilon}{k} \mu_t \left(\frac{\partial \tilde{u}}{\partial r} \right)^2 - C_{\varepsilon_2} \bar{\rho} \frac{\varepsilon^2}{k} \quad (\text{B.6})$$

B.2.2 The Reynolds Stress Closure

With this closure the solution of a modelled turbulence kinetic energy equation is obviated. Instead the solution of the modelled equations for the following quantities is sought: $\widetilde{u''^2}$, $\widetilde{v''^2}$, $\widetilde{w''^2}$ and $\widetilde{u''v''}$. The transformation from Cartesian to cylindrical polar coordinates leads to the appearance of many terms, however, for thin shear layer flows only a single component of the mean strain is taken to be significant, namely $\frac{\partial \tilde{u}}{\partial r}$; terms involving the other components are neglected. Furthermore, the assumption of circumferential symmetry coupled with $\tilde{w} = 0$ imply that $\widetilde{u''w''} = \widetilde{v''w''} = 0$.

The form of the Reynolds stress closure proposed by Jones and Musonge [48] is employed. For $\widetilde{u''v''}$ equation (2.62), subject to the assumptions above, takes the following form in cylindrical polar coordinates:

$$\begin{aligned} \bar{\rho}\tilde{u} \frac{\partial}{\partial x} (\widetilde{u''v''}) + \bar{\rho}\tilde{v} \frac{\partial}{\partial r} (\widetilde{u''v''}) &= \frac{1}{r} \frac{\partial}{\partial r} \left[r C_s \bar{\rho} \frac{k}{\varepsilon} \widetilde{v''^2} \frac{\partial}{\partial r} (\widetilde{u''v''}) \right] \\ &\quad - C_s \bar{\rho} \frac{k}{\varepsilon} \widetilde{w''^2} \frac{\widetilde{u''v''}}{r^2} - \bar{\rho} \widetilde{v''^2} \frac{\partial \tilde{u}}{\partial r} + A_{12}^* \end{aligned} \quad (\text{B.7})$$

and, subject to the simplifications brought about under the thin shear layer approximation, the component A_{12}^* of the tensor A_{ij}^* in equation (2.61) can be written as,

$$A_{12}^* = -C_1 \bar{\rho} \varepsilon \frac{\widetilde{u''v''}}{k} + \left[C_4 k - \left(\frac{3}{2} + C_3 \right) \widetilde{u''^2} + C_3 \widetilde{v''^2} \right] \bar{\rho} \frac{\partial \tilde{u}}{\partial r} \quad (\text{B.8})$$

Equation (B.7) indicates that the gradient modelling of the transport term requires, *inter alia*, the value of k and ε ; k is obtained from the definition, equation (2.38). This in turn implies that the modelled equations for the normal stresses have to be solved,

$$\begin{aligned} \bar{\rho}\tilde{u} \frac{\partial \widetilde{u''^2}}{\partial x} + \bar{\rho}\tilde{v} \frac{\partial \widetilde{u''^2}}{\partial r} &= \frac{1}{r} \frac{\partial}{\partial r} \left(r C_s \bar{\rho} \frac{k}{\varepsilon} \widetilde{v''^2} \frac{\partial \widetilde{u''^2}}{\partial r} \right) - 2 \bar{\rho} \widetilde{u''v''} \frac{\partial \tilde{u}}{\partial r} \\ &\quad - \frac{2}{3} \bar{\rho} \varepsilon + A_{11}^* \end{aligned} \quad (\text{B.9})$$

$$\begin{aligned} \bar{\rho}\tilde{u} \frac{\partial \widetilde{v''^2}}{\partial x} + \bar{\rho}\tilde{v} \frac{\partial \widetilde{v''^2}}{\partial r} &= \frac{1}{r} \frac{\partial}{\partial r} \left(r C_s \bar{\rho} \frac{k}{\varepsilon} \widetilde{v''^2} \frac{\partial \widetilde{v''^2}}{\partial r} \right) - 2 C_s \bar{\rho} \frac{k}{\varepsilon} \widetilde{w''^2} \frac{(\widetilde{v''^2} - \widetilde{w''^2})}{r^2} \\ &\quad - \frac{2}{3} \bar{\rho} \varepsilon + A_{22}^* \end{aligned} \quad (\text{B.10})$$

$$\begin{aligned} \bar{\rho}\tilde{u}\frac{\partial\widetilde{w''^2}}{\partial x} + \bar{\rho}\tilde{v}\frac{\partial\widetilde{w''^2}}{\partial r} &= \frac{1}{r}\frac{\partial}{\partial r}\left(rC_s\bar{\rho}\frac{k}{\varepsilon}\widetilde{v''^2}\frac{\partial\widetilde{w''^2}}{\partial r}\right) + 2C_s\bar{\rho}\frac{k}{\varepsilon}\widetilde{w''^2}\frac{(\widetilde{v''^2} - \widetilde{w''^2})}{r^2} \\ &\quad - \frac{2}{3}\bar{\rho}\varepsilon + A_{33}^* \end{aligned} \quad (\text{B.11})$$

where,

$$A_{11}^* = -C_1\bar{\rho}\varepsilon\left(\frac{\widetilde{u''^2}}{k} - \frac{2}{3}\right) + (C_2 + 2C_3)\bar{\rho}\widetilde{u''v''}\frac{\partial\tilde{u}}{\partial r} \quad (\text{B.12})$$

$$A_{22}^* = -C_1\bar{\rho}\varepsilon\left(\frac{\widetilde{v''^2}}{k} - \frac{2}{3}\right) - 2(C_2 + C_3)\bar{\rho}\widetilde{u''v''}\frac{\partial\tilde{u}}{\partial r} \quad (\text{B.13})$$

$$A_{33}^* = -C_1\bar{\rho}\varepsilon\left(\frac{\widetilde{w''^2}}{k} - \frac{2}{3}\right) - C_2\bar{\rho}\widetilde{u''v''}\frac{\partial\tilde{u}}{\partial r} \quad (\text{B.14})$$

The modelled dissipation rate equation takes the form:

$$\bar{\rho}\tilde{u}\frac{\partial\varepsilon}{\partial x} + \bar{\rho}\tilde{v}\frac{\partial\varepsilon}{\partial r} = \frac{1}{r}\frac{\partial}{\partial r}\left(rC_\varepsilon\bar{\rho}\frac{k}{\varepsilon}\widetilde{v''^2}\frac{\partial\varepsilon}{\partial r}\right) - C_{\varepsilon_1}\bar{\rho}\frac{\varepsilon}{k}\widetilde{u''v''}\frac{\partial\tilde{u}}{\partial r} - C_{\varepsilon_2}\bar{\rho}\frac{\varepsilon^2}{k} \quad (\text{B.15})$$

B.3 Scalar Pdf Transport Equation

The scalar field is calculated using a multi-scalar, single-point, single-time, Eulerian pdf transport equation. For the inhomogeneous turbulent flows under consideration, equation (4.3) becomes:

$$\begin{aligned} \underbrace{\bar{\rho}\tilde{u}\frac{\partial\tilde{P}(\psi)}{\partial x} + \bar{\rho}\tilde{v}\frac{\partial\tilde{P}(\psi)}{\partial r}}_{\text{convection}} &= \underbrace{\frac{1}{r}\frac{\partial}{\partial r}\left\{r\frac{\mu_t}{\sigma_t}\frac{\partial\tilde{P}(\psi)}{\partial r}\right\}}_{\text{diffusion}} - \underbrace{\bar{\rho}\sum_{\alpha=1}^{N+1}\frac{\partial}{\partial\psi_\alpha}\left\{\dot{S}_\alpha^*(\psi)\tilde{P}(\psi)\right\}}_{\text{source I}} \\ &\quad + \underbrace{\sum_{\alpha=1}^{N+1}\frac{\partial}{\partial\psi_\alpha}\left\{\left\langle\frac{\partial J_{i,\alpha}}{\partial x_i}\right|\phi=\psi\right\rangle\tilde{P}(\psi)\right\}}_{\text{source II}} \end{aligned} \quad (\text{B.16})$$

where gradient transport is employed to model the turbulent diffusion term, section 4.12, and μ_t is given by equation (B.4), regardless of the turbulence model being employed. Note that although the source term for reaction is grouped together with the molecular mixing term, the former does not require modelling. This type of grouping is done out of convenience since the transport equations for the various quantities to be solved will then have a similar form; this feature can then be exploited when the finite-difference approximations to these equations are discussed.

Appendix C

The Transformation of the Equations to the (x, ω) Coordinate System

The general form of the equations can be expressed as:

$$\bar{\rho}\tilde{u}\frac{\partial\tilde{\phi}}{\partial x} + \bar{\rho}\tilde{v}\frac{\partial\tilde{\phi}}{\partial y} = \frac{1}{r}\frac{\partial}{\partial y}\left(r\Gamma_{\phi}\frac{\partial\tilde{\phi}}{\partial y}\right) + S_{\phi} \quad (\text{C.1})$$

where $y = r$ for cylindrical polar coordinates and $r = 1$ for Cartesian coordinates. For reasons of clarity and generality, ϕ, u and v are used in the remainder of this appendix to denote the dependent variables; i.e. the 'tilde' is omitted. The application of the *Von Mises* transformation maps the relation $\phi = f(x, y)$ to $\phi = g(x, \Psi)$, where f and g denote functional relations. The transformation does not affect the velocities. The chain rule is used to obtain the following expressions for the differential of ϕ in the two coordinate systems:

$$\delta\phi = \left(\frac{\partial\phi}{\partial x}\right)_y \delta x + \left(\frac{\partial\phi}{\partial y}\right)_x \delta y \quad (\text{C.2})$$

$$\delta\phi = \left(\frac{\partial\phi}{\partial x}\right)_{\Psi} \delta x + \left(\frac{\partial\phi}{\partial\Psi}\right)_x \delta\Psi \quad (\text{C.3})$$

Dividing equation (C.3) by δx and taking the limit as $\delta x \rightarrow 0$ with y held constant ($\delta y = 0$), the following expression for $\left(\frac{\partial\phi}{\partial x}\right)_y$ can be obtained,

$$\left(\frac{\partial\phi}{\partial x}\right)_y = \left(\frac{\partial\phi}{\partial x}\right)_{\Psi} + \left(\frac{\partial\phi}{\partial\Psi}\right)_x \left(\frac{\partial\Psi}{\partial x}\right)_y \quad (\text{C.4})$$

A similar procedure in terms of δy (with x held constant) yields:

$$\left(\frac{\partial\phi}{\partial y}\right)_x = \left(\frac{\partial\phi}{\partial\Psi}\right)_x \left(\frac{\partial\Psi}{\partial y}\right)_x \quad (\text{C.5})$$

The transformation is achieved by substituting equations (C.4) and (C.5), together with the definition of the stream function (equation (5.3)) into (C.1),

$$\begin{aligned} \frac{1}{r}\left(\frac{\partial\Psi}{\partial y}\right)_x \left[\left(\frac{\partial\phi}{\partial x}\right)_{\Psi} + \left(\frac{\partial\phi}{\partial\Psi}\right)_x \left(\frac{\partial\Psi}{\partial x}\right)_y \right] - \frac{1}{r}\left(\frac{\partial\Psi}{\partial x}\right)_y \left(\frac{\partial\phi}{\partial\Psi}\right)_x \left(\frac{\partial\Psi}{\partial y}\right)_x = \\ \frac{1}{r}\left(\frac{\partial\Psi}{\partial y}\right)_x \left\{ \frac{\partial}{\partial\Psi} \left[r\Gamma_{\phi} \left(\frac{\partial\Psi}{\partial y}\right)_x \left(\frac{\partial\phi}{\partial\Psi}\right)_x \right] \right\} + S_{\phi} \end{aligned} \quad (\text{C.6})$$

Equation (C.6) can be simplified to:

$$\left(\frac{\partial \phi}{\partial x}\right)_{\Psi} = \frac{\partial}{\partial \Psi} \left[\rho u r^2 \Gamma_{\phi} \left(\frac{\partial \phi}{\partial \Psi}\right)_{x,x} \right] + \frac{S_{\phi}}{\rho u} \quad (\text{C.7})$$

The transformation from (x, Ψ) to (x, ω) coordinates follows much the same path as the transformation described above. In fact it is possible to go from (x, y) to (x, ω) in one operation; however the derivation is algebraically more involved, and therefore less straightforward to follow.

ω is a dimensionless stream function and defined as¹:

$$\omega \equiv \frac{\Psi - \Psi_I}{\Psi_E - \Psi_I}, \quad \text{where} \quad 0 \leq \omega \leq 1 \quad (\text{C.8})$$

In analogy with the derivation of equations (C.4) and (C.5), the relations relating the (x, Ψ) system to that of (x, ω) can be expressed as,

$$\left(\frac{\partial \phi}{\partial x}\right)_{\Psi} = \left(\frac{\partial \phi}{\partial x}\right)_{\omega} + \left(\frac{\partial \phi}{\partial \omega}\right)_x \left(\frac{\partial \omega}{\partial x}\right)_{\Psi} \quad (\text{C.9})$$

$$\left(\frac{\partial \phi}{\partial \Psi}\right)_x = \left(\frac{\partial \phi}{\partial \omega}\right)_x \left(\frac{\partial \omega}{\partial \Psi}\right)_x \quad (\text{C.10})$$

From equation (C.8) it follows:

$$\left(\frac{\partial \omega}{\partial \Psi}\right)_x = \frac{1}{\Psi_E - \Psi_I} \quad (\text{C.11})$$

$$\begin{aligned} \left(\frac{\partial \omega}{\partial x}\right)_{\Psi} &= \left[(\Psi_E - \Psi_I) \left(-\frac{d\Psi_I}{dx}\right) - (\Psi - \Psi_I) \frac{d}{dx} (\Psi_E - \Psi_I) \right] \frac{1}{(\Psi_E - \Psi_I)^2} \\ &= - \left[\frac{d\Psi_I}{dx} + \omega \frac{d}{dx} (\Psi_E - \Psi_I) \right] \frac{1}{(\Psi_E - \Psi_I)} \\ &= \frac{r_I \dot{m}_I + \omega (r_E \dot{m}_E - r_I \dot{m}_I)}{(\Psi_E - \Psi_I)} \end{aligned} \quad (\text{C.12})$$

Equation (C.12) uses the definitions expressed in equation (5.6). Substituting equations (C.9) to (C.12) into (C.7) leads to the general form of the boundary layer equation that is being solved using the Patankar-Spalding method:

$$(\Psi_E - \Psi_I) \frac{\partial \phi}{\partial x} + (a + b\omega) \frac{\partial \phi}{\partial \omega} = \frac{\partial}{\partial \omega} \left[\frac{\rho u r^2 \Gamma_{\phi}}{(\Psi_E - \Psi_I)} \frac{\partial \phi}{\partial \omega} \right] + \frac{(\Psi_E - \Psi_I)}{\rho u} S_{\phi} \quad (\text{C.13})$$

where $a = r_I \dot{m}_I$, $b = r_E \dot{m}_E - r_I \dot{m}_I$ and $(\Psi_E - \Psi_I)$ is a function of x only. In equation (C.13) the partial derivatives are defined either with respect to ω or x held constant, and for conciseness the dependency has been omitted.

¹The expression written below also appears in section 5.2.3 as equation (5.4)

Appendix D

The Finite-Difference Procedure

D.1 Preliminary Remarks

The general form of the boundary layer equations is derived in appendix C, equation (C.13) and can be expressed as,

$$(\Psi_E - \Psi_I) \frac{\partial \phi}{\partial x} + (a + b\omega) \frac{\partial \phi}{\partial \omega} = \frac{\partial}{\partial \omega} \left[\frac{\rho u r^2 \Gamma_\phi}{(\Psi_E - \Psi_I)} \frac{\partial \phi}{\partial \omega} \right] + \frac{(\Psi_E - \Psi_I)}{\rho u} S_\phi$$

where $a = r_I \dot{m}_I$, $b = r_E \dot{m}_E - r_I \dot{m}_I$ and $(\Psi_E - \Psi_I)$ is a function of x only. In order to construct difference approximations, the control volume shown in figure D.1 is employed.

The typical 'computational molecule' indicates that the solution procedure is of the 'marching-type' i.e. the properties at any particular node are affected by the node directly upstream and by two neighbouring cross-stream nodes; information downstream is not required. The solution procedure is not iterative, which implies that it is computationally economical and the resulting algebraic equations can be solved exactly. It is pointed out in section 5.2.3 that ω is the monotonically increasing, independent cross-stream variable and is defined as a normalised stream function, so that $0 \leq \omega \leq 1$ is always satisfied. Once the ω distribution is generated this remains fixed regardless of the downstream position, i.e. $\omega_i^1 = \omega_i^2 \dots = \omega_i^n = \omega_i^{n+1}$. This condition obviously does not hold for y and Ψ . The following grid/mesh definitions are used in the discretisation of

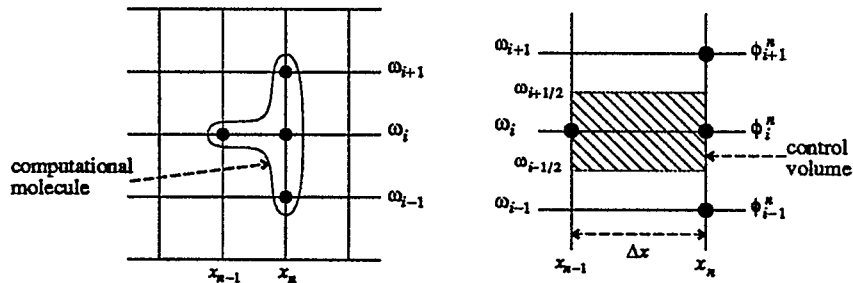


Figure D.1: Schematic of the typical computational molecule and control volume used to derive the difference approximations. The effect of the I and E boundaries are not considered at this stage. The notation ϕ_i^n denotes the value of the variable ϕ at node i ($1 \leq i \leq N+1$) and $x = x_n$. $\omega_{i+1/2} \equiv \frac{1}{2}(\omega_i + \omega_{i+1})$; this implies that the control volume surface lies midway between grid nodes irrespective of whether or not the mesh spacing is uniform.

equation (C.13):

$$\delta\omega_i \equiv \omega_{i+1} - \omega_i, \quad \Delta\omega_i \equiv \frac{1}{2}(\omega_{i+1} - \omega_{i-1}), \quad \delta y_i \equiv y_{i+1} - y_i, \quad \Delta x \equiv x_n - x_{n-1} \quad (\text{D.1})$$

Note that whilst $\omega_{i\pm\frac{1}{2}} \equiv \frac{1}{2}(\omega_i + \omega_{i\pm 1})$ is a definition, the corresponding equation in physical space, namely $y_{i\pm\frac{1}{2}} = \frac{1}{2}(y_i + y_{i\pm 1})$, is an approximation due to the non-linear relationship between ω and y .

D.2 Finite-Difference Approximations for the Internal Nodes

The algebraic approximations to the exact derivatives in equation (C.13) are obtained in the following manner:

- Equation (C.13) is integrated over the control volume, shown in figure D.1, and the result divided by the volume. This latter operation has no effect on the solution and is only carried out so that the fde approximates the pde¹ rather than its integrated form.
- As much of the integration as possible is carried analytically and then assumptions are made about the variation of the relevant properties between the grid nodes, e.g. the algebraic approximations are constructed to be linear in ϕ so that a , b and $\frac{\rho u r^2 \Gamma_\phi}{(\Psi_E - \Psi_I)}$ are based on the upstream value ($x = x_{n-1}$). Once these assumptions are made, the integration can be performed and the discretisation process is complete.

Other methods of obtaining difference approximations do exist (e.g. Taylor series expansion) but the current technique, referred to as a finite-volume or control-volume approach, is commonly employed partly because of its physical appeal. The control volume method automatically generates *conservative* schemes - that is, it satisfies the same conservation law which was used to derive the corresponding pde for *all mesh spacings*. The conservation requirement is achieved by ensuring that the fluxes of any property leaving a control surface is the same as the flux entering the surface of the adjacent control volume. Integrating equation (C.13),

$$\begin{aligned} & \underbrace{\frac{1}{\Delta x \Delta \omega_i} \int_{x_{n-1}}^{x_n} \int_{\omega_{i-\frac{1}{2}}}^{\omega_{i+\frac{1}{2}}} (\Psi_E - \Psi_I) \frac{\partial \phi}{\partial x} d\omega dx}_{\text{Axial convection: I}} \\ & + \underbrace{\frac{1}{\Delta x \Delta \omega_i} \int_{x_{n-1}}^{x_n} \int_{\omega_{i-\frac{1}{2}}}^{\omega_{i+\frac{1}{2}}} (a + b\omega) \frac{\partial \phi}{\partial \omega} d\omega dx}_{\text{Cross-stream convection: II}} = \\ & \underbrace{\frac{1}{\Delta x \Delta \omega_i} \int_{x_{n-1}}^{x_n} \int_{\omega_{i-\frac{1}{2}}}^{\omega_{i+\frac{1}{2}}} \frac{\partial}{\partial \omega} \left(\frac{\rho u r^2 \Gamma_\phi}{(\Psi_E - \Psi_I)} \frac{\partial \phi}{\partial \omega} \right) d\omega dx}_{\text{Cross-stream diffusion: III}} \end{aligned}$$

¹'fde' and 'pde' denote finite-difference equation and partial differential equation, respectively.

$$+ \underbrace{\frac{1}{\Delta x \Delta \omega_i} \int_{x_{n-1}}^{x_n} \int_{\omega_{i-\frac{1}{2}}}^{\omega_{i+\frac{1}{2}}} (\Psi_E - \Psi_I) \frac{S_\phi}{\rho u} d\omega dx}_{\text{Source: IV}} \quad (\text{D.2})$$

The finite-difference analogues of each term in equation (D.2) will now be considered separately.

D.2.1 Axial Convection: I

It is assumed, for the purpose of integrating term I, that $(\Psi_E - \Psi_I) \frac{\partial \phi}{\partial x}$ is invariant with respect to ω over the control volume region. This leads to,

$$\begin{aligned} \text{Term I} &\approx (\Psi_E - \Psi_I)^{n-1} \int_{x_{n-1}}^{x_n} \frac{\partial \phi}{\partial x} dx \\ &\approx (\Psi_E - \Psi_I)^{n-1} \frac{(\phi_i^n - \phi_i^{n-1})}{\Delta x} \end{aligned} \quad (\text{D.3})$$

The approximation amounts to backward Euler differencing and is first-order accurate.

D.2.2 Cross-Stream Convection: II

A fully implicit specification is employed to discretise the cross-stream convection term. This implies that variations of $\frac{\partial \phi}{\partial \omega}$ in the x -direction are ignored. This is inconsistent with the development of the difference approximation for term I where variations in the ω -direction were assumed to be negligible; such seemingly contradictory assumptions are deemed to be of secondary importance if Δx and $\Delta \omega$ are sufficiently 'small', i.e. if the mesh sizes are such that the solution is effectively grid-independent. Thus,

$$\begin{aligned} \text{Term II} &\approx \frac{1}{\Delta \omega_i} \int_{\omega_{i-\frac{1}{2}}}^{\omega_{i+\frac{1}{2}}} (a^{n-1} + b^{n-1} \omega) \frac{\partial \phi^n}{\partial \omega} d\omega \\ &= \underbrace{\frac{1}{\Delta \omega_i} \int_{\omega_{i-\frac{1}{2}}}^{\omega_{i+\frac{1}{2}}} \frac{\partial}{\partial \omega} \{ (a^{n-1} + b^{n-1} \omega) \phi^n \} d\omega}_{\text{IIa}} \\ &\quad - \underbrace{\frac{1}{\Delta \omega_i} \int_{\omega_{i-\frac{1}{2}}}^{\omega_{i+\frac{1}{2}}} \phi^n \frac{\partial}{\partial \omega} (a^{n-1} + b^{n-1} \omega) d\omega}_{\text{IIb}} \end{aligned} \quad (\text{D.4})$$

Consider terms IIa and IIb separately,

$$\begin{aligned} \text{Term IIa} &= \frac{1}{\Delta \omega_i} \left[(a^{n-1} + b^{n-1} \omega) \phi^n \right]_{\omega_{i-\frac{1}{2}}}^{\omega_{i+\frac{1}{2}}} \\ &= \frac{1}{\Delta \omega_i} \left[(a^{n-1} + b^{n-1} \omega_{i+\frac{1}{2}}) \phi_{i+\frac{1}{2}}^n - (a^{n-1} + b^{n-1} \omega_{i-\frac{1}{2}}) \phi_{i-\frac{1}{2}}^n \right] \\ &= \frac{1}{\Delta \omega_i} \left[G_{i+\frac{1}{2}} \left(\frac{\phi_i^n + \phi_{i+1}^n}{2} \right) - G_{i-\frac{1}{2}} \left(\frac{\phi_i^n + \phi_{i-1}^n}{2} \right) \right] \end{aligned} \quad (\text{D.5})$$

where $G_{i\pm\frac{1}{2}} \equiv a^{n-1} + b^{n-1} \left(\frac{\omega_{i\pm 1} + \omega_i}{2} \right)$.

$$\text{Term IIb} \approx \frac{1}{\Delta\omega_i} \phi_i^n \left[a^{n-1} + b^{n-1} \omega \right]_{\omega_{i-\frac{1}{2}}}^{\omega_{i+\frac{1}{2}}} = \frac{1}{\Delta\omega_i} \phi_i^n \left(G_{i+\frac{1}{2}} - G_{i-\frac{1}{2}} \right) \quad (\text{D.6})$$

In arriving at equation (D.5) it is assumed that ϕ is a piecewise linear function of ω between (ω_{i-1}, ω_i) and (ω_i, ω_{i+1}) which implies central differencing. This type of differencing is second-order accurate (in ω space). By combining the two contributions to the cross-stream convection, the approximation becomes:

$$\text{Term II} \approx \frac{1}{\Delta\omega_i} \left[G_{i+\frac{1}{2}} \left(\frac{\phi_{i+1}^n - \phi_i^n}{2} \right) + G_{i-\frac{1}{2}} \left(\frac{\phi_i^n - \phi_{i-1}^n}{2} \right) \right] \quad (\text{D.7})$$

D.2.3 Cross-Stream Diffusion: III

A fully implicit formulation is adopted once again, so that:

$$\begin{aligned} \text{Term III} &\approx \frac{1}{\Delta\omega_i} \int_{\omega_{i-\frac{1}{2}}}^{\omega_{i+\frac{1}{2}}} \frac{\partial}{\partial\omega} \left\{ \left(\frac{\rho ur^2 \Gamma_\phi}{(\Psi_E - \Psi_I)} \right)^{n-1} \frac{\partial \phi^n}{\partial\omega} \right\} d\omega \\ &= \frac{1}{\Delta\omega_i} \left[\left(\frac{\rho ur^2 \Gamma_\phi}{(\Psi_E - \Psi_I)} \right)^{n-1} \frac{\partial \phi^n}{\partial\omega} \right]_{\omega_{i-\frac{1}{2}}}^{\omega_{i+\frac{1}{2}}} \end{aligned} \quad (\text{D.8})$$

$\frac{\partial \phi^n}{\partial\omega}$ is approximated with an assumed linear variation of ϕ between the node points (i.e. central differencing is used), hence,

$$\text{Term III} \approx \frac{1}{\Delta\omega_i} \left[\frac{(\rho ur^2 \Gamma_\phi)_{i+\frac{1}{2}}^{n-1}}{(\Psi_E - \Psi_I)^{n-1}} \left(\frac{\phi_{i+1}^n - \phi_i^n}{\omega_{i+1} - \omega_i} \right) - \frac{(\rho ur^2 \Gamma_\phi)_{i-\frac{1}{2}}^{n-1}}{(\Psi_E - \Psi_I)^{n-1}} \left(\frac{\phi_i^n - \phi_{i-1}^n}{\omega_i - \omega_{i-1}} \right) \right] \quad (\text{D.9})$$

From the definition of the stream function, equation (5.3), $\delta\Psi \equiv \rho ur \delta y$ (ignoring the overbar and tildes for the time being) and from equation (5.4), $\delta\omega = \frac{\delta\Psi}{(\Psi_E - \Psi_I)}$. These expressions can be used in conjunction with the definitions appearing in equation (D.1) to develop the following relations:

$$\delta y_i \equiv \int_i^{i+1} dy = \int_i^{i+1} \frac{d\Psi}{\rho ur} = (\Psi_E - \Psi_I) \int_i^{i+1} \frac{d\omega}{\rho ur} \approx \frac{(\Psi_E - \Psi_I)}{(\rho ur)_{i+\frac{1}{2}}} (\omega_{i+1} - \omega_i) \quad (\text{D.10})$$

A similar analysis can be performed to show that,

$$\delta y_{i-1} = \frac{(\Psi_E - \Psi_I)}{(\rho ur)_{i-\frac{1}{2}}} (\omega_i - \omega_{i-1}) \quad (\text{D.11})$$

Substituting for $(\rho ur)_{i\pm\frac{1}{2}}^{n-1}$ in equation (D.9) using the previously derived relations yields the final form of the approximation for term III:

$$\text{Term III} \approx \frac{1}{\Delta\omega_i} \left[F_{i+\frac{1}{2}} (\phi_{i+1}^n - \phi_i^n) - F_{i-\frac{1}{2}} (\phi_i^n - \phi_{i-1}^n) \right] \quad (\text{D.12})$$

where $F_{i+\frac{1}{2}} \equiv \frac{(r\Gamma_\phi)_{i+\frac{1}{2}}^{n-1}}{\delta y_i^{n-1}}$ and $F_{i-\frac{1}{2}} \equiv \frac{(r\Gamma_\phi)_{i-\frac{1}{2}}^{n-1}}{\delta y_{i-1}^{n-1}}$.

D.2.4 Source Term: IV

The expression that needs to be evaluated is,

$$\text{Term IV} = \frac{1}{\Delta x \Delta \omega_i} \int_{x_{n-1}}^{x_n} \int_{\omega_{i-\frac{1}{2}}}^{\omega_{i+\frac{1}{2}}} (\Psi_E - \Psi_I) \frac{S_\phi}{\rho u} d\omega dx \quad (\text{D.13})$$

By invoking the relations used to derive equations (D.10) and (D.11), term IV can be written as,

$$\text{Term IV} \approx \frac{1}{\Delta x \Delta \omega_i} \int_{x_{n-1}}^{x_n} \int_{y_{i-\frac{1}{2}}^{n-1}}^{y_{i+\frac{1}{2}}^{n-1}} r S_\phi dy dx \approx \frac{1}{\Delta \omega_i} \int_{y_{i-\frac{1}{2}}^{n-1}}^{y_{i+\frac{1}{2}}^{n-1}} r^{n-1} S_\phi^n dy \quad (\text{D.14})$$

The source term is effectively assumed to act at node (x_n, ω_i) - as far as the representation of the pde at the point in question is concerned, there is no approximation introduced with this particular representation. S_ϕ can then be placed outside the integral in equation (D.14). The integration of $r^{n-1} dy$ can be approximated by:

$$\text{Term IV} \approx \frac{1}{\Delta \omega_i} r_i^{n-1} S_{\phi,i}^n \left(y_{i+\frac{1}{2}}^{n-1} - y_{i-\frac{1}{2}}^{n-1} \right) \quad (\text{D.15})$$

The source term is decomposed using a quasi-linearisation²,

$$S_{\phi,i}^n \approx S_{\phi,i}^{n-1} - S_{\phi,i}'^{n-1} \phi_i^n \quad (\text{D.16})$$

The approximation involved in equation (D.16) relates to the representation of $S_{\phi,i}$ at time level n by $S_{\phi,i}$ and $S_{\phi,i}'$ at $n-1$. The quasi-linear decomposition is performed out of convenience. To elaborate slightly more on this form of decomposition, consider equation (C.13) when the evolution of ϕ is assumed to be affected only by the source terms,

$$\frac{d\phi}{dx} = S_\phi \quad (\text{D.17})$$

Discretising this expression using upwind differencing yields,

$$\phi^n \approx \phi^{n-1} + \Delta x S_\phi^n \quad (\text{D.18})$$

In this situation if $S_\phi^n \leq 0$, ϕ^n could become negative - especially if the forward step size is too large. Now consider,

$$\frac{d\phi}{dx} = S_\phi - S_\phi' \phi \quad (\text{D.19})$$

Discretising equation (D.19) so that ϕ , appearing on the rhs, is now based on a time level n ,

$$\phi_n = \frac{\phi_{n-1} + \Delta x S_\phi^{n-1}}{1 + \Delta x S_\phi'^{n-1}} \quad (\text{D.20})$$

If S_ϕ and S_ϕ' are chosen to be greater than zero, then according to equation (D.20) $\phi^n \geq 0$; this is an advantageous property, particularly for quantities like k and ε which are (by definition) positive

²The decomposition is 'quasi-linear' since if $S_{\phi,i}$ and $S_{\phi,i}'$ were independent of ϕ_i , it would be exactly linear.

ϕ	Γ_ϕ	$\mathcal{S}_\phi$	$\mathcal{S}'_\phi$
$\tilde{u}$	μ	$-\frac{1}{r} \frac{\partial}{\partial r} \left(r \bar{\rho} \widetilde{u''v''} \right) + \bar{\rho} g_x$	0
k	$\frac{\mu_t}{\sigma_k}$	$\mu_t \left(\frac{\partial \tilde{u}}{\partial r} \right)^2$	$\bar{\rho} \frac{\varepsilon}{k}$
ε	$\frac{\mu_t}{\sigma_\varepsilon}$	$C_{\varepsilon_1} \frac{\varepsilon}{k} \mu_t \left(\frac{\partial \tilde{u}}{\partial r} \right)^2$	$C_{\varepsilon_2} \bar{\rho} \frac{\varepsilon}{k}$
ε^a	$C_\varepsilon \bar{\rho} \frac{k}{\varepsilon} \widetilde{v''^2}$	$-C_{\varepsilon_1} \frac{\varepsilon}{k} \bar{\rho} \widetilde{u''v''} \frac{\partial \tilde{u}}{\partial r}$	$C_{\varepsilon_2} \bar{\rho} \frac{\varepsilon}{k}$
$\widetilde{u''^2}$	$C_s \bar{\rho} \frac{k}{\varepsilon} \widetilde{v''^2}$	$(C_2 + 2C_3 - 2) \bar{\rho} \widetilde{u''v''} \frac{\partial \tilde{u}}{\partial r} + \frac{2}{3} (C_1 - 1) \bar{\rho} \varepsilon$	$C_1 \bar{\rho} \frac{\varepsilon}{k}$
$\widetilde{w''^2} + \widetilde{v''^2}$	$C_s \bar{\rho} \frac{k}{\varepsilon} \widetilde{v''^2}$	$-(C_2 + 2C_3) \bar{\rho} \widetilde{u''v''} \frac{\partial \tilde{u}}{\partial r} + \frac{4}{3} (C_1 - 1) \bar{\rho} \varepsilon$	$C_1 \bar{\rho} \frac{\varepsilon}{k}$
$\widetilde{w''^2} - \widetilde{v''^2}$	$C_s \bar{\rho} \frac{k}{\varepsilon} \widetilde{v''^2}$	$(3C_2 + 2C_3) \bar{\rho} \widetilde{u''v''} \frac{\partial \tilde{u}}{\partial r}$	$C_1 \bar{\rho} \frac{\varepsilon}{k} + 4C_s \bar{\rho} \frac{k}{\varepsilon} \frac{\widetilde{w''^2}}{r^2}$
$\widetilde{u''v''}$	$C_s \bar{\rho} \frac{k}{\varepsilon} \widetilde{v''^2}$	$\left[C_4 k - \left(\frac{3}{2} C_2 + C_3 \right) \widetilde{u''^2} \right] \bar{\rho} \frac{\partial \tilde{u}}{\partial r} + (C_3 - 1) \widetilde{v''^2} \bar{\rho} \frac{\partial \tilde{u}}{\partial r}$	$C_1 \bar{\rho} \frac{\varepsilon}{k} + C_s \bar{\rho} \frac{k}{\varepsilon} \frac{\widetilde{w''^2}}{r^2}$

Table D.1: The terms grouped into the diffusion coefficient and source terms in the finite-difference formulation are detailed. The equations are written down in appendix B. The value of the model constants are assigned in table 2.1 (page 46).

^aThe contents of this row correspond to the use of the viscous dissipation rate equation with a second moment closure.

definite. For the equations written down in appendix B, the $\mathcal{S}'_\phi$ are constructed to be positive definite. For eddy viscosity closures $\mathcal{S}_\phi$ can also be constructed to be greater than zero thereby ensuring that ϕ^n will not become negative as a result of the source term discretisation. With this particular turbulence model, $\mathcal{S}_\phi$ represents the generation rate of ϕ and $\mathcal{S}'_\phi$ its destruction rate. With the Reynolds stress closures $\mathcal{S}_\phi$ cannot be guaranteed to be positive definite, although very often it is. However, provided Δx is not too large, it is found that this decomposition works well for this particular turbulence model too.

Table D.1 shows the groupings for the various equations that are solved as part of the velocity field calculation. The equations for $\widetilde{v''^2}$ and $\widetilde{w''^2}$ (written down in appendix B) include the term $2C_s \bar{\rho} \frac{k}{\varepsilon} \widetilde{w''^2} \frac{(\widetilde{v''^2} - \widetilde{w''^2})}{r^2}$ which is associated with transport. Near the centre-line $\widetilde{w''^2} = \widetilde{v''^2}$ by symmetry. Therefore, from the numerical point of view, the treatment of $\frac{(\widetilde{v''^2} - \widetilde{w''^2})}{r^2}$ where both the numerator and denominator tend to zero could be problematic. This can be alleviated by solving for $(\widetilde{w''^2} + \widetilde{v''^2})$ which eliminates the troublesome quotient term from its modelled evolution equation. In the case of $(\widetilde{w''^2} - \widetilde{v''^2})$, the term still appears, however, in this case $(\widetilde{w''^2} - \widetilde{v''^2})$ is the subject of the equation to be solved. So if the term $-\frac{(\widetilde{w''^2} - \widetilde{v''^2})}{r^2}$ is very large near the centre-line,

it will be the prominent term in the balance equation for $(\widetilde{w'^2} - \widetilde{v'^2})$ and thereby ensure that $(\widetilde{w'^2} - \widetilde{v'^2}) \rightarrow 0$ as $r \rightarrow 0$. All terms in table D.1 which involve the constant C_s arise from the transport; nevertheless some of these terms are included as $\mathcal{S}_\phi$ and $\mathcal{S}'_\phi$ because they do not conform to the general form for the transport term in equation (C.13), i.e. $\frac{1}{r} \frac{\partial}{\partial r} (r \Gamma_\phi \frac{\partial \phi}{\partial r})$.

Substituting equation (D.16) into (D.15), together with $\Delta y_i \equiv \frac{1}{2}(y_{i+1} - y_{i-1}) \approx (y_{i+\frac{1}{2}} - y_{i-\frac{1}{2}})$,

$$\text{Term IV} \approx \frac{1}{\Delta \omega_i} (\mathcal{S}_{\phi,i}^{n-1} - \mathcal{S}'_{\phi,i} \phi_i^n) r_i^{n-1} \Delta y_i^{n-1} \quad (\text{D.21})$$

D.2.5 The Finite-Difference Equation

Grouping the approximations from equations (D.3,D.7,D.12,D.21) into equation (D.2), and multiplying throughout by $\Delta \omega_i$ yields,

$$\begin{aligned} \underbrace{\frac{(\Psi_E - \Psi_I)^{n-1}}{\Delta x} \Delta \omega_i (\phi_i^n - \phi_i^{n-1})}_{\text{axial convection: I}} &+ \underbrace{G_{i+\frac{1}{2}} \left(\frac{\phi_{i+1}^n - \phi_i^n}{2} \right) + G_{i-\frac{1}{2}} \left(\frac{\phi_i^n - \phi_{i-1}^n}{2} \right)}_{\text{cross-stream convection: II}} \\ &= \underbrace{F_{i+\frac{1}{2}} (\phi_{i+1}^n - \phi_i^n) - F_{i-\frac{1}{2}} (\phi_i^n - \phi_{i-1}^n)}_{\text{cross-stream diffusion: III}} \\ &+ \underbrace{(\mathcal{S}_{\phi,i}^{n-1} - \mathcal{S}'_{\phi,i} \phi_i^n) r_i^{n-1} \Delta y_i^{n-1}}_{\text{source: IV}} \end{aligned} \quad (\text{D.22})$$

In equation (D.22) the terms involving F , $\mathcal{S}_\phi$ and $\mathcal{S}'_\phi$ are dependent on ϕ , whereas the rest of the terms are the same regardless of the dependent variable. This can be put to use in order to minimise the effort of constructing an algorithm for solving for all ϕ . Combining the respective ϕ terms together, the final form of the fde becomes,

$$\begin{aligned} &\left[\frac{\Delta \omega_i}{\Delta x} (\Psi_E - \Psi_I)^{n-1} + \left(F_{i+\frac{1}{2}} - \frac{1}{2} G_{i+\frac{1}{2}} \right) + \left(F_{i-\frac{1}{2}} + \frac{1}{2} G_{i-\frac{1}{2}} \right) + \mathcal{S}'_{\phi,i} r_i^{n-1} \Delta y_i^{n-1} \right] \phi_i^n \\ &= \left(F_{i+\frac{1}{2}} - \frac{1}{2} G_{i+\frac{1}{2}} \right) \phi_{i+1}^n + \left(F_{i-\frac{1}{2}} + \frac{1}{2} G_{i-\frac{1}{2}} \right) \phi_{i-1}^n \\ &+ \left[\frac{\Delta \omega_i}{\Delta x} (\Psi_E - \Psi_I)^{n-1} \phi_i^{n-1} + \mathcal{S}_{\phi,i}^{n-1} r_i^{n-1} \Delta y_i^{n-1} \right] \end{aligned} \quad (\text{D.23})$$

Equation (D.23) can be written more compactly as,

$$\mathcal{E}_i \phi_i^n = \mathcal{P}_i \phi_{i+1}^n + \mathcal{Q}_i \phi_{i-1}^n + \mathcal{J}_i \quad (\text{D.24})$$

where the definitions of the coefficients are clear by comparing equations (D.24) and (D.23). Assuming that there are $N + 1$ grid nodes distributed in the ω direction, equation (D.24) can be applied to all internal nodes ranging from $3 \leq i \leq N - 1$. The treatment of the remaining nodes will be discussed next.

D.3 The Treatment of the Boundaries

Three different types of 'boundaries' appear in the configurations under investigation; these are walls, free stream and axes of symmetry. The flows investigated all involve the jet configuration. Therefore in all cases the I boundary (figure 5.1) is an axis of symmetry. The *turbulent jets* that

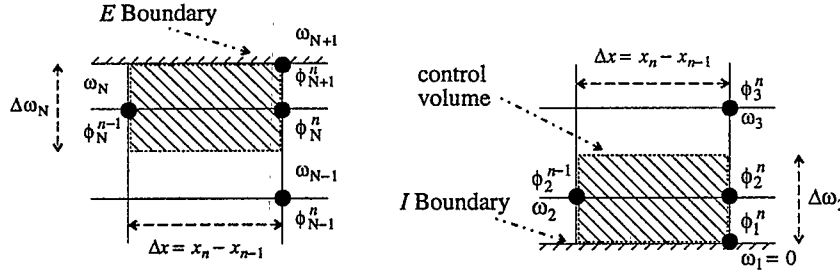


Figure D.2: Schematic of the control volumes used to derive the difference approximations at the I and E boundaries. Note, $\omega_3 - \omega_2 = 2(\omega_2 - \omega_1)$; similarly $\omega_N - \omega_{N-1} = 2(\omega_{N+1} - \omega_N)$. The boundary can be a wall, an axis of symmetry or a free stream.

are calculated all issue into unconfined surrounds, so that the effect of solid boundaries on the evolution of the jet can be ignored; in this case the E boundary corresponds to the free stream. However, a confined *laminar* jet diffusion flame is investigated; in this case the wall shear stress can be calculated in terms of the velocity gradient at the wall.

The construction of the finite-difference approximations follows exactly the same procedure as for the internal nodes. The only difference lies in the form of the control volumes over which the same governing pde's are integrated and then discretised. The control volumes used for the treatment of the I and E boundaries are shown in figure D.2

Integration of equation (D.2) over the region ω_1 to $\omega_{2+\frac{1}{2}}$ leads to,

$$\begin{aligned} & \left[\frac{\Delta\omega_2}{\Delta x} (\Psi_E - \Psi_I)^{n-1} + \left(F_{2+\frac{1}{2}} - \frac{1}{2}G_{2+\frac{1}{2}} \right) + (F_1 + G_1) + S_{\phi,2}^{n-1} r_2^{n-1} \Delta y_2^{n-1} \right] \phi_2^n \\ & = \left(F_{2+\frac{1}{2}} - \frac{1}{2}G_{2+\frac{1}{2}} \right) \phi_3^n + (F_1 + G_1) \phi_1^n \\ & + \left[\frac{\Delta\omega_2}{\Delta x} (\Psi_E - \Psi_I)^{n-1} \phi_2^{n-1} + S_{\phi,2}^{n-1} r_2^{n-1} \Delta y_2^{n-1} \right] \end{aligned} \quad (D.25)$$

where $\Delta y_2 = \frac{1}{2}(y_2 + y_3) - y_1$. At an axis of symmetry or a free stream there is a zero gradient condition, implying that diffusive fluxes are zero too; this condition can be implemented by setting $F_1 = F_{N+1} = 0$. Also $G_1 = a + b\omega_1 = 0$, since there is no entrainment at the axis of symmetry and $G_{N+1} = b$. By analogy, ϕ_N^n is obtained by integrating over the control volume shown in figure D.2, and the result is,

$$\begin{aligned} & \left[\frac{\Delta\omega_N}{\Delta x} (\Psi_E - \Psi_I)^{n-1} + (F_{N+1} - G_{N+1}) + \left(F_{N-\frac{1}{2}} + \frac{1}{2}G_{N-\frac{1}{2}} \right) + S_{\phi,N}^{n-1} r_N^{n-1} \Delta y_N^{n-1} \right] \phi_N^n \\ & = (F_{N+1} + G_{N+1}) \phi_{N+1}^n + \left(F_{N-\frac{1}{2}} + \frac{1}{2}G_{N-\frac{1}{2}} \right) \phi_{N-1}^n \\ & + \left[\frac{\Delta\omega_N}{\Delta x} (\Psi_E - \Psi_I)^{n-1} \phi_N^{n-1} + S_{\phi,N}^{n-1} r_N^{n-1} \Delta y_N^{n-1} \right] \end{aligned} \quad (D.26)$$

where $\Delta y_N = y_{N+1} - \frac{1}{2}(y_N + y_{N-1})$. With regards to the boundary conditions, the properties at node $\omega_{N+1}(=1)$ are given by the free stream values. At the centre-line a zero gradient condition is present, and this can be implemented by imposing a *parabolic* variation of ϕ over this region, i.e. $\phi \sim y^2$. This ensures that $\frac{\partial\phi}{\partial y} \rightarrow 0$, $\frac{\partial^2\phi}{\partial y^2} \rightarrow \text{constant}$, as $y \rightarrow 0$. Integration of the relation $(\rho u r) \delta y = (\Psi_E - \Psi_I) \delta \omega$ (defined in section 5.2.3) with an assumed quadratic variation in ρu near the centre-line indicates that for small values of y , $\omega \sim y^2$. Since $\phi \sim y^2$ is postulated over this region, this implies that $\phi \sim \omega$. So to obtain ϕ_1^n , a linear *extrapolation* can be performed using the known properties at $\omega = \omega_2$ and $\omega = \omega_3$. The conditions at $\omega = \omega_3$ are known from

the solution of the finite-difference equations for the internal nodes, whilst for ω_2 , equation (D.25) can be used since the coefficients of ϕ_1^n are zero.

D.4 The High Lateral Flux Modification

The description of the finite-difference procedure is not complete as yet. The cross-stream transport terms have been approximated using central differencing. The difficulties arising from the use of such a representation *for the convective term* can be highlighted using the one-dimensional steady convection/diffusion problem [42],

$$u \frac{\partial \phi}{\partial x} = \Gamma \frac{\partial^2 \phi}{\partial x^2} \quad u \geq 0, \Gamma \geq 0 \quad (\text{D.27})$$

where u and Γ are constant. With boundary conditions $x = 0, \phi = 0$ and $x = 1, \phi = 1$, the analytical solution demonstrates that ϕ is bounded in $[0, 1]$; this is a physically plausible result. If the equation is solved numerically and the derivatives are approximated using central differencing, then for *cell* Peclet numbers greater than two, ϕ can become negative³. Furthermore, the unboundness increases as Pe_x rises; this manifests itself as spatial oscillations ('wiggles') which could result in negative values for otherwise positive definite quantities. If upwind differencing were employed for the convection this problem would be averted.

The procedure that is adopted for the cross-stream transport (i.e. convection and diffusion) is to employ central differencing when applicable and to switch to upwind where necessary - this type of differencing is referred to as *hybrid* (also known as the *high lateral flux modification* [74]). With reference to equation (D.24), the factor which motivates the high lateral flux modification is that if $\mathcal{P}_i$ or $\mathcal{Q}_i$ become negative, then the weightings of ϕ_{i+1}^n and ϕ_{i-1}^n , respectively, serve to reduce ϕ_i^n . To avoid these coefficients from becoming negative the magnitude of the terms F and G , stemming from the diffusive and convective contributions (respectively), need to be examined more closely. The groupings which need to be taken into consideration are,

$$\begin{array}{lll} i = 2 & (F_1 + G_1), & \left(F_{2+\frac{1}{2}} - \frac{1}{2}G_{2+\frac{1}{2}} \right) \Rightarrow \text{First control volume} \\ i = 3 & \underbrace{\left(F_{3-\frac{1}{2}} + \frac{1}{2}G_{3-\frac{1}{2}} \right)}_{\mathcal{Q}_3}, & \underbrace{\left(F_{3+\frac{1}{2}} - \frac{1}{2}G_{3+\frac{1}{2}} \right)}_{\mathcal{P}_3} \Rightarrow \text{Second control volume} \\ & \vdots & \vdots \\ i = N & \left(F_{N-\frac{1}{2}} + \frac{1}{2}G_{N-\frac{1}{2}} \right), & (F_{N+1} - G_{N+1}) \Rightarrow N^{\text{th}} \text{ control volume} \end{array}$$

These groupings are best understood in terms of the schematic shown in figure D.3.

The manner in which the high lateral flux modification is applied is as follows:

- If $\frac{1}{2}|G_{i+\frac{1}{2}}| \leq F_{i+\frac{1}{2}}$, then no modification is applied. Thus, for example, $\mathcal{P}_2 = F_{2+\frac{1}{2}} - \frac{1}{2}G_{2+\frac{1}{2}} \geq 0$ and $\mathcal{Q}_3 = F_{3-\frac{1}{2}} + \frac{1}{2}G_{3-\frac{1}{2}} \geq 0$. Note that $F_{2+\frac{1}{2}} = F_{3-\frac{1}{2}}$ and $G_{2+\frac{1}{2}} = G_{3-\frac{1}{2}}$ due to the conservative finite-differencing.
- If $\frac{1}{2}|G_{i+\frac{1}{2}}| \geq F_{i+\frac{1}{2}}$, then $F_{i+\frac{1}{2}} \Rightarrow \frac{1}{2}|G_{i+\frac{1}{2}}|$. In this situation, $\mathcal{P}_2 = \frac{1}{2}|G_{2+\frac{1}{2}}| - \frac{1}{2}G_{2+\frac{1}{2}} = G_{2+\frac{1}{2}}$ and $\mathcal{Q}_3 = \frac{1}{2}|G_{2+\frac{1}{2}}| + \frac{1}{2}G_{2+\frac{1}{2}} = 0$ [§].

³The cell Peclet number, Pe_x , is defined as $\frac{u\Delta x}{\Gamma}$.

[§]Note that the definitions of F and G , stemming from equations (D.12) and (D.5) respectively, imply that the former is always positive and the latter negative. This explains why $\mathcal{P}_2 = G_{2+\frac{1}{2}}$ and $\mathcal{Q}_3 = 0$

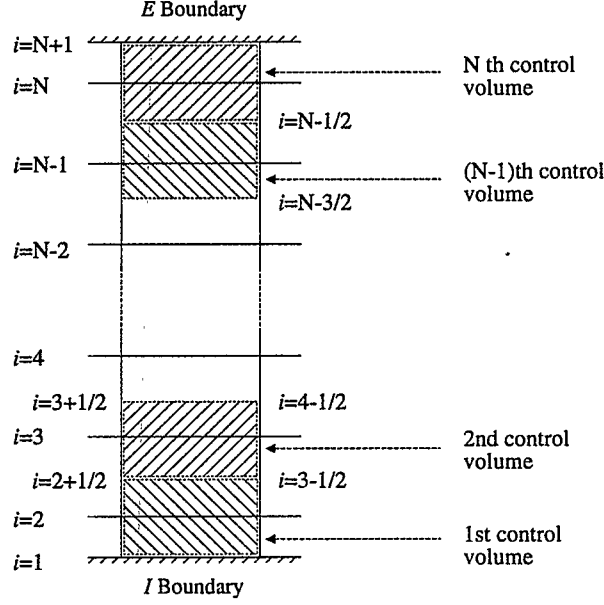


Figure D.3: Schematic of the control volumes along a given radial plane, indicating their relative placement and the associated notation. Because the finite-differencing is *conservative*, the fluxes leaving the top face of the first control volume is identically equal to the fluxes entering the bottom face of the second; i.e. $F_{2+\frac{1}{2}} = F_{3-\frac{1}{2}}$ and $G_{2+\frac{1}{2}} = G_{3-\frac{1}{2}}$

Since $F \geq 0$ and $G \leq 0$, this implies that the condition $\mathcal{P}_i \geq 0$ is always satisfied and that only $\mathcal{Q}_i$ can become negative. However, because the finite-differencing is conservative, even the groupings for $\mathcal{P}$ need to be checked according to the test above, since its fluxes are carried over to the $\mathcal{Q}$ term in the adjacent cell.

The high lateral flux modification can be viewed as a change in the diffusion coefficient, Γ_ϕ . F is effectively changed, but since r and δy are unaffected at a given point, the implication is that Γ_ϕ has been *increased*. In a region of the flow where $\frac{1}{2}|G_{i+\frac{1}{2}}| \geq F_{i+\frac{1}{2}}$, the effect of the modification on equation (D.23) is to represent the transport in terms of $\frac{1}{\Delta\omega_i} G_{i+\frac{1}{2}} (\phi_{i+1}^n - \phi_i^n)$; this is upwind differencing. If upwind differencing were to be employed for the convective terms right from the start, none of the above-mentioned manipulations would be required. However, the advantage of using 'hybrid' differencing is that it allows, at least under certain circumstances, for a more accurate approximation of the governing pde.

D.5 The Calculation of the Cross Stream Distance

The finite-difference procedure described thus far operates in the transformed (x, ω) space. Nevertheless, when the profiles of the variables are to be examined, they should be expressed in terms of physical space coordinates so that a comparison with experimental measurements can be made. Equations (D.12) and (D.21) also show that r is required to compute the coefficients of some of the terms in the finite-difference approximations. Out of convenience, the initial y -distribution is chosen to be uniform, i.e. $\delta y \approx \text{constant}$. Equations (5.3) and (5.4) demonstrate that $\delta\omega \propto r \delta y$, therefore a uniform initial y distribution implies that the ω grid is non-uniform and increases in width in proportion to the distance from the I boundary. Once this ω distribution is constructed, it

is maintained until the end of the simulation; in other words $\delta\omega$ does not change. However, δy is affected by the local density and velocity fields and so there is no reason why this should remain fixed. This is a potential disadvantage of the Patankar-Spalding method in that there is little (if no) control, by the analyst, over the mesh spacing as the flow evolves (particularly when coupled to a Monte Carlo simulation).

The first step is to evaluate $(\Psi_E - \Psi_I)^n$ - this quantity is only a function of the axial distance, so starting with equation (5.6), the following first-order accurate approximation can be written,

$$(\Psi_E - \Psi_I)^n = (\Psi_E - \Psi_I)^{n-1} + (r_I \dot{m}_I - r_E \dot{m}_E) (x_n - x_{n-1}) \quad (D.28)$$

From the definition of ω , equation (5.4), the following differential relation can be expressed,

$$\delta\omega = \frac{\partial\omega}{\partial\Psi} \delta\Psi + \frac{\partial\omega}{\partial\Psi_I} \delta\Psi_I + \frac{\partial\omega}{\partial\Psi_E} \delta\Psi_E \quad (D.29)$$

The objective of this section is to determine the value y for a given axial location. Since Ψ_I and Ψ_E are only functions of x , $\delta\Psi_I = \delta\Psi_E = 0$ under the present circumstances. Furthermore, from the definition of the stream function, equation (5.3), $\delta\Psi \approx \rho u r \delta y$; substituting these relations into equation (D.29),

$$r \delta y = (\Psi_E - \Psi_I) \frac{\delta\omega}{\rho u} \quad (D.30)$$

where $r = r_I + y$. Integration of equation (D.30) together with the definition,

$$\mathcal{I} \equiv (\Psi_E - \Psi_I) \int_o^\omega \frac{d\Omega}{\rho u} \quad (D.31)$$

leads to a quadratic equation in y^2 , which when solved for y has the following (positive) root,

$$y = -r_I + \left(r_I^2 + 2\mathcal{I} \right)^{\frac{1}{2}} \quad (D.32)$$

For an axisymmetric flow with the I boundary lying along the centre-line, $r_I = 0$, so that,

$$y = \sqrt{2\mathcal{I}} \quad (D.33)$$

Consequently, once ρ , u and $(\Psi_E - \Psi_I)$ are known along any cross-stream section, the corresponding y values can be obtained after evaluating the $\mathcal{I}$ integral. The details of how this integral is calculated can be found in the book by Patankar and Spalding [74].

D.6 The Treatment of Confined Flows: Calculating $\frac{dp}{dx}$

In this section the manner in which $\frac{dp}{dx}$ is calculated is briefly described. The details of the implementation are described in [74].

For confined flows the boundary layer and the pressure distribution interact appreciably; in practice $\frac{dp}{dx}$ is unknown and needs to be obtained as part of the calculation procedure. Conventional pressure-correction techniques would pose limitations on the speed and economy of the solution procedure which has so far been achieved (in part) by the use of linear fde's, their grouping into a tri-diagonal matrix and upon the avoidance of iteration. A somewhat heuristic approach was

proposed by Patankar and Spalding [74] so that $\frac{dp}{dx}$ could be approximated without bringing any serious detrimental effect to the efficiency of the calculation procedure.

The idea behind obtaining $\frac{dp}{dx}$ is that *it should take whatever value that will allow the boundaries of the flow to coincide with the walls of the duct*. Consequently $\frac{dp}{dx}$ is initially estimated, a forward step is performed and then, when the resulting cross-section of the flow is found to differ from that which the duct provides, the knowledge of the discrepancy guides the choice of $\frac{dp}{dx}$ for the next step. This practice is 'accurate' provided the step sizes are sufficiently 'small'.

The area of the duct (A_D) is obviously fixed by the geometrical constraints. However, the area of the flow is given by,

$$A_f = 2\pi \int_0^{y_{N+1}} r dr = 2\pi \int_{\Psi_I}^{\Psi_E} \frac{d\Psi'}{\rho u} \quad (D.34)$$

The contributions to the pressure gradient are evaluated through a force-momentum equation balance. The terms contributing to $\frac{dp}{dx}$ are the wall shear stress and a term involving an area gradient, $\frac{dA}{dx}$. So, $\frac{dp}{dx} \propto (\frac{dA}{dx} - |\tau_w|)$ and the contribution from the wall shear stress is generally larger (i.e. more negative) than the area gradient. The latter quantity is defined as:

$$\frac{dA}{dx} \equiv \frac{A_D^n - A_f^{n-1}}{x_n - x_{n-1}} \quad (D.35)$$

From equation (D.35), if $A_f^{n-1} \leq A_D^n$, then $\frac{dA}{dx} \geq 0$, so that the overall value of $\frac{dp}{dx}$ will be less negative. This means that the mean flow velocity *decreases* and by continuity, the area of the *flow* has to increase, thereby counteracting the original discrepancy.

Appendix E

Implementation of the Scalar Pdf Equation

The starting point for the discussion on the implementation of the scalar pdf equation is equation (5.9),

$$\tilde{P}(\underline{\psi}; x + \Delta x, \omega) \approx \underbrace{(\mathbf{I} + \Omega \Delta t)}_{\text{Source}} \underbrace{(\mathbf{I} + \Xi \Delta t)}_{\text{Mixing}} \underbrace{(\mathbf{I} + \mathbf{T} \Delta t)}_{\text{Transport}} \tilde{P}(\underline{\psi}; x, \omega)$$

The operator splitting technique allows for the effect of each physical process on the pdf to be considered *separately*. The simulation of the processes of transport and mixing are discussed in detail in this appendix. The simulation of chemical reaction is discussed in section (5.3.1.3).

E.1 Transport

The effect of transport on the pdf, $\tilde{P}(\underline{\psi}; x, \omega)$ can be expressed in the following manner,

$$\tilde{P}^t(\underline{\psi}; x_t, \omega) = (\mathbf{I} + \mathbf{T} \Delta t) \tilde{P}(\underline{\psi}; x, \omega) \quad (\text{E.1})$$

where x_t is a *notional* position such that $x \leq x_t \leq x + \Delta x$. By this, it is not implied that x_t lies somewhere in physical space at some fraction of the distance between nodes x_n and x_{n-1} (as depicted in figure D.1); it merely constitutes a symbolic notation for the intermediate solution, $\tilde{P}^{t\dagger}$. Equation (E.1) represents a convection/diffusion equation with no source terms. It was pointed out in section 4.13 that the Monte Carlo simulation represents an approximation to the *finite-difference* representation of the pde for the evolution of the pdf. In order to obtain such a fde, the same principles that were employed in appendix D can be used here too. The only significant difference lies in the fact that the Monte Carlo algorithm is an *explicit* scheme - it is difficult to envisage how the particle method could be made implicit. Because the particle simulation is explicit, the same is true for the finite-difference technique. Therefore, the pdf, $\tilde{P}^t(\underline{\psi}; x_t, \omega_i)$, is calculated using the properties at position x_{n-1} , namely, $\tilde{P}_{i\pm 1}^{n-1}(\underline{\psi}; x, \omega_{i\pm 1})$ and $\tilde{P}_i^{n-1}(\underline{\psi}; x, \omega_i)$. In this respect, the computational molecule for this situation is the mirror image of that illustrated

[†]It is important to bear in mind that no real interpretation can be given to the result $\tilde{P}^t$. In fact the solution cannot be assessed until the operations of mixing and chemical reaction have also been applied.

in figure D.1, with x_n being replaced by x_t . For an explicit scheme the fde can be written as:

$$\begin{aligned} \tilde{P}_i^t = & \frac{\Delta x}{\Delta \omega_i (\Psi_E - \Psi_I)^{n-1}} \left\{ \left(F_{i+\frac{1}{2}} - \frac{1}{2} G_{i+\frac{1}{2}} \right) \tilde{P}_{i+1}^{n-1} + \left(F_{i-\frac{1}{2}} + \frac{1}{2} G_{i-\frac{1}{2}} \right) \tilde{P}_{i-1}^{n-1} \right. \\ & \left. + \left[\frac{\Delta \omega_i}{\Delta x} (\Psi_E - \Psi_I)^{n-1} - \left(F_{i+\frac{1}{2}} - \frac{1}{2} G_{i+\frac{1}{2}} \right) - \left(F_{i-\frac{1}{2}} + \frac{1}{2} G_{i-\frac{1}{2}} \right) \right] \tilde{P}_i^{n-1} \right\} \quad (E.2) \end{aligned}$$

where $i = 3, \dots, N-1$. The similarities between equation (E.2) and the implicit form, given by equation (D.23), are clear. For the remaining i nodes, the form of the fde constructed for the boundary control volumes should be employed. The following definitions are introduced in order to simplify the form of equation (E.2).

$$\mathcal{P}_{i+\frac{1}{2}} \equiv \frac{\Delta x}{\Delta \omega_i (\Psi_E - \Psi_I)^{n-1}} \left(F_{i+\frac{1}{2}} - \frac{1}{2} G_{i+\frac{1}{2}} \right) \quad (E.3)$$

$$\mathcal{Q}_{i-\frac{1}{2}} \equiv \frac{\Delta x}{\Delta \omega_i (\Psi_E - \Psi_I)^{n-1}} \left(F_{i-\frac{1}{2}} + \frac{1}{2} G_{i-\frac{1}{2}} \right) \quad (E.4)$$

Probability theory demonstrates that,

$$\tilde{Q}_i^t = \int_{\psi} Q(\psi) \tilde{P}_i^t d\psi \quad (E.5)$$

where Q can denote any prescribed function of the scalar variables. $\tilde{Q}_i^t$ is the density-weighted average of Q at (x_t, ω_i) , based on the solution of the fde. Applying equation (E.5) to (E.2) leads to,

$$\tilde{Q}_i^t = \mathcal{P}_{i+\frac{1}{2}} \tilde{Q}_{i+1}^{n-1} + \mathcal{Q}_{i-\frac{1}{2}} \tilde{Q}_{i-1}^{n-1} + \left(1 - \mathcal{P}_{i+\frac{1}{2}} - \mathcal{Q}_{i-\frac{1}{2}} \right) \tilde{Q}_i^{n-1} \quad (E.6)$$

Let $\hat{Q}_i^t$ denote the ensemble average of Q at the same position due to the application of the transport process via the Monte Carlo simulation, where the ensemble is based on a finite sample size used to represent the pdf. The simulation of transport is achieved by selecting at random a 'certain number' of particles from nodes (x_n, ω_i) and $(x_n, \omega_{i\pm 1})^\dagger$; these particles form the new ensemble at (x_t, ω_i) as depicted in figure 4.2. It is important to note that the nodes do not exchange particles, and that the number of particles at a node remains fixed. By definition of the ensemble average (equation (2.25)),

$$\hat{Q}_i^t = \frac{1}{\mathcal{N}_i} \sum_{j=1}^{\mathcal{N}_i} Q_{i,j}^t \quad (E.7)$$

where $\mathcal{N}_i$ is the number of particles at ω_i [‡] and $Q_{i,j}^t$ is the value of the function Q associated with the properties of the j^{th} particle at (x_t, ω_i) [¶]. The expected value of $\hat{Q}_i^t$ is $\tilde{Q}_i^t$.

Based on the 'dynamics' of the simulation of the transport term, it is possible to write,

$$\hat{Q}_i^t \approx \frac{1}{\mathcal{N}_i} \left[\sum_{j=1}^{n_i^+} Q_{i+1,j}^{n-1} + \sum_{j=n_i^++1}^{n_i^++n_i^-} Q_{i-1,j}^{n-1} + \sum_{j=n_i^++n_i^-+1}^{\mathcal{N}_i} Q_{i,j}^{n-1} \right] \quad (E.8)$$

[†]More precisely, the particles are sampled at random without replacement. This means that, at a given point, when a particle is randomly selected it is no longer allowed to be re-selected until node ω_i has been advanced from x_n to x_t .

[‡]The ω grid distribution does not change once it is generated and the number of particles per node remain fixed (but not necessarily equal). This implies that the number of particles per node depends only on the radial location; hence the notation $\mathcal{N}_i$, which indicates no dependency on x .

[¶]The definition of the ensemble average in equation (2.25) includes the density. However, equation (E.7) does not require these multipliers since the ensemble, $Q_{i,j}^t$, is defined to be representative of the density-weighted pdf. Consequently, the result shown in equation (E.7) automatically leads to a density-weighted statistic.

where n_i^+ and n_i^- are the number of particles selected at random from the ensembles at (x_n, ω_{i+1}) and (x_n, ω_{i-1}) , respectively. Equation (E.8) indicates that $(\mathcal{N}_i - n_i^+ + n_i^-)$ particles are randomly selected from node i at $x = x_{n-1}$ and retained in the new ensemble at $x = x_t$. The remaining particle properties are overwritten by those randomly selected from $\omega_{i\pm 1}$. The ordering of the particles at a node is immaterial, so that the first term on the rhs of equation (E.8) could have been, $\sum_{j=1}^{n_i^-} Q_{i-1,j}^{n-1}$ and consequently the second term, $\sum_{j=n_i^++1}^{n_i^++n_i^-} Q_{i+1,j}^{n-1}$. The following straightforward, yet revealing, manipulation can be applied to equation (E.8),

$$\begin{aligned} \hat{Q}_i^t \approx & \underbrace{\frac{n_i^+}{\mathcal{N}_i} \left[\frac{1}{n_i^+} \sum_{j=1}^{n_i^+} Q_{i+1,j}^{n-1} \right]}_{\text{I}} + \underbrace{\frac{n_i^-}{\mathcal{N}_i} \left[\frac{1}{n_i^-} \sum_{j=n_i^++1}^{n_i^++n_i^-} Q_{i-1,j}^{n-1} \right]}_{\text{II}} \\ & + \underbrace{\frac{(\mathcal{N}_i - n_i^+ - n_i^-)}{\mathcal{N}_i} \left[\frac{1}{(\mathcal{N}_i - n_i^+ - n_i^-)} \sum_{j=n_i^++n_i^-+1}^{\mathcal{N}_i} Q_{i,j}^{n-1} \right]}_{\text{III}} \end{aligned} \quad (\text{E.9})$$

If the mathematical expectation of equation (E.9) is taken, in the limit of $\mathcal{N}_i$ (and hence n_i^- and n_i^+) tending to infinity, terms I, II and III approach $\tilde{Q}_{i+1}^{n-1}$, $\tilde{Q}_{i-1}^{n-1}$ and $\tilde{Q}_i^{n-1}$, respectively. At the same time, $\hat{Q}_i^t \rightarrow \tilde{Q}_i^t$, so that in the limit equation (E.9) can be expressed as:

$$\tilde{Q}_i^t = \frac{n_i^+}{\mathcal{N}_i} \tilde{Q}_{i+1}^{n-1} + \frac{n_i^-}{\mathcal{N}_i} \tilde{Q}_{i-1}^{n-1} + \left(1 - \frac{n_i^+}{\mathcal{N}_i} - \frac{n_i^-}{\mathcal{N}_i} \right) \tilde{Q}_i^{n-1} \quad (\text{E.10})$$

If the Monte Carlo simulation for the transport operation is to approximate the finite-difference solution, then clearly the results of equations (E.6) and (E.10) should be the same; this is the basic requirement which ensures equivalence between the two approaches. By comparing these two equations (and using the definitions expressed in equations (E.3) and (E.4)), the following fundamental relations hold,

$$\frac{n_i^+}{\mathcal{N}_i} = \frac{\Delta x}{\Delta \omega_i (\Psi_E - \Psi_I)^{n-1}} \left(F_{i+\frac{1}{2}} - \frac{1}{2} G_{i+\frac{1}{2}} \right) \quad (\text{E.11})$$

$$\frac{n_i^-}{\mathcal{N}_i} = \frac{\Delta x}{\Delta \omega_i (\Psi_E - \Psi_I)^{n-1}} \left(F_{i-\frac{1}{2}} + \frac{1}{2} G_{i-\frac{1}{2}} \right) \quad (\text{E.12})$$

The discussion presented in the previous paragraphs and culminating in equations (E.11) and (E.12) describe how the solution at (x_{n-1}, ω_i) can be advanced to (x_t, ω_i) for the internal nodes. The treatment of $i = 1$ and $i = N + 1$ will now be described. In the present context the pdf equation is applied to turbulent jet diffusion flames issuing into unconfined surrounds, so that $i = 1$ corresponds to a symmetry axis and $i = N + 1$ is the free stream. The free stream condition is the most straightforward to specify, since the flow in that region is unaffected by the evolution of the jet. Since $\frac{\partial \bar{p}}{\partial x} = 0$, this means that the properties in this region are invariant with downstream position, and the pdf there is a δ function. This latter condition implies that all $\mathcal{N}_{N+1}$ particles have the same value, or alternatively, only one particle is sufficient to represent the properties there. At the centre-line the zero gradient condition is used to characterise the scalar boundary condition. This condition is achieved by requiring that the ensemble at (x_t, ω_2) is exactly the same as that at (x_t, ω_1) .

For the treatment of the scalar field the finite-difference scheme (and hence the Monte Carlo simulation) is fully explicit. It is well-known that to ensure the stability of such schemes, restrictions have to be placed on the forward step size. The stability requirements for the application of the Monte Carlo simulation stem from the need to avoid $n_i^+ \leq 0$, $n_i^- \leq 0$ and $\mathcal{N}_i - (n_i^+ + n_i^-) \leq 0$. However, from equations (E.11) and (E.12) it is clear that $n_i^\pm \propto \Delta x$. The manner in which the forward step size is chosen is simple and ad-hoc, partly because the main motivation in choosing Δx is that the solution is grid-independent, but also because the semi-implicit finite-difference procedure for the velocity field does not suffer from the step size restrictions of the Monte Carlo simulation. As the jet spreads downstream of the issuing nozzle its width increases, and based on this $\Delta x \equiv \ell y_{N+1}$, where y_{N+1} is the outermost radial position of the jet and ℓ is an arbitrary constant ($\ell \approx 0.01$). Δx is prevented from increasing too rapidly by specifying,

$$\Delta x_{new} = \min(\ell y_{N+1}, 1.05 \Delta x_{old}) \quad (\text{E.13})$$

In principle, if the forward step size is too large, the constraint,

$$\mathcal{N}_i - (n_i^+ + n_i^-) \geq 0 \quad (\text{E.14})$$

could be violated. Let Δx_i^* be the step size suitable for the Monte Carlo simulation of transport such that the inequality expressed in equation (E.14) is satisfied for all i . Dividing equation (E.14) by $\mathcal{N}_i$ and substituting equations (E.11) and (E.12) yields,

$$1 - \frac{\Delta x_i^*}{\Delta \omega_i (\Psi_E - \Psi_I)^{n-1}} \underbrace{\left(F_{i+\frac{1}{2}} - \frac{1}{2} G_{i+\frac{1}{2}} + F_{i-\frac{1}{2}} + \frac{1}{2} G_{i-\frac{1}{2}} \right)}_{\equiv \mathcal{H}_{i\pm\frac{1}{2}}} \geq 0 \quad (\text{E.15})$$

Whilst equation (E.14) constitutes the minimum requirement which can be placed on the selection of the particles, it can be argued that to obtain a 'meaningful' ensemble for $\hat{Q}_i^t$ (based on equation (E.9)), it is necessary to have finite numbers of particles selected from all the participating nodes. This implies that equation (E.15), which relates to the fractional number of particles sampled from node i , can be written as,

$$1 - \frac{\Delta x_i^*}{\Delta \omega_i (\Psi_E - \Psi_I)^{n-1}} \mathcal{H}_{i\pm\frac{1}{2}} = \Lambda_t \quad (\text{E.16})$$

where $0 \leq \Lambda_t \leq 1$, but a more plausible choice is $\Lambda_t = \frac{1}{3}$. In other words, Δx_i^* must be such that *in the event* $\Delta x_i^* \leq \Delta x^{\dagger\dagger}$ at least one third of the particles should come from (x_{n-1}, ω_i) , and the remainder from nodes $(x_{n-1}, \omega_{i\pm 1})$. When $\Delta x_i^* \geq \Delta x$, the implication of Λ_t is of no consequence. In fact tests indicated that the results appear to be quite insensitive to the precise value of Λ_t - for example starting with $\Lambda_t = \frac{1}{3}$, it was found that doubling and halving this value had virtually no observable effect on the results. This may be an indication that the sample size was suitably large and/or the forward step size was sufficiently small. To implement Δx_i^* into the solution procedure the following *integer* parameter is defined,

$$\mathcal{K}_t \equiv \text{nint} \left(\frac{\Delta x}{\Delta x_i^*} + \frac{1}{2} \right) \quad (\text{E.17})$$

where *nint* denotes the nearest integer and the addition of ' $\frac{1}{2}$ ' is there to yield the most conservative estimate for $\mathcal{K}_t$. Δx_i^* depends on the radial position, i , but in this procedure a single value of $\mathcal{K}_t$ is

^{††}Note that Δx is given by equation (E.13) and Δx_i^* from equation (E.16).

needed which satisfies equation (E.16) for all i . To achieve this, the most conservative (and hence largest) value of $\mathcal{K}_t$ is used. Substituting the smallest value of Δx_t^* (referred to as $\Delta x_{t,min}^*$) from equation (E.16) into (E.17) gives,

$$\mathcal{K}_{t,max} = \max \left[\text{nint} \left(\frac{\Delta x}{\Delta x_{t,min}^*} + \frac{1}{2} \right), 1 \right] \quad (\text{E.18})$$

So the transport operation is applied $\mathcal{K}_{t,max}$ times (with a forward step size of $\Delta x_{t,min}^*$). *During any one of these substeps, the mixing and reaction operations are also applied*¹.

Finally it is worth pointing out the following significant difference between the finite-difference approach and the corresponding Monte Carlo simulation. With the former technique the accuracy of the result increases as the grid size is refined, i.e. as $\Delta x, \delta\omega \rightarrow 0$. For the Monte Carlo procedure with a finite sample size, as $\Delta x \rightarrow 0$ the number of particles being randomly selected will tend to zero (as implied by equations (E.11) and (E.12)), with the result that there is very little observable transport. Clearly the missing link in this case is that as $\Delta x, \delta\omega \rightarrow 0$, this needs to be matched with $\mathcal{N}_i \rightarrow \infty, \forall i$.

E.2 Molecular Mixing

In this section the following operation is considered,

$$\tilde{\underline{P}}^m(\underline{\psi}; x_m, \omega) = (\mathbf{I} + \Xi \Delta t) \tilde{\underline{P}}^t(\underline{\psi}; x_t, \omega) \quad (\text{E.19})$$

This follows from equation (E.1). The mixing models are discussed in sections 4.6 and 4.7. Only the implementation features of modified Curl (with $\mathcal{A}(x) = 1$) and LMSE are considered in this appendix. Unlike the transport operation, molecular mixing is applied at a point in physical space to the notional particles residing there. Therefore internodal particle interactions do not occur. This exemplifies how molecular mixing brings about the evolution of the pdf in composition space and not physical space.

E.2.1 Modified Curl

This model can be summarised through the schematic in figure E.1, where the particle properties *after* the mixing operation are given by equation (4.20).

At a given point in physical space, comprising nodes $\omega_2, \dots, \omega_N$, the number of particles selected randomly without replacement is given by (cf. equation (4.16))²:

$$\mathcal{N}_{s,i} = 2\mathcal{B} \left(\frac{\Delta x}{\tilde{u}_i} \right) \frac{\varepsilon_i}{k_i} \mathcal{N}_i, \quad i = 2, \dots, N \quad (\text{E.20})$$

where $\mathcal{B} = 3$ for modified Curl. For any given pair of randomly selected particles, a pseudo-random number generator is again used to select a value for x , the quantity which controls the extent of mixing, as specified in equation (4.20). This same value of x is employed to calculate all the new scalar properties ($\alpha = 1, \dots, N+1$).

¹In fact the mixing operation also imposes forward step size restrictions, as described in section E.2. Therefore the whole operation involving transport, mixing and reaction can be applied more than $\mathcal{K}_{t,max}$ times depending on the value of $\mathcal{K}_{m,max}$ defined in the section dealing with the mixing terms

²Note that a distinction is made at this stage between the number of particles and *pairs* of particles randomly selected.

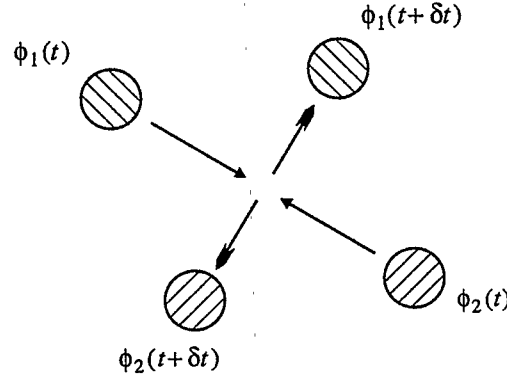


Figure E.1: Schematic of the mixing operation using modified Curl. The randomly selected particles are represented in terms of the properties $\phi_1(t)$ and $\phi_2(t)$. They mix along a line joining them in composition space and jump to the new properties $\phi_1(t + \delta t)$ and $\phi_2(t + \delta t)$. The extent of mixing is constrained by either no change in their original values, to full mixing at their common mean value.

It was pointed out in relation to equations (E.11) and (E.12) that the condition expressed in equation (E.14) can be violated since n_i^+ and n_i^- are proportional to Δx . A similar situation arises with regards to equation (E.20); $\mathcal{N}_{s,i}$ can be greater than $\mathcal{N}_i$ if Δx is too large or $\tilde{u}_i$ too small. To avoid such unphysical situations, the following requirement is imposed (in analogy with equation (E.16)),

$$\frac{\mathcal{N}_{s,i}}{\mathcal{N}_i} = 2\mathcal{B} \left(\frac{\Delta x_m^*}{\tilde{u}_i} \right) \frac{\varepsilon_i}{k_i} \equiv \Lambda_m, \quad i = 2, \dots, N \quad (\text{E.21})$$

where Δx_m^* is the step size which satisfies $\frac{\mathcal{N}_{s,i}}{\mathcal{N}_i} = \Lambda_m$ ($0 \leq \Lambda_m \leq 1$). Sensitivity analysis shows that the choice of Λ_m does not affect the final result significantly, and in fact by choosing Δx to be sufficiently small, the value of Λ_m becomes immaterial since the basic condition $\mathcal{N}_{s,i} \geq \mathcal{N}_i$ would not be violated. $\Lambda_m = \frac{1}{2}$ was chosen for the results presented in chapter 6 using modified Curl. In analogy with equations (E.17) and (E.18), $\mathcal{K}_{m,max}$ is defined as,

$$\mathcal{K}_{m,max} \equiv \max \left\{ nint \left[\max \left(\frac{\frac{\Delta x}{\tilde{u}_i}}{\frac{\Delta x_m^*}{\tilde{u}_i}} + \frac{1}{2} \right), \mathcal{K}_{t,max} \right] \right\} \quad (\text{E.22})$$

where $\frac{\Delta x_m^*}{\tilde{u}_i}$ and $\mathcal{K}_{t,max}$ are obtained from equations (E.21) and (E.18) respectively.

E.2.2 Linear Mean Square Estimation (LMSE) Closure

The starting point for discussing the implementation of this model is equation (4.14), re-written as:

$$\frac{d}{dt} \phi_{\alpha,i}^{(p)} = -\frac{C_D}{2} \frac{\varepsilon_i}{k_i} \left(\phi_{\alpha,i}^{(p)} - \langle \phi_{\alpha,i} \rangle \right) \quad \begin{cases} (p) = 1, \dots, \mathcal{N}_i \\ \alpha = 1, \dots, N+1 \\ i = 2, \dots, N \end{cases} \quad (\text{E.23})$$

This model is deterministic in the sense that a certain number of particles do not have to be randomly selected for mixing; *all* the particles are subjected to equation (E.23) at a given node i and for all scalar properties. Equation (E.23) can be discretised in the following manner,

$$\phi_{\alpha,i}^{(p),n+1} - \phi_{\alpha,i}^{(p),n} \approx -\frac{C_D}{2} \frac{\varepsilon_i}{k_i} \delta t_i \left(\phi_{\alpha,i}^{(p),n} - \langle \phi_{\alpha,i} \rangle^n \right) + \mathcal{O}(\delta t^2) \quad (\text{E.24})$$

where $\delta t_i \equiv \frac{\Delta x^\circ}{\tilde{u}_i}$ and $\Delta x^\circ = \frac{\Delta x}{\mathcal{K}_{t,max}}$ as given by equations (E.13) and (E.18). One potential source of problems with the above form of discretisation is that if δt_i is too large $\phi_{\alpha,i}^{(p)}$ can become unbounded. However, such a problem has not been encountered with the use of the above equation, presumably because the forward step size has been sufficiently small to avoid such problems. Nevertheless, to eliminate such undesirable features associated with the discretisation, the term on the rhs of equation (E.24) involving $\phi_{\alpha,i}^{(p),n}$ can be replaced with $\phi_{\alpha,i}^{(p),n+1}$. Solving for $\phi_{\alpha,i}^{(p),n+1}$ yields:

$$\phi_{\alpha,i}^{(p),n+1} = \frac{\phi_{\alpha,i}^{(p),n} + \frac{C_D}{2} \frac{\varepsilon_i}{k_i} \delta t_i \langle \phi_{\alpha,i} \rangle^n}{1 + \frac{C_D}{2} \frac{\varepsilon_i}{k_i} \delta t_i} \quad (\text{E.25})$$

With the use of equation (E.25), $\phi_{\alpha,i}^{(p),n+1} \geq 0$ regardless of the value of δt_i . In addition as $\delta t_i \rightarrow \infty$, $\phi_{\alpha,i}^{(p),n+1} \rightarrow \langle \phi_{\alpha,i} \rangle^n$, which implies that the result remains bounded.

It has already been pointed out that $\delta t_i \equiv \frac{\Delta x^\circ}{\tilde{u}_i}$ where $\Delta x^\circ = \frac{\Delta x}{\mathcal{K}_{t,max}}$. It is possible to impose a further constraint by comparing the relative sizes of δt_i and the mixing time scale. The latter time scale is defined as $(\frac{C_D}{2} \frac{\varepsilon_i}{k_i})^{-1}$. To develop an expression for such a constraint, the following definition is introduced:

$$\delta t_i^* \equiv \Lambda'_m \left(\frac{C_D}{2} \frac{\varepsilon_i}{k_i} \right)^{-1}, \quad 0 \leq \Lambda'_m \leq 1 \quad (\text{E.26})$$

where δt_i^* is the required time step at node i defined as some fraction of the mixing time scale. In this case a parameter, $\mathcal{K}_{m,max}$, as defined for the coalescence-dispersion model (equation (E.22)), can be used:

$$\mathcal{K}_{m,max} \equiv \max \left\{ \text{rint} \left[\max \left(\frac{\frac{\Delta x^\circ}{\tilde{u}_i}}{\delta t_i^*} \right) + \frac{1}{2} \right], \mathcal{K}_{t,max} \right\} \quad (\text{E.27})$$

The introduction of $\Lambda'_m \leq 1$ can only add to the run times and the effect on the results is hardly detectable for the inhomogeneous flows considered.

E.3 Simulation of Homogeneous, Isotropic Turbulence

In the present context, by homogeneous, isotropic turbulence it is meant a flow where the scalar pdf is invariant with position. Consequently there is no mean strain imposed on the flow, and the pdf evolves only in composition space; the dependence of quantities such as ε , k and $\mathcal{N}$ on position disappears. The simulation of transport, described in section E.1, becomes redundant as does the value of the local velocity, $\tilde{u}_i$. So for equations (E.20) and (E.25) the time step is now provided in terms of some fraction of the characteristic turbulent time scale ($\frac{k}{\varepsilon}$). A suitably small fraction is chosen (typically $\frac{1}{100}$ th) in order to ensure accurate solutions, and because of this $\mathcal{K}_{m,max}$ is always unity.

In the calculations reported in this thesis the value of $\frac{k}{\varepsilon}$ can be allowed to evolve with time ; based on the conditions prevailing in the homogeneous and isotropic turbulence, equations (2.39) and (2.50) reduce to:

$$\frac{dk}{dt} = -\varepsilon \quad (\text{E.28})$$

$$\frac{d\varepsilon}{dt} = -C_{\varepsilon_2} \frac{\varepsilon^2}{k} \quad (\text{E.29})$$

In equation (E.29) ε^2 can be eliminated by substituting from (E.28); furthermore, $\frac{d\varepsilon}{dt}$ can be substituted using equation (E.28) differentiated with respect to time. The final result is:

$$\frac{d^2 k}{dt^2} = C_{\varepsilon_2} \frac{\left(\frac{dk}{dt}\right)^2}{k} \quad (\text{E.30})$$

By introducing the substitution $\zeta = \frac{dk}{dt}$, equation (E.30) can be integrated using a separation of variables technique. Introducing the initial conditions specified above (with $t_o = 0$) leads to,

$$k = k_o \left[1 + (C_{\varepsilon_2} - 1) \left(\frac{\varepsilon}{k} \right)_o t \right]^{\frac{-1}{C_{\varepsilon_2} - 1}} \quad (\text{E.31})$$

Hence from equation (E.28),

$$\varepsilon = \varepsilon_o \left[1 + (C_{\varepsilon_2} - 1) \left(\frac{\varepsilon}{k} \right)_o t \right]^{\frac{-C_{\varepsilon_2}}{C_{\varepsilon_2} - 1}} \quad (\text{E.32})$$

Equations (E.31) and (E.32) were also used by Lakshmisha et al. [57] in their work relating to homogeneous, isotropic turbulence. Dividing equation (E.31) by (E.32) leads to the expression for the evolution of the characteristic turbulent time scale,

$$\frac{k}{\varepsilon} = \left(\frac{k}{\varepsilon} \right)_o + (C_{\varepsilon_2} - 1) t \quad (\text{E.33})$$

The implication of equation (E.33) is that the characteristic time scale of the energy containing motions increases with time - this is indicative of a decaying turbulence. The choice of the value of $\left(\frac{k}{\varepsilon}\right)_o$ is arbitrary since the aim of the homogeneous, isotropic turbulence calculations is to investigate the qualitative behaviour of the mixing models. In this respect the use of equation (E.33) could be altogether avoided and be replaced by a constant turbulence field (i.e. $\frac{k}{\varepsilon}$ is constant) - the qualitative behaviour of the results would be the same.

E.4 Evaluation of the Statistical Properties from the Discrete Ensemble

In this thesis predictions have been compared against a wide range of measurements in turbulent jet diffusion flames. The experimental data comes in the form of density-weighted or unweighted statistics. In certain cases the form of the data is dictated by the measurement technique; for example sampling probe measurements of species concentrations are conventionally viewed as being close to a density-weighted average, whilst fine-wire thermocouple measurements provide unweighted statistics. The transported pdf approach is advantageous in that both forms of statistics can be evaluated.

It is convenient to solve a density-weighted pdf equation when calculating variable density flows³. Formally, for any variable which is a function of the scalar field, $\mathcal{G} = \mathcal{G}(\phi)$,

$$\tilde{\mathcal{G}} \equiv \widetilde{\mathcal{G}(\phi)} = \int_{\psi} \mathcal{G}(\psi) \tilde{P}(\psi) d\psi = \lim_{N \rightarrow \infty} \frac{1}{N} \sum_{p=1}^N \mathcal{G}(\phi^{(p)}) \quad (\text{E.34})$$

³The is because the models for the turbulent transport and molecular mixing can then be carried over from the constant density situation, with the assumption that the effects associated with the fluctuating density field are accounted for through the density-weighting.

where $\mathcal{G}(\underline{\phi}^{(p)})$ is the value of the variable $\mathcal{G}$ for the p^{th} particle which has the property set $\underline{\phi}^{(p)}$. To obtain the unweighted average, $\bar{\mathcal{G}}$, the following can be applied:

$$\bar{\mathcal{G}} = \overline{\left(\frac{\mathcal{G}}{\varrho}\right)} \equiv \bar{\varrho} \left(\frac{\mathcal{G}}{\varrho}\right) = \bar{\varrho} \int_{\underline{\psi}} \frac{\mathcal{G}(\underline{\psi})}{\varrho(\underline{\psi})} \tilde{P}(\underline{\psi}) d\underline{\psi} = \bar{\varrho} \left[\lim_{\mathcal{N} \rightarrow \infty} \frac{1}{\mathcal{N}} \sum_{p=1}^{\mathcal{N}} \frac{\mathcal{G}(\underline{\phi}^{(p)})}{\varrho(\underline{\phi}^{(p)})} \right] \quad (\text{E.35})$$

In order to make use of equation (E.35), $\bar{\varrho}$ would have to be evaluated; the use of $\mathcal{G} = \varrho$ in equation (E.35) leads to the trivial result $\bar{\varrho} = \bar{\varrho}$. Instead, consider the following equation,

$$\int_{\underline{\psi}} \frac{1}{\varrho(\underline{\psi})} \tilde{P}(\underline{\psi}) d\underline{\psi} = \lim_{\mathcal{N} \rightarrow \infty} \frac{1}{\mathcal{N}} \sum_{p=1}^{\mathcal{N}} \frac{1}{\varrho(\underline{\phi}^{(p)})} \quad (\text{E.36})$$

The density-weighted pdf is defined as: $\bar{\varrho} \tilde{P}(\underline{\psi}) \equiv \varrho(\underline{\psi}) P(\underline{\psi})$, and since $\int_{\underline{\psi}} P(\underline{\psi}) d\underline{\psi} = 1$, it follows that,

$$\bar{\varrho} = \left[\lim_{\mathcal{N} \rightarrow \infty} \frac{1}{\mathcal{N}} \sum_{p=1}^{\mathcal{N}} \frac{1}{\varrho(\underline{\phi}^{(p)})} \right]^{-1} \quad (\text{E.37})$$

The fluctuating field statistics is given by,

$$\overline{\mathcal{G}''^n} = \int_{\underline{\psi}} (\mathcal{G}(\underline{\psi}) - \bar{\mathcal{G}})^n \tilde{P}(\underline{\psi}) d\underline{\psi} = \lim_{\mathcal{N} \rightarrow \infty} \frac{1}{\mathcal{N}} \sum_{p=1}^{\mathcal{N}} (\mathcal{G}(\underline{\phi}^{(p)}) - \bar{\mathcal{G}})^n \quad (\text{E.38})$$

The measurements and predictions are mainly confined to the mean and root-mean-square (rms) quantities, and so attention will focus on the case when $n = 2$. Equation (E.38) cannot be directly modified along the lines of equation (E.35) to yield an expression for $\overline{\mathcal{G}'^2}$, primarily because $\underline{\phi}^{(p)}$ refers to properties which are density-weighted and so $\mathcal{G}' \neq (\mathcal{G}(\underline{\phi}^{(p)}) - \bar{\mathcal{G}})$. A more straightforward approach is now described; the starting point is the following basic relations:

$$\left. \begin{aligned} \mathcal{G}'' &= \mathcal{G} - \bar{\mathcal{G}} \\ \mathcal{G}' &= \mathcal{G} - \bar{\mathcal{G}} \end{aligned} \right\} \mathcal{G}'' = (\bar{\mathcal{G}} - \bar{\mathcal{G}}) + \mathcal{G}' \quad (\text{E.39})$$

Squaring and averaging equation (E.39) leads to:

$$\begin{aligned} \overline{\mathcal{G}''^2} &= (\bar{\mathcal{G}} - \bar{\mathcal{G}})^2 + 2(\bar{\mathcal{G}} - \bar{\mathcal{G}}) \underbrace{\overline{\mathcal{G}'}}_{=0} + \overline{\mathcal{G}'^2} \\ \Rightarrow \overline{\mathcal{G}'^2} &= \overline{\mathcal{G}''^2} - (\bar{\mathcal{G}} - \bar{\mathcal{G}})^2 \end{aligned} \quad (\text{E.40})$$

where $\overline{\mathcal{G}''^2}$ can also be expressed in terms of the ensemble properties as follows:

$$\overline{\mathcal{G}''^2} = \bar{\varrho} \left[\lim_{\mathcal{N} \rightarrow \infty} \frac{1}{\mathcal{N}} \sum_{p=1}^{\mathcal{N}} \frac{(\mathcal{G}(\underline{\phi}^{(p)}) - \bar{\mathcal{G}})^2}{\varrho(\underline{\phi}^{(p)})} \right] \quad (\text{E.41})$$

The higher order statistics could be obtained in a similar manner.

Given the ensemble of particle properties, it is a straightforward matter to obtain the pdf. In order calculate the pdf of a property ϕ_α (referred to as $\tilde{P}(\psi_\alpha)$), the scalar bounds of ϕ_α are first split up into intervals; the width of these intervals, $\Delta\psi_\alpha$, affects the 'smoothness' of the resulting pdf. Defining $\mathcal{N}_{\psi_\alpha}$ as the number of particles which satisfy the condition $\psi_\alpha \leq \underline{\phi}^{(p)} \leq \psi_\alpha + \Delta\psi_\alpha$,

the probability (or frequency of occurrence) of any particle falling into this interval is $\frac{\mathcal{N}_{\psi_\alpha}}{\mathcal{N}}$. This, by definition, is approximately $\tilde{P}(\psi_\alpha)\Delta\psi_\alpha$, so that,

$$\tilde{P}(\psi_\alpha) = \lim_{\substack{\Delta\psi_\alpha \rightarrow 0 \\ \mathcal{N} \rightarrow \infty}} \left(\frac{\mathcal{N}_{\psi_\alpha}}{\mathcal{N}\Delta\psi_\alpha} \right) \quad (\text{E.42})$$

In deriving equation (E.37), the relation between the density-weighted and unweighted pdf was established. Integrating this over the entire composition space ψ_β ($\beta = 1, \dots, N+1$), except for $\beta = \alpha$ yields:

$$\begin{aligned} \int \tilde{P}(\underline{\psi}) d\psi_1 \cdots d\psi_{\alpha-1} d\psi_{\alpha+1} \cdots d\psi_{N+1} &= \frac{1}{\bar{\varrho}} \int \varrho(\underline{\psi}) P(\underline{\psi}) d\psi_1 \cdots d\psi_{\alpha-1} d\psi_{\alpha+1} \cdots d\psi_{N+1} \\ \Rightarrow P(\psi_\alpha) &= \frac{\bar{\varrho} \tilde{P}(\psi_\alpha)}{\langle \varrho(\underline{\phi}) | \phi_\alpha = \psi_\alpha \rangle} \end{aligned} \quad (\text{E.43})$$

$\tilde{P}(\psi_\alpha)$ can still be obtained from equation (E.42). $\langle \varrho(\underline{\phi}) | \phi_\alpha = \psi_\alpha \rangle$ is the mean density *conditioned on the event* $\psi_\alpha \leq \phi_\alpha \leq \psi_\alpha + d\psi_\alpha$. In practice this implies that those particles falling into the region $\psi_\alpha \leq \phi_\alpha^{(p)} \leq \psi_\alpha + \Delta\psi_\alpha$ are collected, *and based on this sub-ensemble* the mean density is calculated using equation (E.37) (with $\mathcal{N}$ replaced by $\mathcal{N}_{\psi_\alpha}$).

Appendix F

The Determination of the Scalar Bounds for Tabulating the Chemistry

F.1 Introduction

The bounds for the allowable composition states are required in order to tabulate the chemistry on an 'once and for all' basis. The saving on cpu time for the Monte Carlo simulation of the pdf equation is substantial. The number of species present depends on the reduced scheme that is being employed; the requirement of mass conservation and the modelling assumption of equal diffusivity imply that the dimensionality of the problem can be reduced by three scalars. The remaining scalars that have to be specified are referred to as the independent scalars.

Initially the discussion focuses on the global hydrocarbon scheme of Jones and Lindstedt [46]. The modifications required to account for the presence of N_2 in the fuel stream is also presented. The derivation of the bounds is described in detail, since it also forms the foundation for the construction of the bounds relating to the systematically reduced scheme of Chelliah et al. [10].

F.2 Composition Bounds for the Global Hydrocarbon Scheme

The global hydrocarbon scheme [46] incorporates the following species: C_xH_y , O_2 , CO , CO_2 , H_2 , H_2O and N_2 . A sensible choice for the first independent scalar is the mixture fraction since this quantity plays a central role in characterising the composition at a point in relation to the initial condition in the fuel and air streams. Furthermore, for a two-stream mixing process, the mixture fraction is related to other *strictly conserved* scalars by:

$$n_\alpha = (1 - \xi) n_{\alpha,2} + \xi n_{\alpha,1} \quad (F.1)$$

where '1' refers to the fuel stream, '2' to the oxidant stream and n_α can denote *element* specific mole number or enthalpy ($J\ Kg^{-1}$ of mixture). The mixture fraction remains constant during the chemical reaction. For each of the element species (C, H, O and N) the mass conservation equation is now constructed. These are subsequently used to construct the bounds for the reactive species.

F.2.1 Element Conservation

Carbon

Denoting $\mathcal{N}_\alpha$ as the number of atoms (or molecules) of specie α present in the reactive mixture at any position and time, the carbon element satisfies the following conservation equation (given the species that are present in the mixture):

$$\mathcal{N}_C = x\mathcal{N}_{C_xH_y} + \mathcal{N}_{CO} + \mathcal{N}_{CO_2} \quad (F.2)$$

Dividing equation (F.2) by Avogadro's constant ($N_A \approx 6.022 \times 10^{26} \text{ Kmol}^{-1}$) and subsequently by the total mass of the mixture present at that instant leads to an equation for the specific mole number (Kmol Kg^{-1}),

$$n_C = xn_{C_xH_y} + n_{CO} + n_{CO_2} \quad (F.3)$$

In terms of the carbon element, equation (F.1) becomes $n_C = n_{C,1}\xi$, where the specific mole number of carbon atoms in the fuel stream is given by, $n_{C,1} = \frac{xY_{C_xH_y,1}}{W_{C_xH_y}}$ (and $Y_{C_xH_y,1} = 1$)[†]. Substituting the relations for n_C and $n_{C,1}$ into equation (F.3) the conservation of atomic species for carbon can be written as:

$$xn_{C_xH_y} + n_{CO} + n_{CO_2} = \frac{x\xi}{W_{C_xH_y}} \quad (F.4)$$

Nitrogen

Nitrogen is a conserved scalar and is therefore linearly related to the local value of the mixture fraction. For this specie, equation (F.1) reduces to:

$$n_N = (1 - \xi)n_{N,2} \quad (F.5)$$

The only source of nitrogen atoms is from nitrogen molecules since this element is neither generated nor consumed by the chemical reaction; consequently $n_N = 2n_{N_2}$. In the *air stream*,

$$n_{N_2,2}W_{N_2} + n_{O_2,2}W_{O_2} = 1 \quad (F.6)$$

Due to the equal diffusivity assumption, the oxygen-to-nitrogen ratio is constant and is given by:

$$\frac{n_{O_2,2}}{n_{N_2,2}} = \frac{n_{O,2}}{n_{N,2}} = \frac{n_O}{n_N} \equiv \psi \approx 0.265 \quad (F.7)$$

Using equations (F.5) to (F.7) it can be shown that:

$$n_{N_2} = \frac{(1 - \xi)}{(W_{N_2} + \psi W_{O_2})} \quad (F.8)$$

For the remainder of the elements only the final result will be given since the basic procedure has been outlined in the paragraphs above.

Hydrogen

$$\begin{aligned} n_H &= yn_{C_xH_y} + 2n_{H_2O} + 2n_{H_2} = n_{H,1}\xi = \frac{y\xi}{W_{C_xH_y}} \\ \Rightarrow yn_{C_xH_y} + 2(n_{H_2O} + n_{H_2}) &= \frac{y\xi}{W_{C_xH_y}} \end{aligned} \quad (F.9)$$

[†] $Y_{C_xH_y,1} = 1$ when the fuel stream is not diluted with other species.

Oxygen

$$\begin{aligned} n_{\text{O}} &= 2n_{\text{O}_2} + n_{\text{CO}} + 2n_{\text{CO}_2} + n_{\text{H}_2\text{O}} = n_{\text{O},2}(1 - \xi) \\ \Rightarrow 2n_{\text{O}_2} + n_{\text{CO}} + 2n_{\text{CO}_2} + n_{\text{H}_2\text{O}} &= \frac{2\mathcal{U}(1 - \xi)}{(W_{\text{N}_2} + \mathcal{U}W_{\text{O}_2})} \end{aligned} \quad (\text{F.10})$$

F.2.2 Bounds for the Independent Scalars

As pointed out in section 5.3.1.3, the choice of the independent scalars is not unique. Since one of the aims of this research work is the prediction of CO and H₂, it seems sensible to try to include these as independent scalars. The aim in constructing the scalar bounds is to produce a hierarchy of coupled equations which can scan the entire composition space of the mixture; i.e. any composition state which satisfies mass conservation and $\frac{n_{\text{C}}}{n_{\text{H}}} = \frac{x}{y}$ and $\frac{n_{\text{O}}}{n_{\text{N}}} = \mathcal{U}$ can be located somewhere within the allowable scalar domain that is constructed below. In order to quantify the bounds it is necessary to invoke conditions which may never be reached in a flame. This practice is necessary so that the most conservative concentration limits can be specified. However, at the same time, this highlights a potential weakness of the approach in that the distribution of the scalar domain can be wasteful, since some regions are never accessed.

F.2.2.1 Bounds for the Mixture Fraction

The bounds for ξ are straightforward to assign since they are constrained, by definition, to be zero in the air stream and unity in the fuel stream.

$$\xi^{\max} = 1, \quad \xi^{\min} = 0 \quad (\text{F.11})$$

F.2.2.2 Bounds for $n_{\text{C}_x\text{H}_y}$ given ξ

From equation (F.4), $n_{\text{C}_x\text{H}_y}^{\max}$ occurs when $n_{\text{CO}} = n_{\text{CO}_2} = 0$.

$$n_{\text{C}_x\text{H}_y}^{\max} = \frac{\xi}{W_{\text{C}_x\text{H}_y}} \quad (\text{F.12})$$

The minimum amount of C_xH_y is subject to oxygen availability. According to equation (F.4), for a given ξ , there are $\frac{x\xi}{W_{\text{C}_x\text{H}_y}}$ Kmol of carbon atoms present per unit mass, and from equation (F.10), there are $\frac{2\mathcal{U}(1-\xi)}{(W_{\text{N}_2} + \mathcal{U}W_{\text{O}_2})}$ Kmol of oxygen atoms per unit mass of mixture. Now, if

$$\frac{2\mathcal{U}(1 - \xi)}{(W_{\text{N}_2} + \mathcal{U}W_{\text{O}_2})} \geq \frac{x\xi}{W_{\text{C}_x\text{H}_y}} \quad (\text{F.13})$$

then $n_{\text{C}_x\text{H}_y}^{\min} = 0$. However, if inequality (F.13) is not satisfied, then there will still be $\frac{x\xi}{W_{\text{C}_x\text{H}_y}} - \frac{2\mathcal{U}(1-\xi)}{(W_{\text{N}_2} + \mathcal{U}W_{\text{O}_2})}$ carbon atoms left over. Within the constraints of the global hydrocarbon scheme, this implies that these left over carbon atoms have to be attached to hydrogen in the form of C_xH_y. In this case,

$$n_{\text{C}_x\text{H}_y}^{\min} = \frac{\xi}{W_{\text{C}_x\text{H}_y}} - \frac{2\mathcal{U}(1 - \xi)}{x(W_{\text{N}_2} + \mathcal{U}W_{\text{O}_2})} \quad (\text{F.14})$$

But if oxygen satisfies the availability condition (F.13), then the rhs of equation (F.14) would be zero or negative. Since negative concentrations are unphysical, the minimum amount of C_xH_y can be generally expressed as follows:

$$n_{C_xH_y}^{min} = \max \left\{ \frac{\xi}{W_{C_xH_y}} - \frac{2\mathcal{U}(1-\xi)}{x(W_{N_2} + \mathcal{U}W_{O_2})}, 0 \right\} \quad (F.15)$$

The oxygen availability criterion assumes a one-to-one combination of carbon and oxygen atoms in order to determine the minimum C_xH_y concentration. Clearly this leads to a more conservative estimate for the minimum amount of C_xH_y than a C:O combination of 1:2 (i.e. to form CO_2) which would require fewer carbon atoms to be combined with the available oxygen atoms.

F.2.2.3 Bounds for n_{CO} given ξ and $n_{C_xH_y}$

Equation (F.4) indicates that in the balance for the C-atom the species C_xH_y , CO and CO_2 appear. If CO is chosen as the independent scalar then CO_2 can no longer be independent too, since knowledge of ξ , $n_{C_xH_y}$ and n_{CO} is sufficient to determine n_{CO_2} . Note, however, that as long as the level of CO is not fixed, the bounds for both CO and CO_2 can be determined. Let,

$$\mathcal{L} \equiv \frac{\xi}{W_{C_xH_y}} - n_{C_xH_y} \quad (F.16)$$

where $\mathcal{L}$ is constant when ξ and $n_{C_xH_y}$ are specified. Substituting equation (F.16) into (F.4),

$$n_{CO} + n_{CO_2} = x\mathcal{L} \quad (F.17)$$

From equation (F.17), $n_{CO}^{max} = x\mathcal{L}$ when $n_{CO_2}^{min} = 0$. This implies that the oxygen atoms are attached to all the remaining carbon atoms (in the form CO). This may appear to be a pathological situation, but it serves the purpose of quantifying the maximum allowable CO concentration given the constraints. Thus,

$$n_{CO}^{max} = x\mathcal{L} \quad (F.18)$$

Equation (F.18) is *not* subject to oxygen availability; in principle there has to be enough oxygen to form $x\mathcal{L}$ Kmol of $CO^\dagger$, otherwise, given that the level of C_xH_y has been fixed, this would have to imply the presence of C-radicals in the mixture, which is not permitted by the reduced scheme. The maximum amount of CO_2 is subject to oxygen availability. If,

$$\frac{\mathcal{U}(1-\xi)}{(W_{N_2} + \mathcal{U}W_{O_2})} \geq x\mathcal{L} \quad (F.19)$$

then $n_{CO_2}^{max} = x\mathcal{L}$ and $n_{CO}^{min} = 0$. However, if the above inequality is not satisfied, then clearly $n_{CO_2}^{max} \neq x\mathcal{L}$; in this case some CO would also have to be present in order to avoid having C-atoms left over. In trying to find the bounds for n_{CO}^{min} (and hence $n_{CO_2}^{max}$) subject to oxygen availability, it is necessary to assume that $n_{O_2} = n_{H_2O} = 0$ so that all the oxygen is placed at the disposal of CO and CO_2 . So from equation (F.10) with $n_{O_2} = n_{H_2O} = 0$,

$$n_{CO}^{min} + 2n_{CO_2}^{max} = \frac{2\mathcal{U}(1-\xi)}{(W_{N_2} + \mathcal{U}W_{O_2})} \quad (F.20)$$

[†]The minimum bound on the fuel ensures that this condition is satisfied

Eliminating n_{CO} from equation (F.20) with (F.17) gives,

$$n_{\text{CO}_2}^{\text{max}} = \frac{2\mathcal{U}(1 - \xi)}{(W_{\text{N}_2} + \mathcal{U}W_{\text{O}_2})} - x\mathcal{L} \quad (\text{F.21})$$

The oxygen availability requirement, inequality (F.19), can be re-expressed as:

$$\frac{2\mathcal{U}(1 - \xi)}{(W_{\text{N}_2} + \mathcal{U}W_{\text{O}_2})} - x\mathcal{L} \geq x\mathcal{L} \quad (\text{F.22})$$

Although equation (F.21) expresses the maximum amount of CO_2 when there is insufficient O_2 , inequality (F.22) indicates that if there is enough oxygen present, expression (F.21) becomes invalid since $n_{\text{CO}_2}^{\text{max}}$ then becomes larger than the *maximum allowable* amount of CO_2 ($= x\mathcal{L}$). In order to express the level of $n_{\text{CO}_2}^{\text{max}}$ in a general manner, the following can be used,

$$n_{\text{CO}_2}^{\text{max}} = \min \left\{ \frac{2\mathcal{U}(1 - \xi)}{(W_{\text{N}_2} + \mathcal{U}W_{\text{O}_2})} - x\mathcal{L} , x\mathcal{L} \right\} \quad (\text{F.23})$$

Combining equations (F.23) and (F.20) yields the bounds for the minimum amount of CO ,

$$n_{\text{CO}}^{\text{min}} = \max \left\{ 2x\mathcal{L} - \frac{2\mathcal{U}(1 - \xi)}{(W_{\text{N}_2} + \mathcal{U}W_{\text{O}_2})} , 0 \right\} \quad (\text{F.24})$$

F.2.2.4 Bounds for n_{H_2} given ξ , $n_{\text{C}_x\text{H}_y}$ and n_{CO}

In this section n_{H_2} is treated as the independent scalar. Knowledge of ξ , $n_{\text{C}_x\text{H}_y}$, n_{CO} and n_{H_2} permits the evaluation of H_2O , CO_2 and O_2 . Having specified ξ , $n_{\text{C}_x\text{H}_y}$ and n_{CO} , the amount of hydrogen atoms left to appear as H_2O and H_2 is given by equation (F.9),

$$n_{\text{H}}^* \equiv \underbrace{\left(\frac{y\xi}{W_{\text{C}_x\text{H}_y}} - yn_{\text{C}_x\text{H}_y} \right)}_{y\mathcal{L}} = 2(n_{\text{H}_2\text{O}} + n_{\text{H}_2}) \quad (\text{F.25})$$

The amount of oxygen atoms left over is given by equation (F.10),

$$n_{\text{O}}^* \equiv \frac{2\mathcal{U}(1 - \xi)}{(W_{\text{N}_2} + \mathcal{U}W_{\text{O}_2})} - n_{\text{CO}} - 2n_{\text{CO}_2} = 2n_{\text{O}_2} + n_{\text{H}_2\text{O}} \quad (\text{F.26})$$

Eliminating n_{CO_2} in equation (F.26) using (F.17) leads to,

$$\frac{2\mathcal{U}(1 - \xi)}{(W_{\text{N}_2} + \mathcal{U}W_{\text{O}_2})} + n_{\text{CO}} - 2x\mathcal{L} = 2n_{\text{O}_2} + n_{\text{H}_2\text{O}} \quad (\text{F.27})$$

The concentration of O_2 is minimised as more oxygen atoms attach themselves to hydrogen atoms in the form of H_2O . If oxygen is abundant supply then in principle it can attach itself to *all* the H-atoms so that $n_{\text{H}_2}^{\text{min}} = 0$, $n_{\text{O}_2}^{\text{min}} \geq 0$ and $n_{\text{H}_2\text{O}}$ is at a maximum. This is achieved if $n_{\text{O}}^* \geq \frac{1}{2}n_{\text{H}}^*$, since $\mathcal{N}$ hydrogen atoms require at least $\frac{\mathcal{N}}{2}$ oxygen atoms to form $\frac{\mathcal{N}}{2}$ H_2O molecules with no hydrogen left over. Substituting $n_{\text{H}_2}^{\text{min}} = 0$ into equation (F.25),

$$n_{\text{H}_2\text{O}}^{\text{max}} = \frac{y}{2}\mathcal{L} \quad (\text{F.28})$$

Using the above result in equation (F.27) yields,

$$n_{O_2}^{min} = \frac{\mathcal{U}(1-\xi)}{(W_{N_2} + \mathcal{U}W_{O_2})} + \frac{1}{2}n_{CO} - \left(x + \frac{y}{4}\right)\mathcal{L} \quad (F.29)$$

If $n_O^* \leq \frac{1}{2}n_H^*$ this means there is not enough oxygen atoms to combine with all the H_2 molecules available, so that $n_{H_2}^{min} \geq 0$, $n_{O_2}^{min} = 0$ and n_{H_2O} is at another maximum, but of a smaller magnitude to that prescribed by equation (F.28). Substituting $n_{O_2}^{min} = 0$ into equation (F.27),

$$n_{H_2O}^{max} = \frac{2\mathcal{U}(1-\xi)}{(W_{N_2} + \mathcal{U}W_{O_2})} + n_{CO} - 2x\mathcal{L} \quad (F.30)$$

This can then be used with equation (F.25) to give,

$$n_{H_2}^{min} = \left(\frac{y}{2} + 2x\right)\mathcal{L} - \frac{2\mathcal{U}(1-\xi)}{(W_{N_2} + \mathcal{U}W_{O_2})} - n_{CO} \quad (F.31)$$

Depending on the abundance of O_2 , different expressions have been determined for the bounds of $n_{H_2}^{min}$, $n_{O_2}^{min}$ and $n_{H_2O}^{max}$. Mathematically, the overriding constraints are clearly the conditions $n_O^* \leq \frac{1}{2}n_H^*$ and $n_O^* \geq \frac{1}{2}n_H^*$. Consider the latter condition with equations (F.25) and (F.26) replacing n_H^* and n_O^* , respectively:

$$\frac{2\mathcal{U}(1-\xi)}{(W_{N_2} + \mathcal{U}W_{O_2})} + n_{CO} - 2x\mathcal{L} \geq \frac{y}{2}\mathcal{L} \quad (F.32)$$

The lhs of the above inequality represents $n_{H_2O}^{max}$ (from equation (F.30)) based on limited oxygen availability, whereas the rhs is the expression for $n_{H_2O}^{max}$ when the oxygen is plentiful (equation (F.28)). The implication of this implausible result is that expression (F.30) is valid provided that its value does not exceed $\frac{y}{2}\mathcal{L}$, since at that cross-over point, a new regime of oxygen availability is operative. These remarks are embodied in the following expression:

$$n_{H_2O}^{max} = \min \left\{ \frac{2\mathcal{U}(1-\xi)}{(W_{N_2} + \mathcal{U}W_{O_2})} + n_{CO} - 2x\mathcal{L} , \frac{y}{2}\mathcal{L} \right\} \quad (F.33)$$

Re-arranging inequality (F.32) as follows:

$$\left(\frac{y}{2} + 2x\right)\mathcal{L} - \frac{2\mathcal{U}(1-\xi)}{(W_{N_2} + \mathcal{U}W_{O_2})} - n_{CO} \leq 0 \quad (F.34)$$

The rhs of equation (F.34) is the same as the rhs of equation (F.31), and the implication of this comparison is that the latter expression should not be used when the local oxygen concentration is plentiful; under those circumstances, $n_{H_2}^{min} = 0$. In summary,

$$n_{H_2}^{min} = \max \left\{ \left(\frac{y}{2} + 2x\right)\mathcal{L} - \frac{2\mathcal{U}(1-\xi)}{(W_{N_2} + \mathcal{U}W_{O_2})} - n_{CO} , 0 \right\} \quad (F.35)$$

It is clear that inequality (F.32) is consistent with the use of equation (F.29) to the extent that both require $n_{O_2}^{min} \geq 0$. However, consider when oxygen is not present in sufficient quantities, i.e. $n_O^* \leq \frac{1}{2}n_H^*$. The use of equation (F.29) then leads to $n_{O_2}^{min} \leq 0$. This is remedied by writing:

$$n_{O_2}^{min} = \max \left\{ \frac{\mathcal{U}(1-\xi)}{(W_{N_2} + \mathcal{U}W_{O_2})} + \frac{1}{2}n_{CO} - \left(x + \frac{y}{4}\right)\mathcal{L} , 0 \right\} \quad (F.36)$$

The magnitude of $n_{\text{H}_2}^{max}$ has yet to be determined. But this is a straightforward problem to resolve since n_{O_2} can be maximised by keeping all the oxygen atoms as O_2 . This implies that, $n_{\text{H}_2\text{O}}^{min} = 0$. Substituting this result into equation (F.25) yields:

$$\boxed{n_{\text{H}_2}^{max} = \frac{y}{2}\mathcal{L}} \quad (\text{F.37})$$

F.2.3 Determination of the Dependent Scalar Concentrations

Having established the bounds for all of the independent scalars, it remains to specify the concentration of the dependent scalars. At this stage the value of the following quantities are known: ξ , $n_{\text{C}_x\text{H}_y}$, n_{CO} and n_{H_2} . From equation (F.17),

$$n_{\text{CO}_2} = x\mathcal{L} - n_{\text{CO}} \quad (\text{F.38})$$

From equation (F.25),

$$n_{\text{H}_2\text{O}} = \frac{y}{2}\mathcal{L} - n_{\text{H}_2} \quad (\text{F.39})$$

Finally, from equations (F.27) and (F.39),

$$n_{\text{O}_2} = \frac{\mathcal{U}(1-\xi)}{(W_{\text{N}_2} + \mathcal{U}W_{\text{O}_2})} + \frac{1}{2}n_{\text{CO}} - \left(x + \frac{y}{4}\right)\mathcal{L} + \frac{1}{2}n_{\text{H}_2} \quad (\text{F.40})$$

F.2.4 Modifications for Flames with Nitrogen-Diluted Fuel Streams

In this section the modifications that are required to account for the N_2 -dilution of the fuel stream are described. The effect manifest itself in the equations expressing the conservation of elemental species. The subsequent analysis to obtain the bounds for the independent scalars is the same as in section F.2.2 and will not be presented.

Due to the presence of N_2 in the fuel stream, the oxygen-to-nitrogen ratio is no longer equal to $\mathcal{U}$. However, this does *not* mean that the concept of equal diffusivity has to be relaxed. Equation (F.1) for the oxygen and nitrogen elements can be expressed as:

$$n_{\text{O}} = (1-\xi)n_{\text{O},2} \quad (\text{F.41})$$

$$n_{\text{N}} = (1-\xi)n_{\text{N},2} + \xi n_{\text{N},1} \quad (\text{F.42})$$

where subscript '2' refers to the air stream so that $\frac{n_{\text{O},2}}{n_{\text{N},2}} = \mathcal{U}$ and,

$$n_{\text{N},1} = 2 \frac{Y_{\text{N}_2,1}}{W_{\text{N}_2}} = \frac{2}{W_{\text{N}_2}} \frac{\dot{m}_{\text{N}_2,1}}{\dot{m}_{\text{fuel},1} + \dot{m}_{\text{N}_2,1}} \quad (\text{F.43})$$

where the mass flow rates are given in table 6.3. Dividing equation (F.41) by (F.42) leads to:

$$\frac{n_{\text{O}}}{n_{\text{N}}} = \frac{(1-\xi)n_{\text{O},2}}{(1-\xi)n_{\text{N},2} + \xi n_{\text{N},1}} = \frac{(1-\xi)}{\frac{(1-\xi)}{\mathcal{U}} + \frac{n_{\text{N},1}}{n_{\text{O},2}}\xi} \quad (\text{F.44})$$

The above expression clearly demonstrates that $\frac{n_{\text{O}}}{n_{\text{N}}}$ is no longer fixed to the value of $\mathcal{U}$. This means that the implication of $\frac{n_{\text{O}}}{n_{\text{N}}}$ being constant is only true when the fuel and air travel in separate initial streams.

The effect of these changes on the conservation of elemental species will now be presented.

Carbon

Equation (F.3) is still valid under these circumstances.

$$n_C = x n_{C_x H_y} + n_{CO} + n_{CO_2} = n_{C,1} \xi \quad (F.45)$$

where the specific mole number of the carbon atoms in the fuel stream is now given by

$$n_{C,1} = \frac{x Y_{C_x H_y,1}}{W_{C_x H_y}} = \frac{x}{W_{C_x H_y}} \left(\frac{\dot{m}_{fuel,1}}{\dot{m}_{fuel,1} + \dot{m}_{N_2,1}} \right) \quad (F.46)$$

Therefore equation (F.46) indicates that $Y_{C_x H_y,1} \neq 1$ unless $\dot{m}_{N_2,1} = 0$. Substituting the relation for $n_{C,1}$ into equation (F.45) yields:

$$x n_{C_x H_y} + n_{CO} + n_{CO_2} = \frac{x \xi}{W_{C_x H_y}} \left(\frac{\dot{m}_{fuel,1}}{\dot{m}_{fuel,1} + \dot{m}_{N_2,1}} \right) \quad (F.47)$$

Nitrogen

Dividing equation (F.42) by two yields the following relation involving nitrogen *molecules*,

$$n_{N_2} = (1 - \xi) n_{N_2,2} + \xi n_{N_2,1} \quad (F.48)$$

$n_{N_2,1}$ can be obtained using (F.43) and $n_{N_2,2}$ is essentially the result of equation (F.8) evaluated at $\xi = 0$. Thus,

$$n_{N_2} = \frac{(1 - \xi)}{(W_{N_2} + U W_{O_2})} + \left(\frac{\dot{m}_{N_2,1}}{\dot{m}_{fuel,1} + \dot{m}_{N_2,1}} \right) \frac{\xi}{W_{N_2}} \quad (F.49)$$

Hydrogen

Using the results described above, leading to equations (F.9) and (F.47), the conservation equation for the specific mole number of hydrogen atoms can be written down by inspection:

$$y n_{C_x H_y} + 2 (n_{H_2O} + n_{H_2}) = \frac{y \xi}{W_{C_x H_y}} \left(\frac{\dot{m}_{fuel,1}}{\dot{m}_{fuel,1} + \dot{m}_{N_2,1}} \right) \quad (F.50)$$

Oxygen

The oxygen balance equation is unaffected by the fact that the fuel stream is now diluted with nitrogen. Therefore equation (F.10) is still valid.

It is quite clear that the effect of the nitrogen dilution on the conservation equations for the elemental species is quite simple. The manner in which the bounds are subsequently constructed follows *exactly* the same pattern as the one described in section F.2.2; the analytical form for the bounds are very similar too, affected slightly by the quotient involving the mass flow rates of fuel and N_2 .

F.3 Composition Bounds for the Systematically Reduced Methane/Air Scheme

F.3.1 Conservation of Atomic Species

The species involved in this mechanism are: $C_x H_y$, O_2 , CO , CO_2 , H_2 , H_2O , CH_3 , H and N_2 . The results for the conservation of atomic species only affect carbon and hydrogen; the relations for

oxygen atoms and nitrogen molecules (equations (F.10) and (F.8) are still valid:

$$n_{\text{CH}_4} + n_{\text{CO}} + n_{\text{CO}_2} + n_{\text{CH}_3} = \frac{\xi}{W_{\text{CH}_4}} \quad (\text{F.51})$$

$$4n_{\text{CH}_4} + 2n_{\text{H}_2\text{O}} + 2n_{\text{H}_2} + 3n_{\text{CH}_3} + n_{\text{H}} = \frac{4\xi}{W_{\text{CH}_4}} \quad (\text{F.52})$$

F.3.2 Bounds for the Independent Scalars

The bounds for the mixture fraction are the same as those specified in equation (F.11). There is also no change for the upper bound of methane, as given by equation (F.12) for the case where $x = 1$ and $y = 4$. The lower bound deserves further mention since it highlights an important difference between the way the bounds are generated for the global scheme and for the current one. If the oxygen level is abundant, then as before, the lower bound for the hydrocarbon fuel will be zero. However, if the oxygen supply is limited, then in principle $(\frac{\xi}{W_{\text{CH}_4}} - \frac{2U(1-\xi)}{(W_{\text{N}_2} + UW_{\text{O}_2})}) \text{ Kmol Kg}^{-1}$ of carbon atoms are left over. Whilst with the global scheme this would have to remain as CH_4 molecules, the current scheme allows for the presence of CH_3 and H . Therefore in theory, the most conservative value for $n_{\text{CH}_4}^{\min}$ is zero since the surplus fuel can appear in the form of CH_3 and H . The problem with such a specification is that for rich mixtures unrealistically high levels of radical species would be present. For lean mixtures, the proportion of radical to non-radical species could also be unrealistic. The validity of tabulating such pathological composition states comes under question. The solution of the rate equations is also rendered more difficult as suggested by the method of obtaining the mixture temperature (described in appendix H); LSODE also indicates that the set of ODE's may be more 'stiff' too, since in certain cases converged solutions could not be obtained. It was found that the simplest approach would be to use the same bounds as specified in equation (F.15), implying that no decomposition of CH_4 into CH_3 and H is permitted when the oxygen is not in an abundant supply.

The bounds for CH_3 are now discussed. The fact that the upper bound for CH_4 is $\frac{\xi}{W_{\text{CH}_4}}$ implies that $n_{\text{CH}_3}^{\min} = 0$. At this stage it is assumed that ξ and n_{CH_4} have been specified, so that concentration of carbon atoms available is:

$$\mathcal{L}^a \equiv \frac{\xi}{W_{\text{CH}_4}} - n_{\text{CH}_4} \quad (\text{F.53})$$

The following arbitrary and ad-hoc constraint is imposed for the upper bound of CH_3 ,

$$\boxed{\text{if } \mathcal{L}^a \geq n_{\text{CH}_3}^{\lim}, \text{ then } n_{\text{CH}_3}^{\max} = n_{\text{CH}_3}^{\lim} \text{ otherwise } n_{\text{CH}_3}^{\max} = \mathcal{L}^a} \quad (\text{F.54})$$

The quantity $n_{\text{CH}_3}^{\lim}$ is a constant - the concentration of CH_3 is effectively clipped so that it is never allowed to go above the value of $n_{\text{CH}_3}^{\lim}$. A similar quantity is defined for the H -radical discussed further on. For the time being it is pointed that these clipped values are chosen by performing laminar flame simulations. The results and discussion (section 6) indicate that the predicted levels of H and CH_3 in the laminar flame calculations are, in the best cases, an order of magnitude above those obtained with differential diffusion effects included. It therefore seems plausible to include these values as the very maximum that can be allowed for the construction of the table.

Introducing the following definition,

$$\mathcal{L}^b = \frac{\xi}{W_{\text{CH}_4}} - n_{\text{CH}_4} - n_{\text{CH}_3} \quad (\text{F.55})$$

it can be shown using the same ideas as those invoked in section F.2.2.3 that the bounds for CO are the same as those specified in equations (F.18) and (F.24), with $\mathcal{L}$ replaced by $\mathcal{L}^b$.

The bounds for the H-radical are generated along much the same lines as for CH_3 . The minimum amount is necessarily zero, i.e. $n_{\text{H}}^{\min} = 0$. The available 'pool' of hydrogen atoms, $n_{\text{H}}^{\#}$, is given by: $n_{\text{H}}^{\#} \equiv 4\mathcal{L}^a - 3n_{\text{CH}_3}$. Consequently $n_{\text{H}}^{\min}$ is forced to satisfy,

$$\boxed{\text{if } n_{\text{H}}^{\#} \geq n_{\text{H}}^{\lim}, \text{ then } n_{\text{H}}^{\max} = n_{\text{H}}^{\lim} \text{ otherwise } n_{\text{H}}^{\max} = n_{\text{H}}^{\#}} \quad (\text{F.56})$$

At this stage ξ , n_{CH_4} , n_{CH_3} , n_{CO} and n_{H} have been specified, leaving n_{H_2} . The amounts of oxygen and hydrogen left over are given by,

$$n_{\text{H}}^* = 2n_{\text{H}_2\text{O}} + 2n_{\text{H}_2} = 4\mathcal{L}^a - 3n_{\text{CH}_3} - n_{\text{H}} \quad (\text{F.57})$$

$$n_{\text{O}}^* = 2n_{\text{O}_2} + n_{\text{H}_2\text{O}} = \frac{2\mathcal{U}(1 - \xi)}{(W_{\text{N}_2} + \mathcal{U}W_{\text{O}_2})} - n_{\text{CO}} - 2n_{\text{CO}_2} \quad (\text{F.58})$$

The manner in which the lower bound for H_2 is determined centres around the degree of oxygen availability. If $n_{\text{H}}^* \leq 2n_{\text{O}}^*$ then all the hydrogen atoms can attach themselves to oxygen atoms so that $n_{\text{H}_2} = 0$. However, if $n_{\text{H}}^* \geq 2n_{\text{O}}^*$ then $n_{\text{O}_2} = 0$. Using these relations, the following expression for $n_{\text{H}_2}^{\min}$ can be obtained:

$$\boxed{n_{\text{H}_2}^{\min} = \max \left\{ \frac{n_{\text{H}}^*}{2} - n_{\text{O}}^*, 0 \right\}} \quad (\text{F.59})$$

To maximise H_2 , $n_{\text{H}_2\text{O}} = 0$ so that (from equation (F.57)),

$$\boxed{n_{\text{H}_2}^{\max} = \frac{1}{2}n_{\text{H}}^* = 2\mathcal{L}^a - \frac{3}{2}n_{\text{CH}_3} - \frac{1}{2}n_{\text{H}}} \quad (\text{F.60})$$

Having established the bounds, it remains to specify n_{CO_2} , $n_{\text{H}_2\text{O}}$ and n_{O_2} in terms of the independent scalars. Substituting equation (F.55) into (F.51) gives,

$$n_{\text{CO}_2} = \mathcal{L}^b - n_{\text{CO}} \quad (\text{F.61})$$

From equation (F.52) the concentration of H_2O can be obtained,

$$n_{\text{H}_2\text{O}} = \frac{1}{2} \left(\frac{4\xi}{W_{\text{CH}_4}} - 4n_{\text{CH}_4} - 3n_{\text{CH}_3} - 2n_{\text{H}_2} - n_{\text{H}} \right) \quad (\text{F.62})$$

and finally, since the oxygen atom balance relation is unaffected *in form* by the presence of the radical species, equation (F.10) can be used to write,

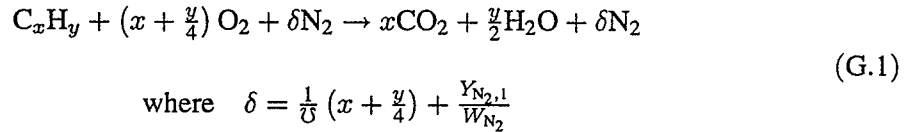
$$n_{\text{O}_2} = \frac{1}{2} \left[\frac{2\mathcal{U}(1 - \xi)}{(W_{\text{N}_2} + \mathcal{U}W_{\text{O}_2})} - n_{\text{CO}} - 2n_{\text{CO}_2} - n_{\text{H}_2\text{O}} \right] \quad (\text{F.63})$$

Appendix G

The Stoichiometric Mixture Fraction of a Hydrocarbon

In this appendix the derivation of equation (6.18) is presented. This result also reduces to equation (6.8) for the case when there is no dilution of the fuel stream by nitrogen. So for the purposes of the current derivation it is assumed that the fuel stream contains an amount $n_{N_2,1}$ of nitrogen[†], and from mass conservation it follows that: $n_{C_xH_y,1} = (1 - Y_{N_2,1})W_{C_xH_y}^{-1}$.

The stoichiometric balance equation for the burnt-out of the hydrocarbon is,



Analytical relations can now be constructed based on the following requirements:

- For a given initial value of $\frac{n_C}{n_H}$ and $\frac{n_O}{n_N}$, chemical reaction does not alter these ratios. This is a physical requirement, not an assumption.
- Overall mass conservation, i.e. the mass fractions sum to unity.

Since the carbon-to-hydrogen ratio remains fixed, it is possible to write,

$$\frac{n_C}{n_H} = \frac{x}{y} = \frac{n_{CO_2}}{2n_{H_2O}} \quad (G.2)$$

For the oxygen-to-nitrogen ratio it is important to specify amounts which imply that fuel and oxygen are present in stoichiometric proportions. Therefore if there are $n_{C_xH_y,1}$ Kmol of fuel per Kg of mixture, there also has to be $(x + \frac{y}{4})n_{C_xH_y,1}$ Kmol of O_2 (per Kg of mixture) present for complete burnt-out.

$$\frac{n_O}{n_N} = \frac{2(x + \frac{y}{4})n_{C_xH_y,1}}{2[\frac{1}{O}(x + \frac{y}{4})n_{C_xH_y,1} + \frac{Y_{N_2,1}}{W_{N_2}}]} = \frac{n_{H_2O} + 2n_{CO_2}}{2n_{N_2}} \quad (G.3)$$

Substituting equation (G.2) into (G.3) to eliminate n_{H_2O} yields,

$$\frac{n_{CO_2}}{n_{N_2}} = xn_{C_xH_y,1} \left[\frac{1}{O} \left(x + \frac{y}{4} \right) n_{C_xH_y,1} + \frac{Y_{N_2,1}}{W_{N_2}} \right]^{-1} \quad (G.4)$$

[†]In the current notation, stream '1' is assumed to contain fuel and nitrogen so that $n_{N_2,1}$ can be quantified using equation (F.43). Subscript '2' denotes the air stream where the oxygen-to-nitrogen ratio, $\frac{n_{O_2,2}}{n_{N_2,2}} \equiv \psi \approx 0.265$.

Defining the mixture fraction in terms of the carbon element,

$$\xi \equiv \frac{n_C}{n_{C,1}} \quad (G.5)$$

Assuming complete combustion, $n_C = n_{CO_2}$ (i.e. $n_{C_xH_y} = 0$ since it is all consumed for the case when $\xi = \xi_{st}$). So equation (G.5) becomes,

$$\xi_{st} = \frac{n_{CO_2}}{xn_{C_xH_y,1}} \quad (G.6)$$

The requirement for overall mass conservation can be expressed as:

$$n_{CO_2}W_{CO_2} + n_{H_2O}W_{H_2O} + n_{N_2}W_{N_2} = 1 \quad (G.7)$$

Note that O_2 and C_xH_y do not appear in equation (G.7) since they have been completely consumed. Substituting equations (G.2,G.4,G.6) into (G.7) leads to,

$$\xi_{st} = \frac{1}{xn_{C_xH_y,1}W_{CO_2} + \frac{y}{2}n_{C_xH_y,1}W_{H_2O} + \left[\frac{1}{U}\left(x + \frac{y}{4}\right)n_{C_xH_y,1} + \frac{Y_{N_2,1}}{W_{N_2}}\right]W_{N_2}} \quad (G.8)$$

The first two terms in the denominator of equation (G.8) can be manipulated as follows:

$$\begin{aligned} xn_{C_xH_y,1}W_{CO_2} + \frac{y}{2}n_{C_xH_y,1}W_{H_2O} &= n_{C_xH_y,1}(xW_C + xW_{O_2} + yW_H + \frac{y}{4}W_{O_2}) \\ &= n_{C_xH_y,1}W_{C_xH_y} + \left(x + \frac{y}{4}\right)n_{C_xH_y,1}W_{O_2} \end{aligned} \quad (G.9)$$

Introducing equation (G.9) into (G.8) leads to the following expression for the stoichiometric mixture fraction,

$$\xi_{st} = \frac{1}{\left(W_{O_2} + \frac{W_{N_2}}{U}\right)\left(x + \frac{y}{4}\right)n_{C_xH_y,1} + \underbrace{Y_{N_2,1} + Y_{C_xH_y,1}}_{=1}} \quad (G.10)$$

When there is no dilution of the fuel stream, $Y_{N_2,1} = 0$, $Y_{C_xH_y,1} = 1$ and $n_{C_xH_y,1} = W_{C_xH_y}^{-1}$; this gives rise to equation (6.8). In addition, using equation (F.43)

$$n_{C_xH_y,1} = \frac{(1 - Y_{N_2,1})}{W_{C_xH_y}} = \frac{1}{W_{C_xH_y}} \left(1 - \frac{\dot{m}_{N_2,1}}{\dot{m}_{fuel,1} + \dot{m}_{N_2,1}}\right) \quad (G.11)$$

Substitution of equation (G.11) into (G.10) leads to (6.18).

Appendix H

Details of the Thermochemistry

H.1 Introduction

In this appendix the following areas of the calculation procedure are dealt with:

- The determination of the mixture temperature. The temperature is central to the prediction of flames, particularly since the reaction rates are such a strongly non-linear function of this quantity.
- For a generalised reversible reaction, the derivation of the expression for the equilibrium constant, $K_c(T)$, is presented.
- By way of example, the method of obtaining the reverse rate in reaction step (iii), equation (5.15), is discussed. In addition, due to analytical form of the reaction rate expressions, it is shown how these are 'clipped' in order to avoid singular behaviour as the concentration of the some of the species tend to zero.

H.2 Determination of the Mixture Temperature

The enthalpy per unit mass of mixture, h (in J/Kg), is the primary variable required to calculate the mixture temperature. It is related to the composition via,

$$h = \sum_{\alpha=1}^N Y_{\alpha} h_{\alpha} \quad (\text{H.1})$$

This expression is also used in equation (2.8). N denotes the number of species present in the reactive mixture. For the global hydrocarbon scheme $N = 7$ whilst for the systematically reduced mechanism $N = 9$. h_{α} is the enthalpy of specie α *per unit mass of specie α* . The polynomial functions presented in the work of Gordon and McBride [32] are used for evaluating H_{α}° , defined as the enthalpy of specie α *per unit Kmol of specie α* [†],

$$H_{\alpha}^{\circ} = \left(c_{\alpha,1}T + \frac{c_{\alpha,2}}{2}T^2 + \frac{c_{\alpha,3}}{3}T^3 + \frac{c_{\alpha,4}}{4}T^4 + \frac{c_{\alpha,5}}{5}T^5 + c_{\alpha,6} \right) R$$

[†]The superscript 'o' indicates the *standard state*. Different definitions exist for this; for example the state of the gas at one atmosphere (unit pressure) or the state of an ideal gas at unit fugacity as used in [32]. However, for an ideal gas the enthalpy is only a function of the temperature, so that the magnitude of the pressure is of no consequence.

$$= \left(\sum_{f=1}^5 \frac{c_{\alpha,f}}{f} T^f + c_{\alpha,6} \right) R \quad (\text{H.2})$$

The basis for the calculation of the temperature relies on the fact that the value of h is unaffected by chemical reaction. This relationship does not generally hold for the enthalpy per unit Kmol of the mixture. Therefore the conversion from H_{α}° (J/Kmol) to h_{α} (J/Kg) is necessary, and is achieved through the relation $H_{\alpha}^{\circ} = h_{\alpha} W_{\alpha}$; substituting this into equation (H.1),

$$\begin{aligned} h &= \sum_{\alpha=1}^N n_{\alpha} H_{\alpha}^{\circ} \\ &= \left(\sum_{\alpha=1}^N \sum_{f=1}^5 n_{\alpha} \frac{c_{\alpha,f}}{f} T^f + \sum_{\alpha=1}^N n_{\alpha} c_{\alpha,6} \right) R \end{aligned} \quad (\text{H.3})$$

Equation (H.3) needs to be solved for T ; the equation is clearly non-linear in T and a Newton-Raphson technique is used to solve this equation. This procedure worked very well for the global hydrocarbon scheme, i.e. convergence to a sufficiently accurate solution was obtained in up to three iterations. For the systematically reduced scheme it was found that in composition states having uncharacteristically high levels of CH_3 and/or H in relation to the other species (such as CO and H_2), the procedure could converge to temperatures below 300K (or even above 2500K). One possible reason for this is that the initial specification of h through an *unburnt* fuel/air mixture is not suitable for specifying the enthalpy of a mixture which evolves to such a pathological composition state. This partly motivated the need for clipping the bounds for the radical concentrations (described in section F.3), in order to reduce the number of occurrences of such pathological states.

H.3 Determination of the Equilibrium Constant

In this section, most of the results that are quoted (without derivation) relate to the basic principles of chemical thermodynamics. The works of Gordon and McBride [32] and Everdell [26] were used as references.

Many different equilibrium constants can be defined, however, $K_c(T)$ appears to be the most useful since it is evaluated in terms of species concentrations and can therefore be used more easily in the context of reaction rate kinetics. Unless stated otherwise, the following S.I. units are employed: Kg, m, s, Kelvins, J and Kmol. The gas constant consistent with these units is $R = 8314.3 \text{ J Kmol}^{-1} \text{K}^{-1}$. In order to calculate $K_c(T)$ a few basic concepts need to be introduced. The Gibbs free energy, g (in J/Kg), is defined as:

$$g \equiv h - Ts \quad (\text{H.4})$$

where s is the entropy per unit mass of mixture ($\text{J Kg}^{-1} \text{K}^{-1}$). The chemical potential of specie α , $\vartheta_{\alpha}^{\dagger}$ (in J/Kmol), can be defined in terms of the Gibbs free energy,

$$\vartheta_{\alpha} \equiv \left(\frac{\partial g}{\partial n_{\alpha}} \right)_{p,T,n_1,\dots,n_{\alpha-1},n_{\alpha+1},\dots,n_N} \quad (\text{H.5})$$

[†]It is conventional to use the symbol μ to denote the chemical potential, but in this thesis μ is already being used for the dynamic viscosity.

For an ideal gas it can be shown [26] that,

$$\vartheta_{\alpha}(T, p) = \vartheta_{\alpha}^{\circ}(T) + RT \ln p_{\alpha} \quad (\text{H.6})$$

where $\vartheta_{\alpha}^{\circ}(T)$ is the *standard* chemical potential of the component as a pure gas at *one atmosphere*^{††} (unit pressure) and at temperature T - the definition constrains $\vartheta_{\alpha}^{\circ}$ to be independent of pressure and only a function of temperature. p_{α} is the partial pressure exerted by specie α . For an ideal gas, Dalton's law of partial pressures implies that $\sum_{\alpha=1}^N p_{\alpha} = p$ and $p_{\alpha} = X_{\alpha}p$, where X_{α} ($= n_{\alpha}(\sum_{\beta} n_{\beta})^{-1}$) is the mole fraction of specie α . Substituting this into equation (H.6) gives,

$$\vartheta_{\alpha}(T, p) = \underbrace{\vartheta_{\alpha}^{\circ}(T) + RT \ln p}_{\equiv \vartheta_{\alpha}^{*}(T, p)} + RT \ln X_{\alpha} \quad (\text{H.7})$$

Consider the following reaction,



At *equilibrium* the sums of the chemical potentials of the reacting species on both sides of the chemical equation are equal [26],

$$a\vartheta_{A,eq} + b\vartheta_{B,eq} + \dots = c\vartheta_{C,eq} + d\vartheta_{D,eq} + \dots \quad (\text{H.9})$$

where the subscript 'eq' indicates the condition at chemical equilibrium. Such a qualification is not required for the *standard* chemical potential since it is only a function of the temperature. Substituting equation (H.6) into (H.9) yields:

$$\begin{aligned} a\vartheta_A^{\circ} + aRT \ln p_{A,eq} + b\vartheta_B^{\circ} + bRT \ln p_{B,eq} + \dots &= c\vartheta_C^{\circ} + cRT \ln p_{C,eq} + \dots \\ \Rightarrow a\vartheta_A^{\circ} + b\vartheta_B^{\circ} + \dots - c\vartheta_C^{\circ} - d\vartheta_D^{\circ} &= RT \ln \left(\frac{p_{C,eq}^c p_{D,eq}^d \dots}{p_{A,eq}^a p_{B,eq}^b \dots} \right) \\ \Rightarrow \frac{p_{C,eq}^c p_{D,eq}^d \dots}{p_{A,eq}^a p_{B,eq}^b \dots} &= \exp \left(\sum_{\alpha=1}^N \nu'_{\alpha} \frac{\vartheta_{\alpha}^{\circ}}{RT} - \sum_{\alpha=1}^N \nu''_{\alpha} \frac{\vartheta_{\alpha}^{\circ}}{RT} \right) \equiv K_p(T) \end{aligned} \quad (\text{H.10})$$

In equation (H.10), ν'_{α} and ν''_{α} are the stoichiometric coefficients of specie α on the left and right sides (respectively) of the chemical equation (H.8). $K_p(T)$ is an equilibrium constant in terms of partial pressures. Since the various $\vartheta_{\alpha}^{\circ}$ are independent of pressure, the same is true for $K_p(T)$. Equation (H.10) is one form of the *Law of Mass Action*. Expressing this law in terms of mole fractions, i.e. in terms of equation (H.7),

$$K_X(T, p) \equiv \frac{\prod_{\alpha=1}^N X_{\alpha,eq}^{\nu''_{\alpha}}}{\prod_{\alpha=1}^N X_{\alpha,eq}^{\nu'_{\alpha}}} \quad (\text{H.11})$$

$$= p^{(\sum_{\alpha=1}^N \nu'_{\alpha} - \sum_{\alpha=1}^N \nu''_{\alpha})} K_p(T) \quad (\text{H.12})$$

The second relation follows from $p_{\alpha} = X_{\alpha}p$ where the unit of pressure is atmospheres; equation (H.11) indicates that $K_X = K_p$ if the system is at unit pressure, and according to (H.12) the

^{††}Note that in equation (H.6), the units of the pressure are in atmospheres, not in Pascals.

equality still holds if there is no change in the overall mole number (i.e. $\sum_{\alpha=1}^N \nu'_\alpha = \sum_{\alpha=1}^N \nu''_\alpha$). From the definition of the mole fraction the following can be written,

$$\prod_{\alpha=1}^N X_{\alpha,eq}^{\nu'_\alpha} = \prod_{\alpha=1}^N \frac{n_{\alpha,eq}^{\nu'_\alpha}}{\left(\sum_{\beta=1}^N n_{\beta,eq}\right)^{\nu'_\alpha}} = \frac{\prod_{\alpha=1}^N n_{\alpha,eq}^{\nu'_\alpha}}{\left(\sum_{\beta=1}^N n_{\beta,eq}\right)^{\sum_{\alpha=1}^N \nu'_\alpha}} \quad (\text{H.13})$$

Substituting equation (H.13) (for $\nu'_\alpha = \nu'_\alpha$ and $\nu''_\alpha = \nu''_\alpha$) into (H.11) leads to:

$$K_X(T, p) = \left(\sum_{\beta=1}^N n_{\beta,eq}\right)^{\left(\sum_{\alpha=1}^N \nu'_\alpha - \sum_{\alpha=1}^N \nu''_\alpha\right)} \frac{\prod_{\alpha=1}^N n_{\alpha,eq}^{\nu''_\alpha}}{\prod_{\alpha=1}^N n_{\alpha,eq}^{\nu'_\alpha}} \quad (\text{H.14})$$

The specific mole number at equilibrium, $n_{\alpha,eq}$ (Kmol/Kg), is related to the molar concentration, $C_{\alpha,eq}$ (Kmol/m³), through the mixture density, viz: $C_{\alpha,eq} = \rho n_{\alpha,eq}$. Substituting this into equation (H.14):

$$K_X(T, p) = \left(\rho \sum_{\beta=1}^N n_{\beta,eq}\right)^{\left(\sum_{\alpha=1}^N \nu'_\alpha - \sum_{\alpha=1}^N \nu''_\alpha\right)} \frac{\prod_{\alpha=1}^N C_{\alpha,eq}^{\nu''_\alpha}}{\underbrace{\prod_{\alpha=1}^N C_{\alpha,eq}^{\nu'_\alpha}}_{\equiv K_c(T)}} \quad (\text{H.15})$$

The equation of state for component α in an ideal gas mixture is, $p_\alpha = \rho n_\alpha R' T$, where $R' = \frac{R}{p_o} = \frac{8314.3}{1.013 \times 10^5} \text{ atm m}^3 \text{Kmol}^{-1} \text{K}^{-1}$. This modification is required since the unit of atmospheres is being used for the pressure. Introducing the equation of state in order to eliminate $n_{\beta,eq}$ from (H.15) and summing over the partial pressures yields:

$$K_X(T, p) = K_c(T) \left(\frac{p_o p}{RT}\right)^{\left(\sum_{\alpha=1}^N \nu'_\alpha - \sum_{\alpha=1}^N \nu''_\alpha\right)} \quad (\text{H.16})$$

Eliminating $K_X(T, p)$ from equation (H.16) using (H.12),

$$K_c(T) = K_p(T) \left(\frac{RT}{p_o p}\right)^{\left(\sum_{\alpha=1}^N \nu'_\alpha - \sum_{\alpha=1}^N \nu''_\alpha\right)} p^{\left(\sum_{\alpha=1}^N \nu'_\alpha - \sum_{\alpha=1}^N \nu''_\alpha\right)} \quad (\text{H.17})$$

$K_p(T)$ is given by equation (H.10) so that,

$$\begin{aligned} K_c(T) &= \left(\frac{RT}{p_o}\right)^{\left(\sum_{\alpha=1}^N \nu'_\alpha - \sum_{\alpha=1}^N \nu''_\alpha\right)} \exp\left(\sum_{\alpha=1}^N \nu'_\alpha \frac{\vartheta_\alpha^{\circ'}}{RT} - \sum_{\alpha=1}^N \nu''_\alpha \frac{\vartheta_\alpha^{\circ''}}{RT}\right) \\ &= \exp\left[\left(\sum_{\alpha=1}^N \nu'_\alpha - \sum_{\alpha=1}^N \nu''_\alpha\right) \ln\left(\frac{RT}{p_o}\right)\right] \exp\left(\sum_{\alpha=1}^N \nu'_\alpha \frac{\vartheta_\alpha^{\circ'}}{RT} - \sum_{\alpha=1}^N \nu''_\alpha \frac{\vartheta_\alpha^{\circ''}}{RT}\right) \end{aligned} \quad (\text{H.18})$$

p_o is a reference pressure and therefore a constant. This means that K_c is a function of the temperature alone. Its final form is expressed as:

$$K_c(T) = \exp\left[\sum_{\alpha=1}^N \nu'_\alpha \frac{\vartheta_\alpha^{\circ'}}{RT} - \sum_{\alpha=1}^N \nu''_\alpha \frac{\vartheta_\alpha^{\circ''}}{RT} + \left(\sum_{\alpha=1}^N \nu'_\alpha - \sum_{\alpha=1}^N \nu''_\alpha\right) \ln\left(\frac{RT}{p_o}\right)\right] \quad (\text{H.19})$$

Polynomial expressions (obtained from the JANAF and NASA Lewis databases) are employed to evaluate the enthalpy and entropy of the species as a function of temperature. The procedure follows the work of Gordon and McBride [32], and in section H.2 the polynomial expression for the enthalpy is written down (equation (H.2)). The form of the expressions for the entropy is,

$$\frac{s_{\alpha}^{\circ}}{R} = c_{\alpha,1} \ln T + c_{\alpha,2} T + \frac{c_{\alpha,3}}{2} T^2 + \frac{c_{\alpha,4}}{3} T^3 + \frac{c_{\alpha,5}}{4} T^4 + c_{\alpha,7} \quad (\text{H.20})$$

The entropy of the mixture, s ($\text{J K g}^{-1} \text{K}^{-1}$), is given by: $s = \sum_{\alpha=1}^N n_{\alpha} s_{\alpha}$. From the definition of the Gibbs free energy and the chemical potential it can be shown [26] that,

$$g \equiv h - Ts = \sum_{\alpha=1}^N \vartheta_{\alpha} n_{\alpha} \quad (\text{H.21})$$

and in addition the following identity can be derived:

$$-\left(\frac{\partial s}{\partial n_{\alpha}}\right)_{p,T,n_1,\dots,n_{\alpha-1},n_{\alpha+1},\dots,n_N} = \left(\frac{\partial \vartheta_{\alpha}}{\partial T}\right)_{p,n_1,\dots,n_{\alpha-1},n_{\alpha},n_{\alpha+1},\dots,n_N} \quad (\text{H.22})$$

Applying equation (H.22) to (H.7) yields,

$$s_{\alpha} = s_{\alpha}^{\circ} - R \ln X_{\alpha} - R \ln p \quad (\text{H.23})$$

In obtaining equation (H.3) it is shown that $h = \sum_{\alpha=1}^N n_{\alpha} H_{\alpha}^{\circ}$. Substituting this, together with equations (H.7) and (H.23) into (H.21) yields an expression for the standard chemical potential of species α ,

$$\boxed{\frac{\vartheta_{\alpha}^{\circ}}{RT} = \frac{H_{\alpha}^{\circ}}{RT} - \frac{s_{\alpha}^{\circ}}{R}} \quad (\text{H.24})$$

H.4 Example: Determination of a Reverse Rate Using the Global Scheme

Consider reaction step (iii) of equation (5.15),



From equation (H.19) it is clear that,

$$K_c^{(iii)}(T) = \exp \left[\frac{1}{RT} \left(\vartheta_{\text{H}_2}^{\circ} + \frac{1}{2} \vartheta_{\text{O}_2}^{\circ} - \vartheta_{\text{H}_2\text{O}}^{\circ} \right) + \frac{1}{2} \ln \left(\frac{RT}{p_o} \right) \right] \quad (\text{H.26})$$

Equation (5.16) indicates that,

$$\dot{\omega}_{f,(iii)} = k_{f,(iii)} [\text{H}_2]^{\frac{1}{2}} [\text{O}_2]^{\frac{9}{4}} [\text{H}_2\text{O}]^{-1}$$

and using the data in table 5.1, the *forward* rate constant (for C_3H_8 , as an example) is given by:

$$k_{f,(iii)} = 3 \times 10^{16} \frac{1}{T} \exp \left(-\frac{40,000}{R^*T} \right) \quad (\text{H.27})$$

The use of Kilocalories for the activation energy (as indicated in table 5.1) implies that in the reaction rate expression the gas constant (R^*) has to have units of Kcal Kmol⁻¹ K⁻¹. Since 1 J is approximately 4.187 calories, this means that $R^* = \frac{R}{4187}$.

Knowing $\dot{\omega}_{f,(iii)}$ and $K_c^{(iii)}(T)$ is enough to evaluate $\dot{\omega}_{b,(iii)}$. The reasoning is as follows: the reverse rate should be specified in such a way so that when (or if) chemical equilibrium is attained, the following condition is satisfied:

$$K_c^{(iii)}(T) = \frac{[\text{H}_2\text{O}]_{eq}}{[\text{H}_2]_{eq} [\text{O}_2]_{eq}^{\frac{1}{2}}} \quad (\text{H.28})$$

By inspection, the following form of the reverse rate will ensure that when $\dot{\omega}_{f,(iii)}|_{eq} = \dot{\omega}_{b,(iii)}|_{eq}$, then equation (H.28) is satisfied:

$$\dot{\omega}_{b,(iii)} = \frac{k_{f,(iii)}}{K_c^{(iii)}(T)} \frac{[\text{O}_2]^{\frac{7}{4}}}{[\text{H}_2]^{\frac{1}{2}}} \quad (\text{H.29})$$

The analysis is not yet complete. Unlike the reaction rate expressions appearing in the systematically reduced scheme (which are formed from elementary rates), in the present global scheme equation (5.16) indicates that as $[\text{H}_2\text{O}] \rightarrow 0$, $\dot{\omega}_{f,(iii)} \rightarrow \infty$ and as $[\text{H}_2] \rightarrow 0$, $\frac{\partial \dot{\omega}_{f,(iii)}}{\partial [\text{H}_2]} \rightarrow \infty$. In order to avoid this the reaction rate is arbitrarily 'clipped' when the mole fraction of H_2 or H_2O falls below 10⁻³%. Quantifying this parameter in terms of a specific mole number (Kmol/Kg),

$$\Upsilon \equiv \frac{\sum_{\alpha=1}^N n_{\alpha}}{1 \times 10^5} \quad (\text{H.30})$$

When $[\text{H}_2\text{O}] \leq \varrho \Upsilon$,

$$\begin{aligned} \dot{\omega}'_{f,(iii)} &= k_{f,(iii)} \frac{[\text{H}_2]^{\frac{1}{2}} [\text{O}_2]^{\frac{9}{4}}}{[\text{H}_2\text{O}]} \times \frac{[\text{H}_2\text{O}]}{\varrho \Upsilon} \\ &= k'_{f,(iii)} [\text{H}_2]^{\frac{1}{2}} [\text{O}_2]^{\frac{9}{4}} \end{aligned} \quad (\text{H.31})$$

The form of the reverse rate, consistent with equations (H.28) and (H.31) is given by,

$$\dot{\omega}'_{b,(iii)} = \frac{k'_{f,(iii)}}{K_c^{(iii)}(T)} \frac{[\text{H}_2\text{O}] [\text{O}_2]^{\frac{7}{4}}}{[\text{H}_2]^{\frac{1}{2}}} \quad (\text{H.32})$$

In order to avoid the problems created by $[\text{H}_2] \leq \varrho \Upsilon$, the exponent of the hydrogen concentration in equation (5.16) needs to be at least unity, so that when the expression is differentiated with respect to $[\text{H}_2]$, this concentration does not appear in the denominator. The modification that is applied when $[\text{H}_2] \leq \varrho \Upsilon$ takes the form,

$$\begin{aligned} \dot{\omega}''_{f,(iii)} &= k_{f,(iii)} \frac{[\text{H}_2]^{\frac{1}{2}} [\text{O}_2]^{\frac{9}{4}}}{[\text{H}_2\text{O}]} \times \frac{[\text{H}_2]^{\frac{1}{2}}}{(\varrho \Upsilon)^{\frac{1}{2}}} \\ &= k''_{f,(iii)} \frac{[\text{H}_2] [\text{O}_2]^{\frac{9}{4}}}{[\text{H}_2\text{O}]} \end{aligned} \quad (\text{H.33})$$

It can then be shown that $\dot{\omega}''_{b,(iii)} = k''_{f,(iii)} (K_c^{(iii)})^{-1} [\text{O}_2]^{\frac{7}{4}}$. However, equations (H.32) and (H.33) would still lead to problems if $[\text{H}_2]$ or $[\text{H}_2\text{O}]$ tended to zero, respectively. So the following

situation needs to be catered for too: when $[\text{H}_2]$ and $[\text{H}_2\text{O}]$ are less than $\varrho\Upsilon$,

$$\begin{aligned}\dot{\omega}_{f,(iii)}''' &= k_{f,(iii)} \frac{[\text{H}_2]^{\frac{1}{2}} [\text{O}_2]^{\frac{9}{4}}}{[\text{H}_2\text{O}]} \times \frac{[\text{H}_2]^{\frac{1}{2}}}{(\varrho\Upsilon)^{\frac{1}{2}}} \times \frac{[\text{H}_2\text{O}]}{\varrho\Upsilon} \\ &= k_{f,(iii)}''' [\text{H}_2] [\text{O}_2]^{\frac{9}{4}}\end{aligned}\tag{H.34}$$

with the result that $\dot{\omega}_{b,(iii)}''' = k_{f,(iii)}''' (K_c^{(iii)})^{-1} [\text{H}_2\text{O}] [\text{O}_2]^{\frac{7}{4}}$.

Figures Relating to Calculations

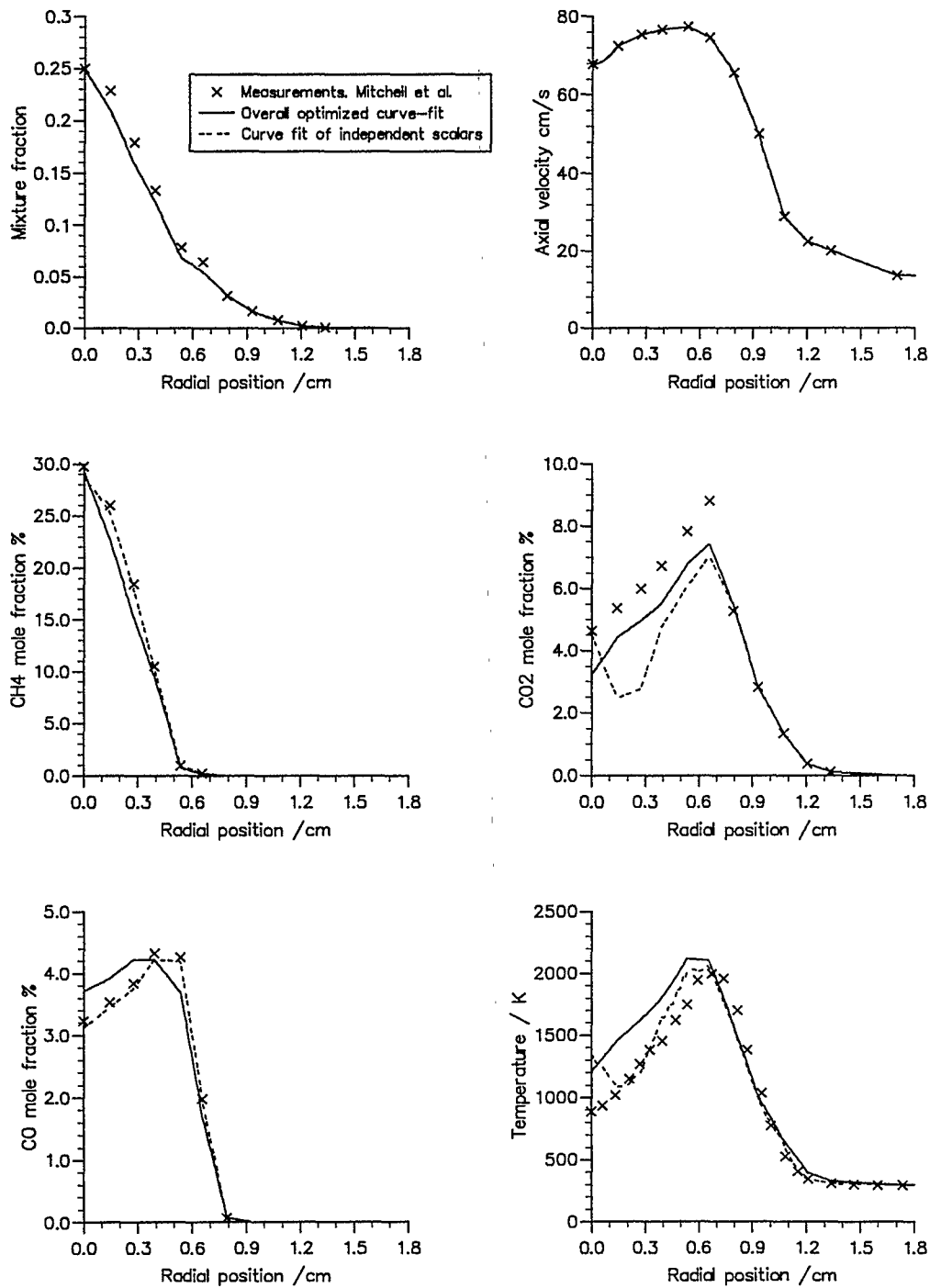


Figure 6.4: Initial conditions for ξ , CH_4 , CO , axial velocity, CO_2 and temperature specified at the first measuring station ($x = 12\text{mm}$) of the flame of Mitchell et al. [69].

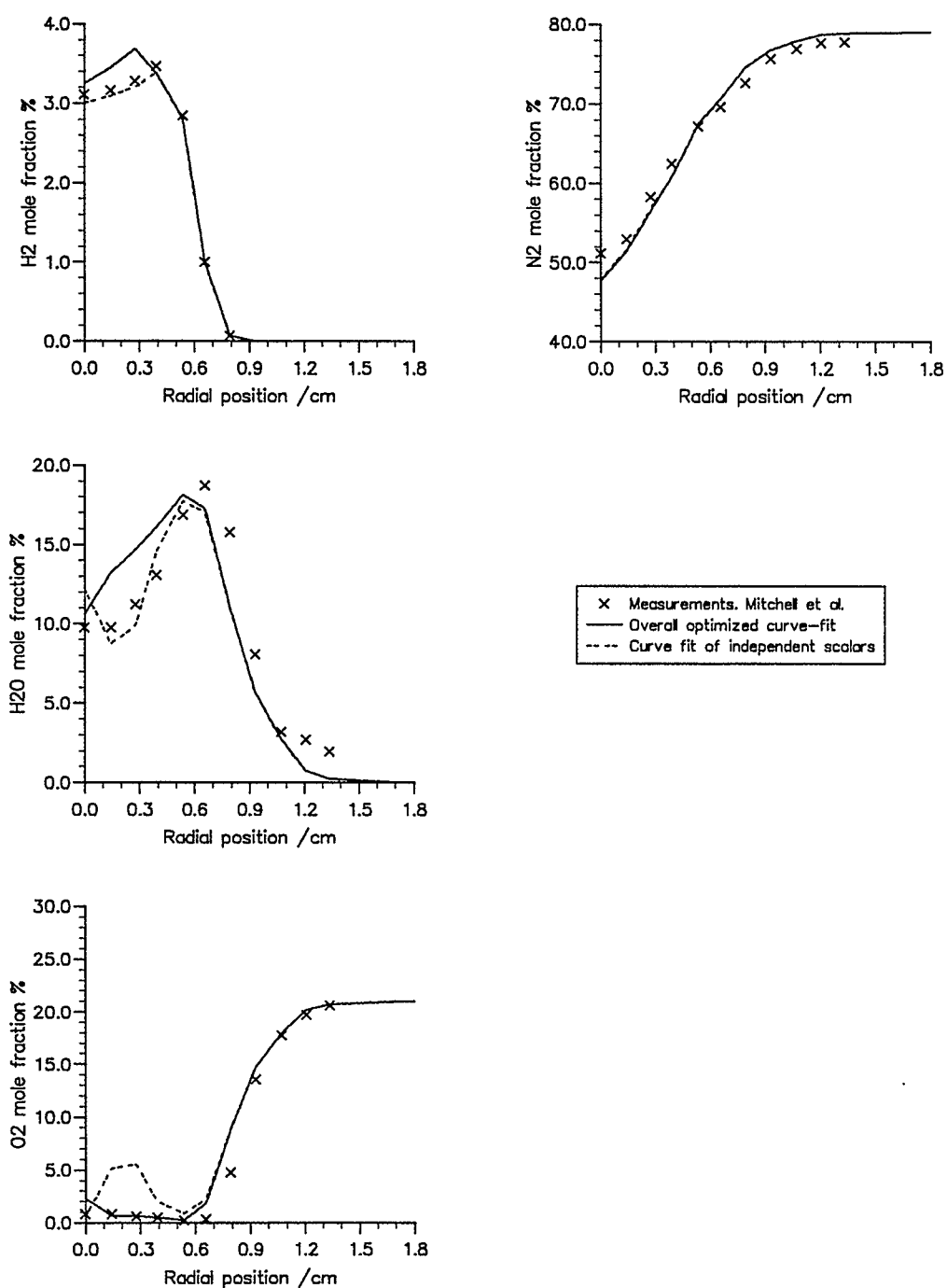


Figure 6.5: Initial conditions for H₂, H₂O, O₂ and N₂ specified at the first measuring station ($x = 12\text{mm}$) of the flame of Mitchell et al. [69].

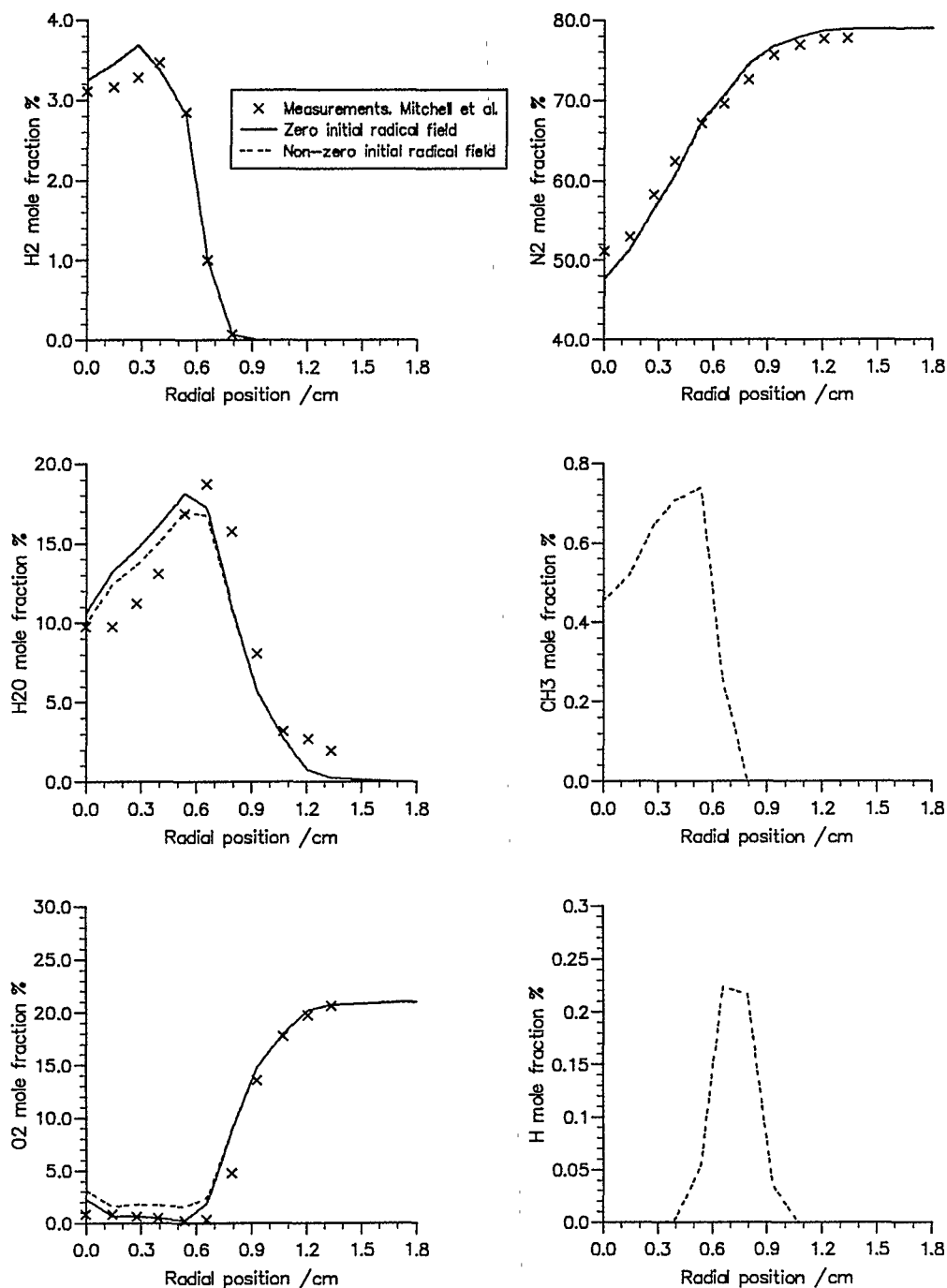


Figure 6.6: Initial conditions for H_2 , H_2O , O_2 , N_2 , CH_3 and H specified at the first measuring station ($x = 12\text{mm}$) of the flame of Mitchell et al. [69]. The effect of specifying radical species in the initial condition is investigated.

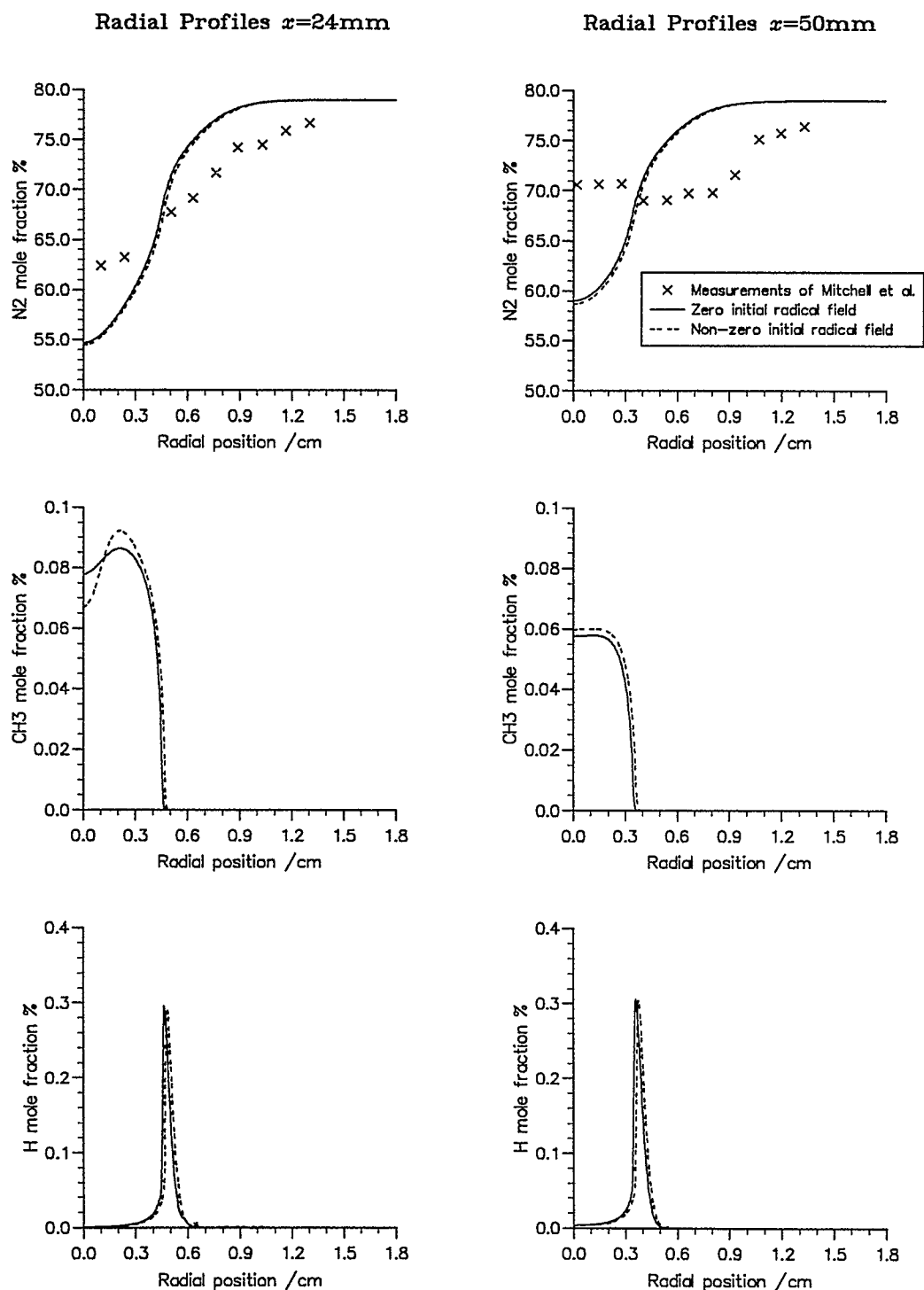


Figure 6.7: Radial profiles of N_2 , CH_3 and H from the calculation of the laminar flame of Mitchell et al. [69] using the reduced scheme of Chelliah et al. [10]; the interpolation table is *not* used. The effect of the two initial conditions specified in figure 6.6 is investigated.

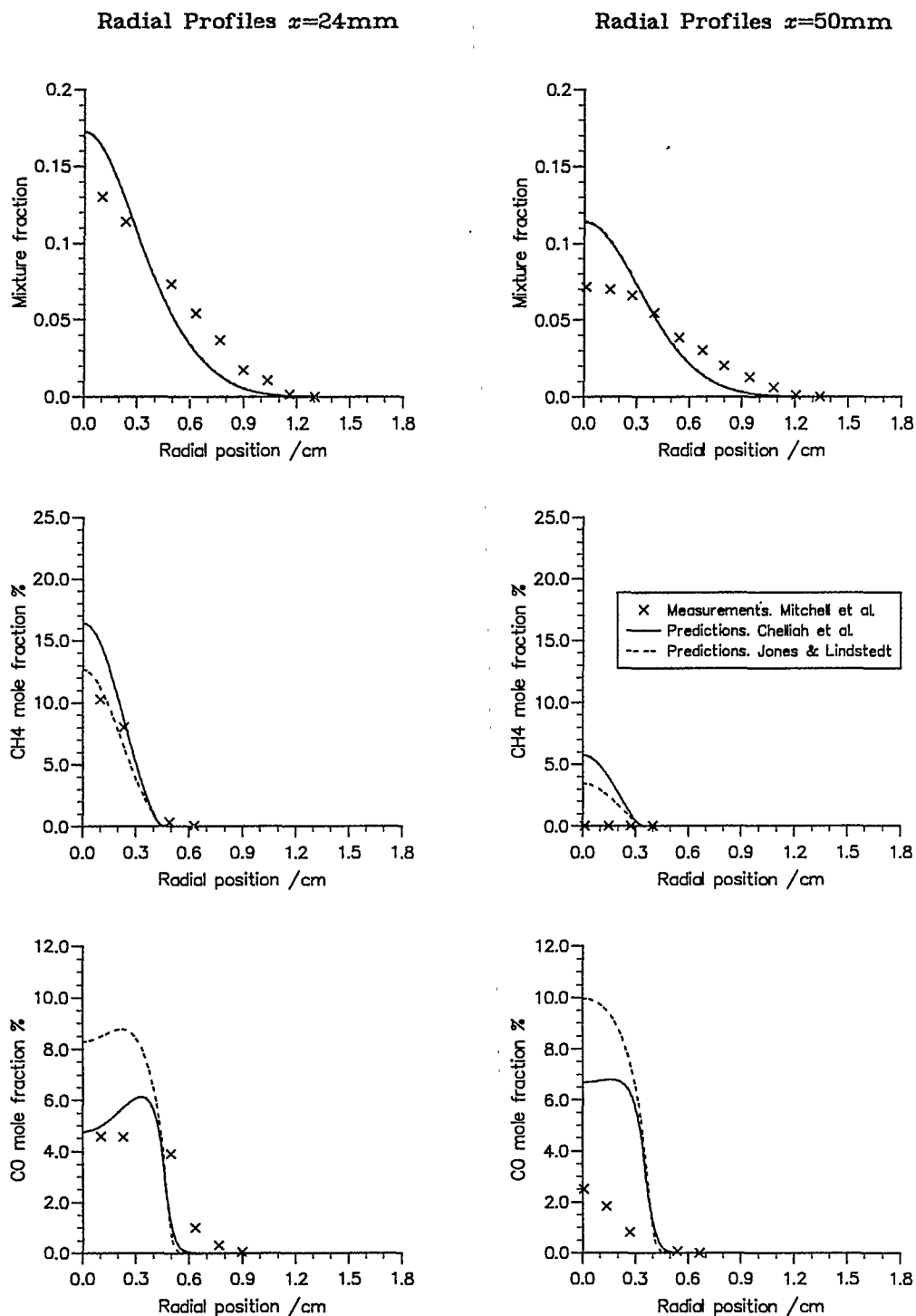


Figure 6.8: Radial profiles of ξ , CH_4 and CO from the calculation of the laminar flame of Mitchell et al. [69] using the reduced schemes of Jones and Lindstedt [46] and Chelliah et al. [10]; the interpolation table is *not* used.

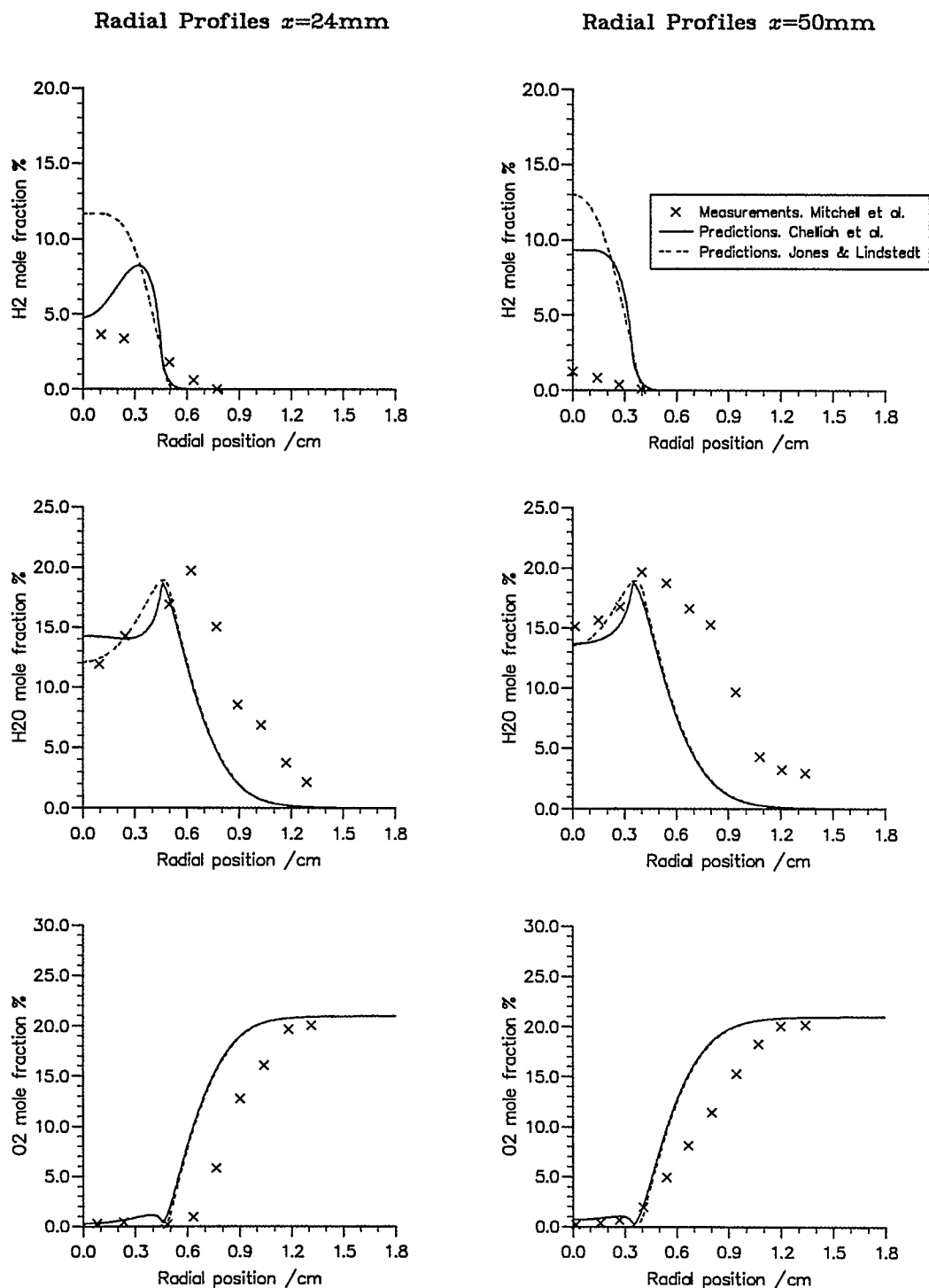


Figure 6.9: Radial profiles of H_2 , H_2O and O_2 from the calculation of the laminar flame of Mitchell et al. [69] using the reduced schemes of Jones and Lindstedt [46] and Chelliah et al. [10]; the interpolation table is *not* used.

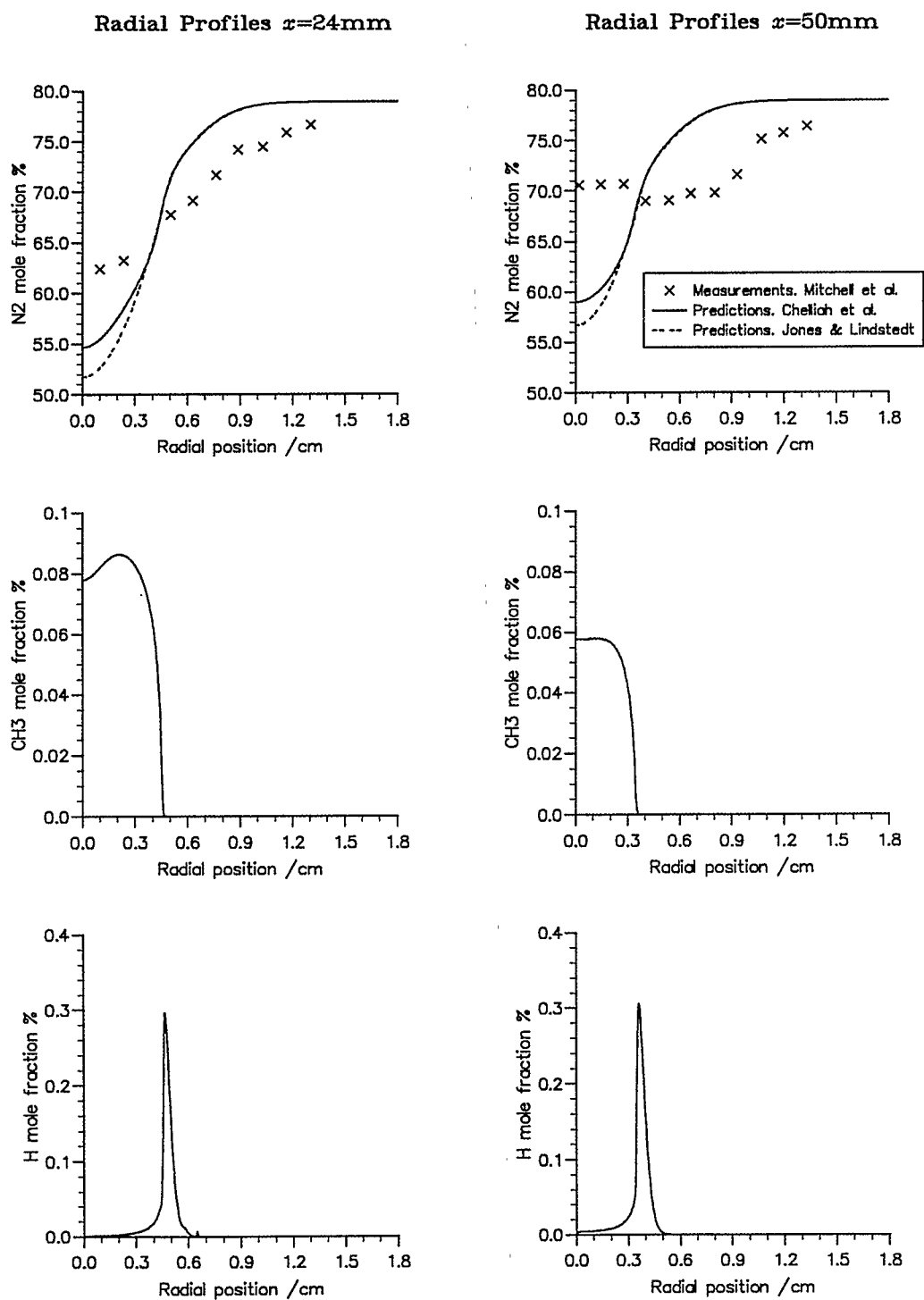


Figure 6.10: Radial profiles of N_2 , CH_3 and H from the calculation of the laminar flame of Mitchell et al. [69] using the reduced schemes of Jones and Lindstedt [46] and Chelliah et al. [10]; the interpolation table is *not* used.

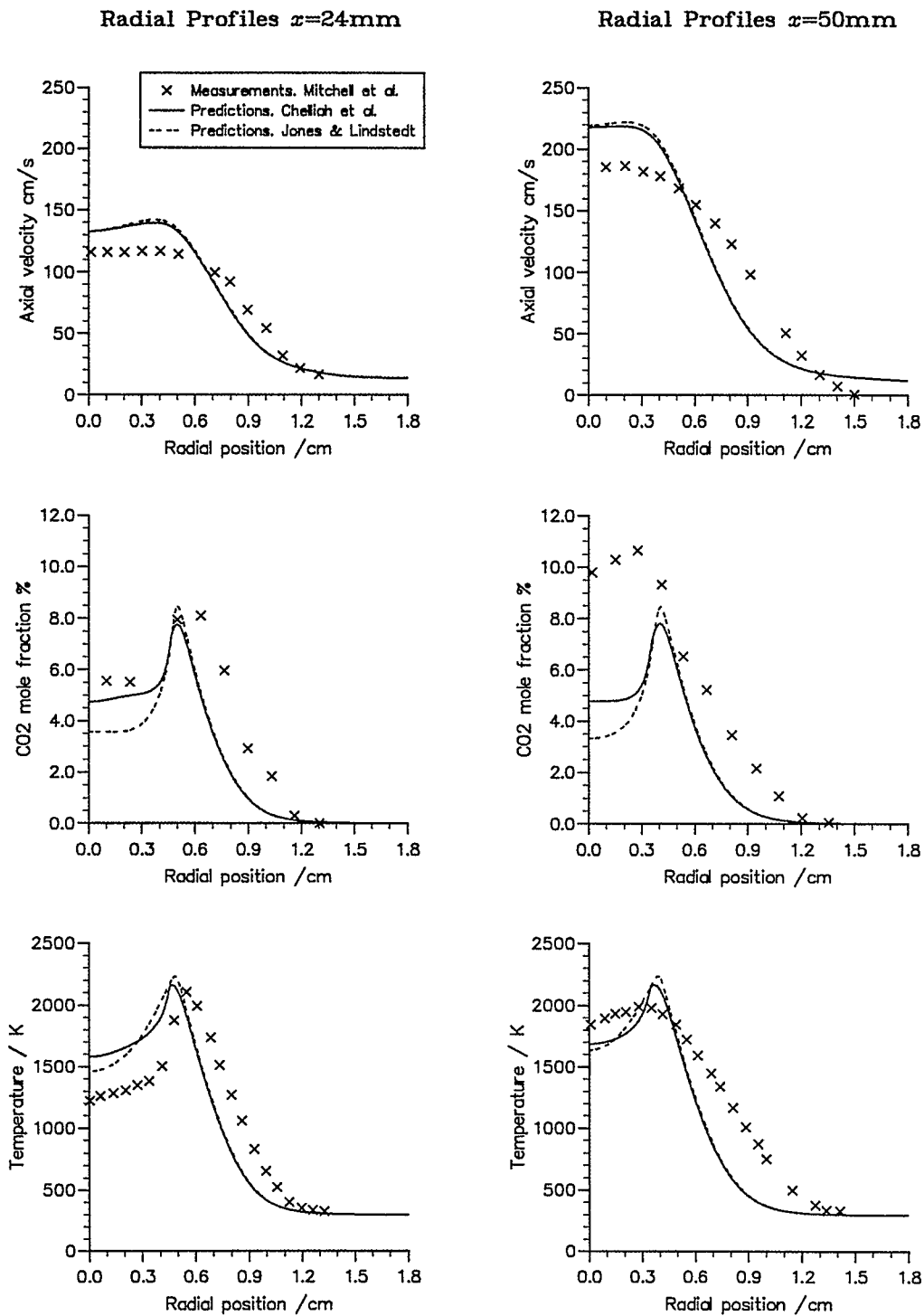


Figure 6.11: Radial profiles of axial velocity, CO₂ and temperature from the calculation of the laminar flame of Mitchell et al. [69] using the reduced schemes of Jones and Lindstedt [46] and Chelliah et al. [10]; the interpolation table is *not* used.

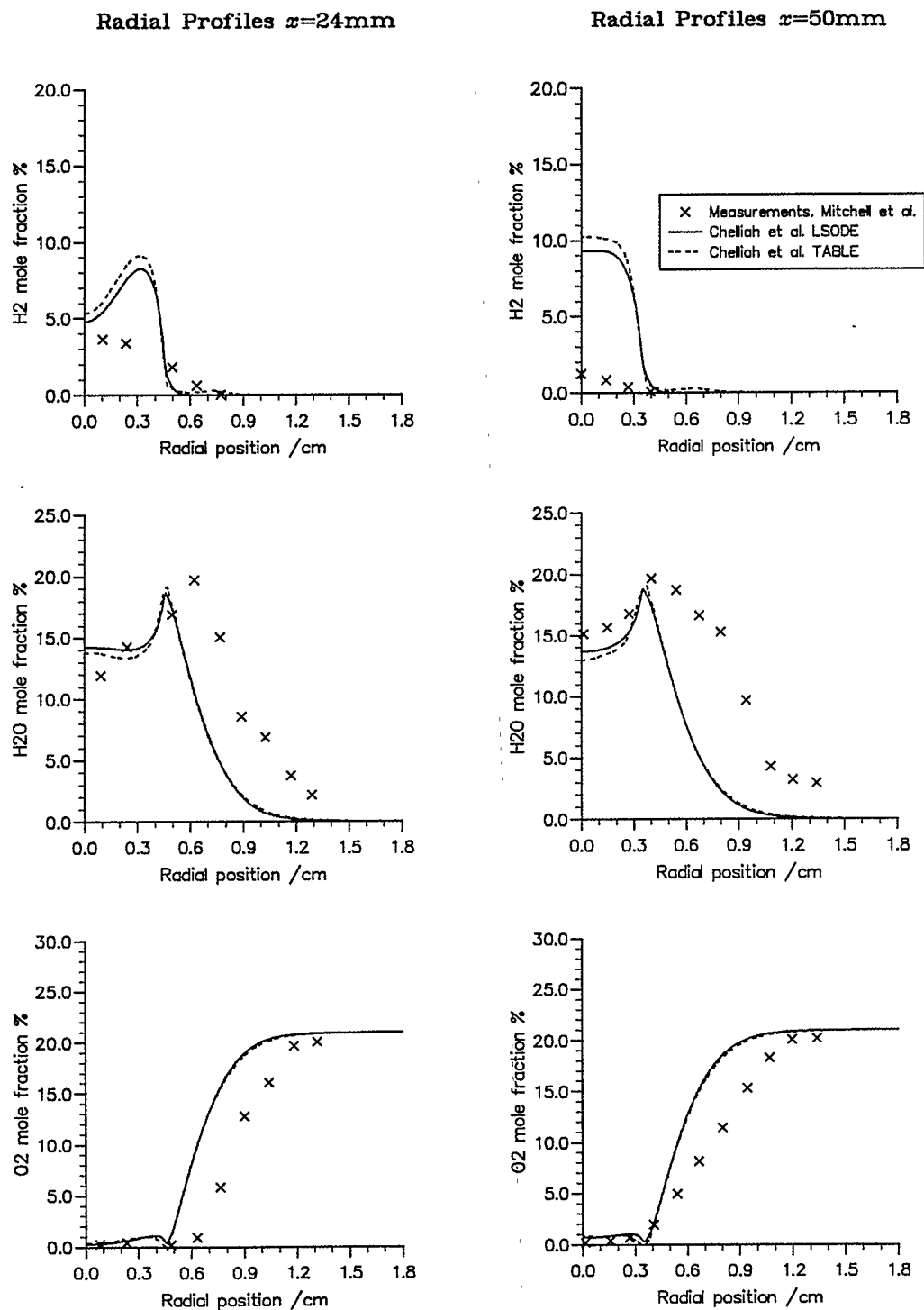


Figure 6.12: Radial profiles of H_2 , H_2O and O_2 from the calculation of the laminar flame of Mitchell et al. [69] using the reduced scheme of Chelliah et al. [10]; the results from LSODE and the 'look-up' (interpolation) table are compared.

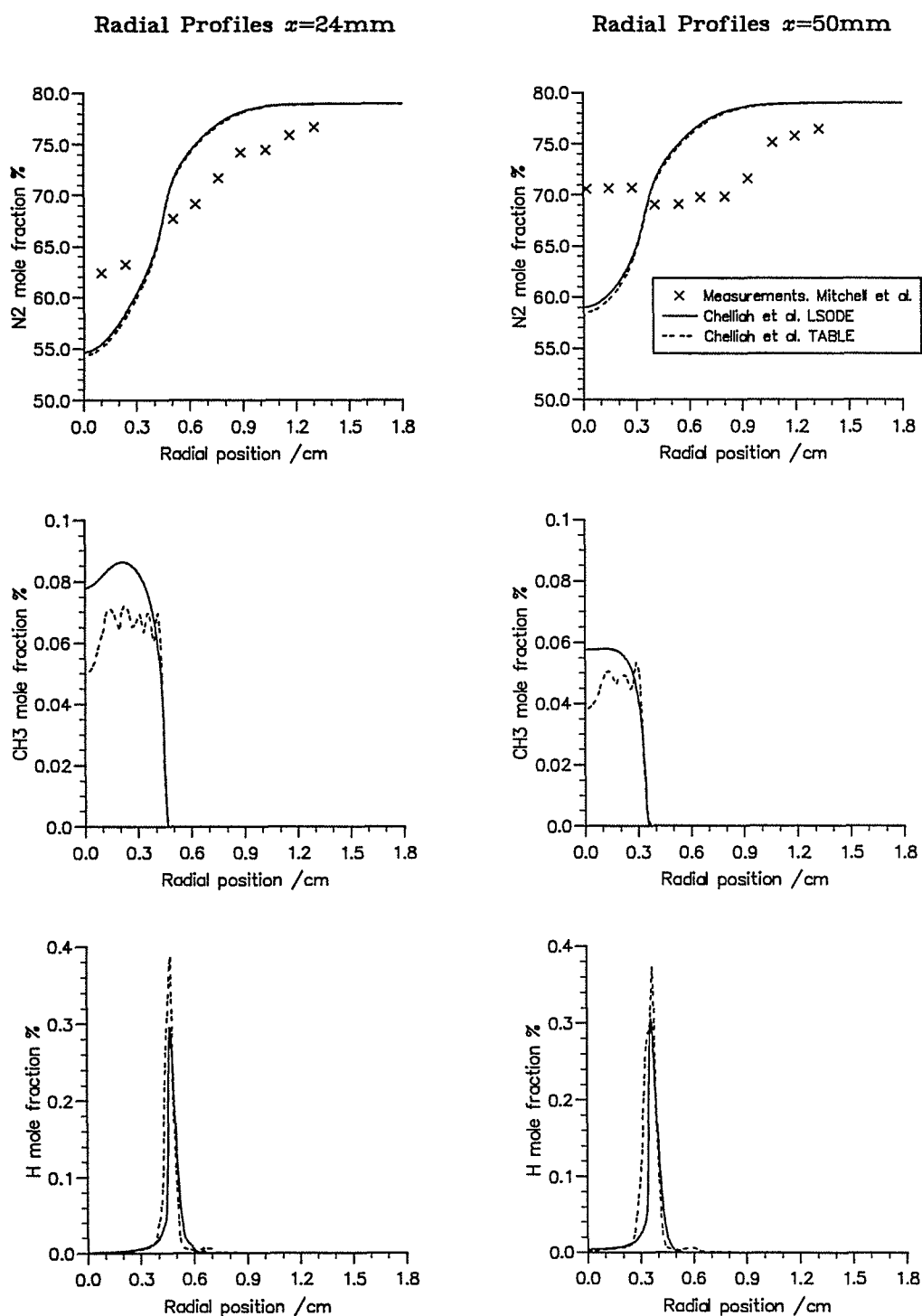


Figure 6.13: Radial profiles of N_2 , CH_3 and H from the calculation of the laminar flame of Mitchell et al. [69] using the reduced scheme of Chelliah et al. [10]; the results from LSODE and the 'look-up' (interpolation) table are compared.

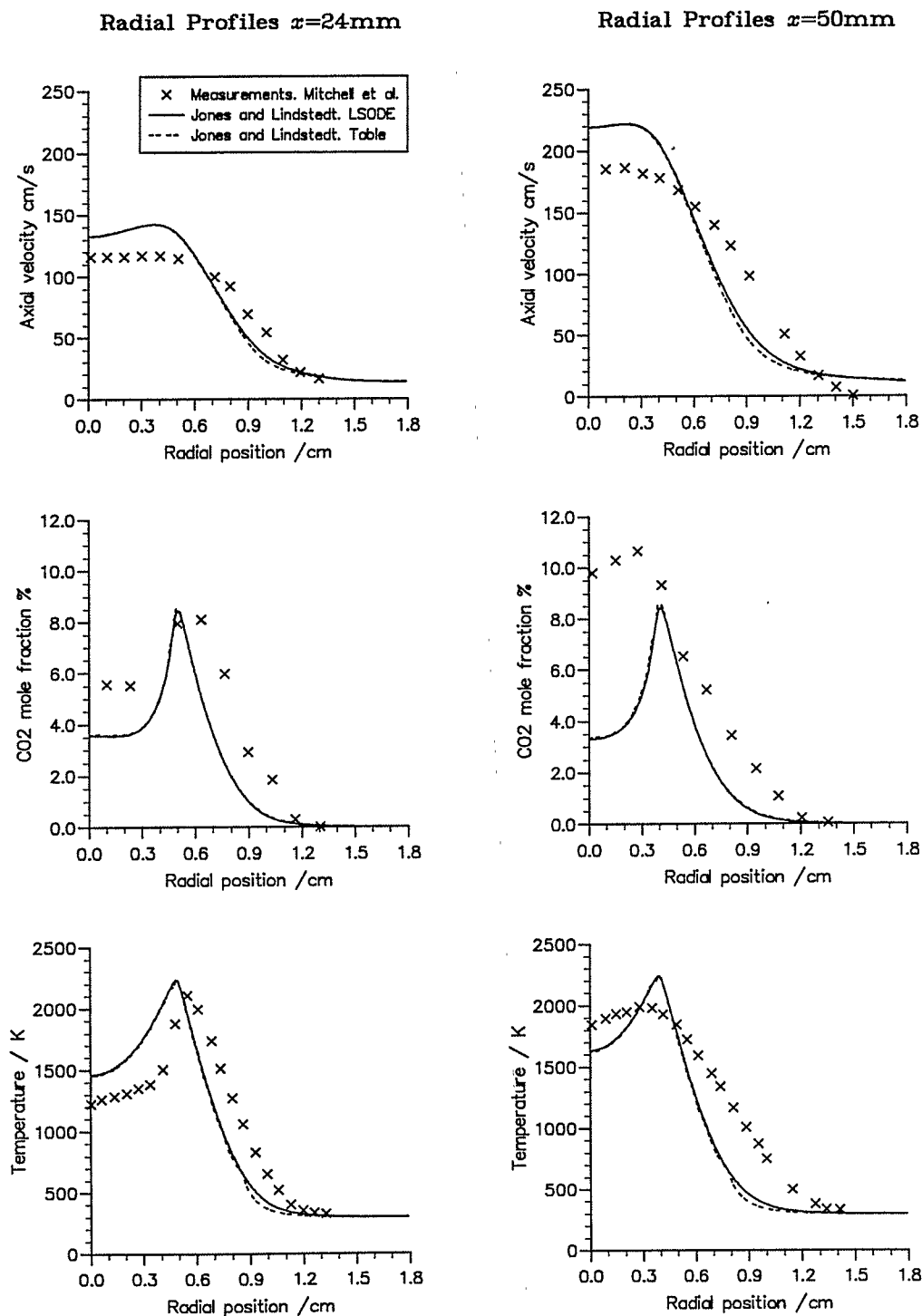


Figure 6.14: Radial profiles of axial velocity, CO₂ and temperature from the calculation of the laminar flame of Mitchell et al. [69] using the reduced scheme of Jones and Lindstedt [46]; the results from LSODE and the 'look-up' (interpolation) table are compared.

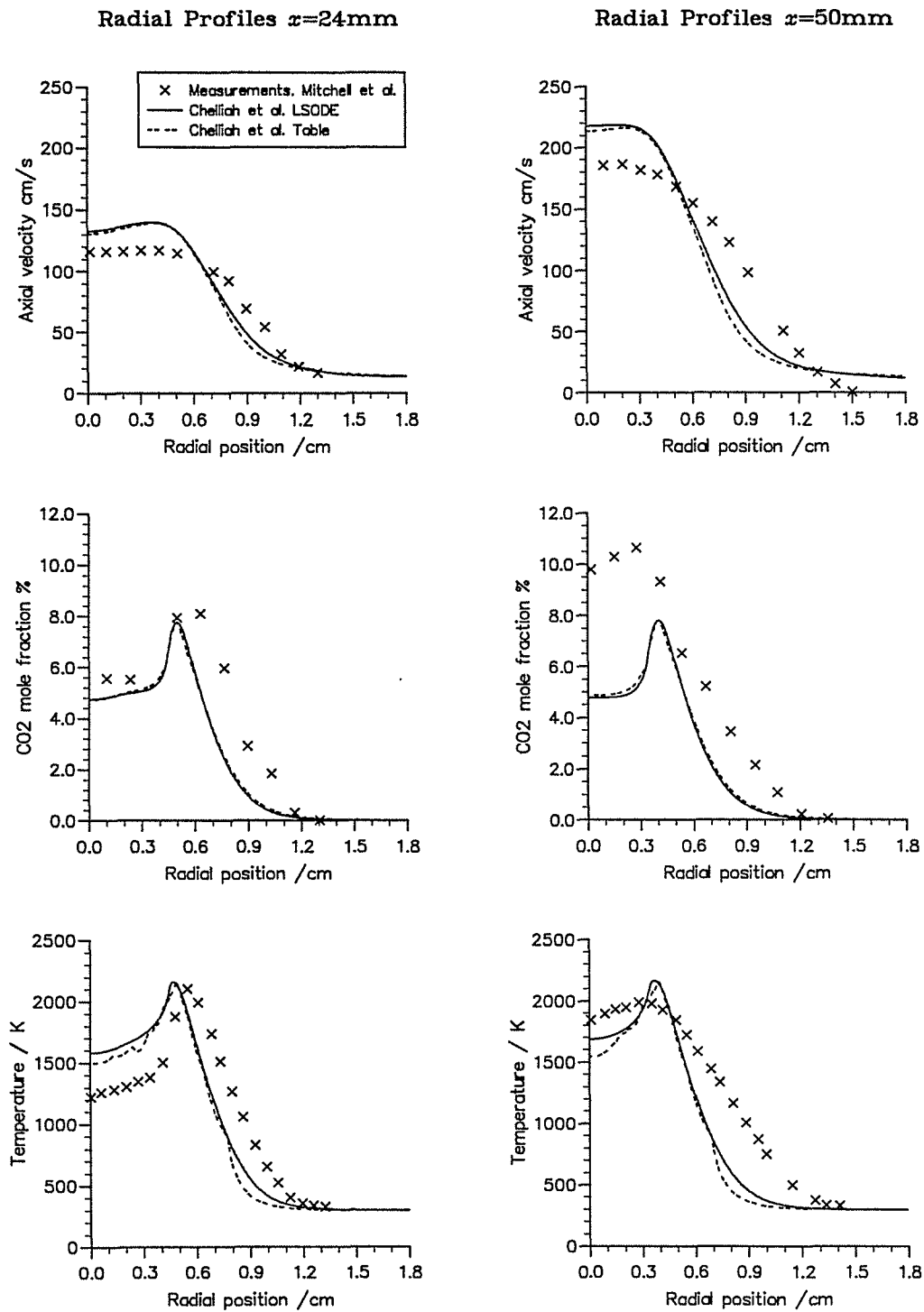


Figure 6.15: Radial profiles of axial velocity, CO₂ and temperature from the calculation of the laminar flame of Mitchell et al. [69] using the reduced scheme of Chelliah et al. [10]; the results from LSODE and the 'look-up' (interpolation) table are compared.

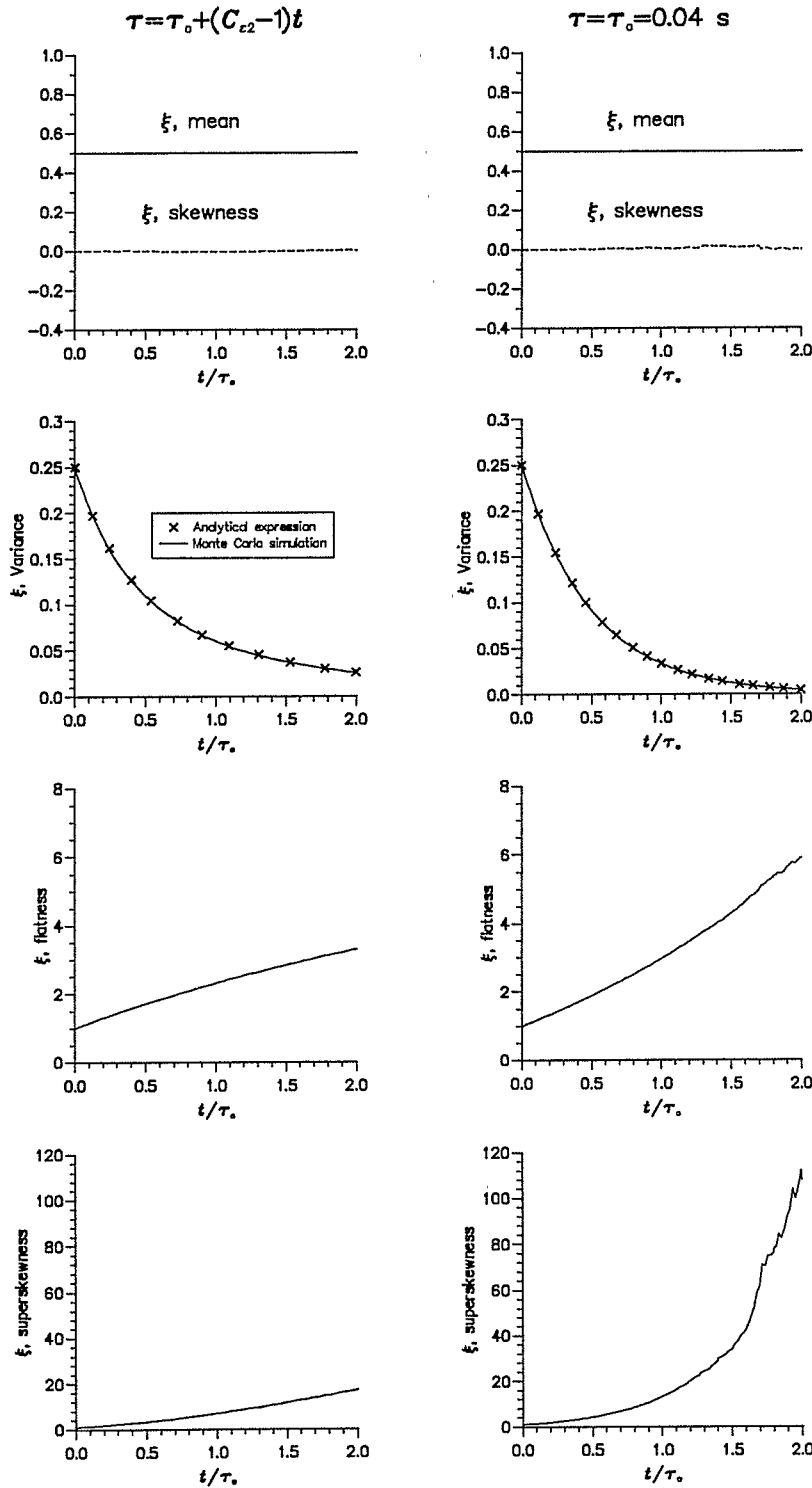


Figure 6.16: Predictions of the passive scalar statistics in homogeneous, isotropic and inert turbulence. Modified Curl is employed with a double δ -function initial condition and with two characteristic turbulent time scales ($\equiv \frac{k}{\epsilon}$).

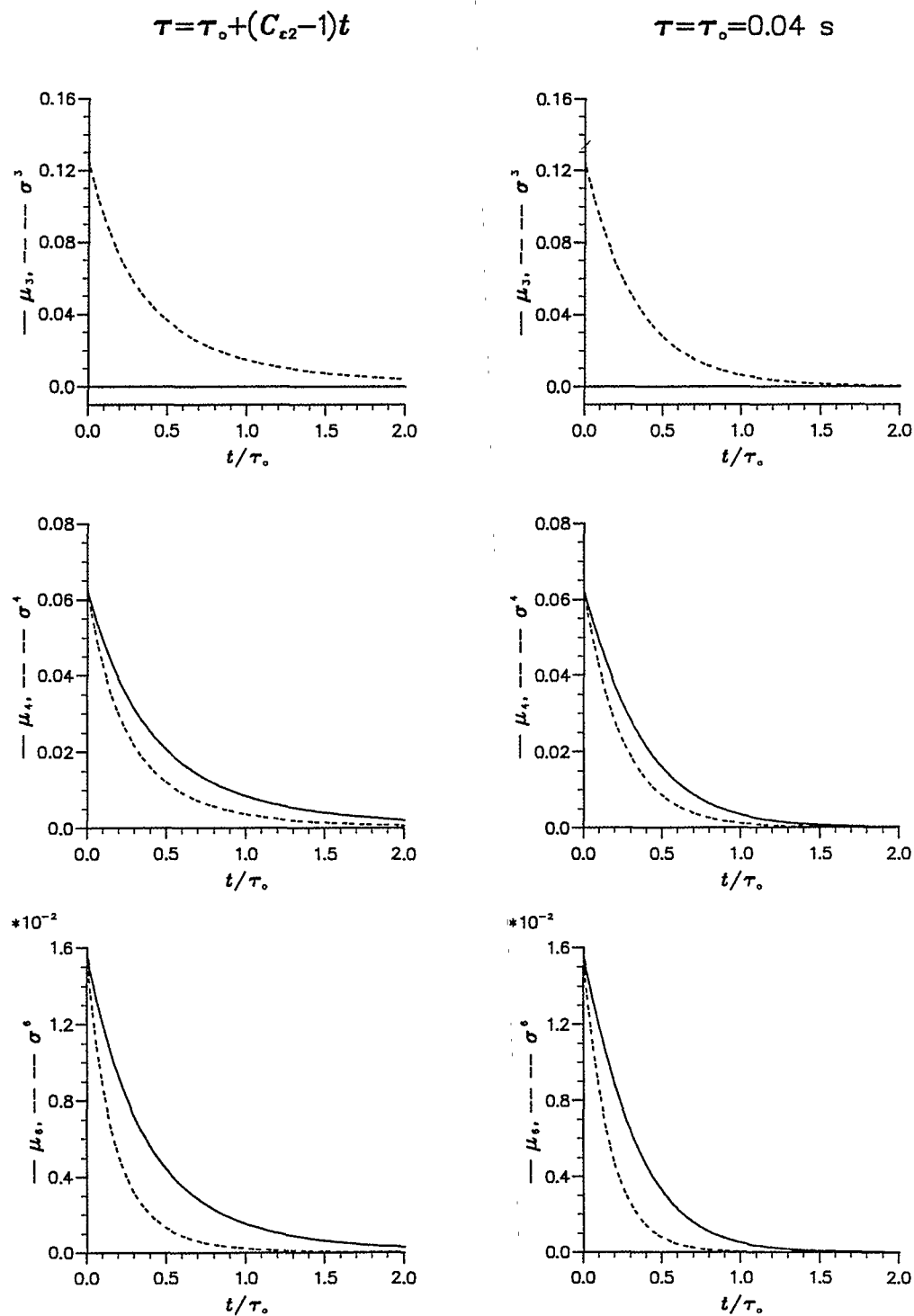


Figure 6.18: Passive scalar statistics in homogeneous, isotropic and inert turbulence. Modified Curl is employed with a double δ -function initial condition and with two characteristic turbulent time scales ($\equiv \frac{k}{\epsilon}$). The evolution of the individual moments are shown.

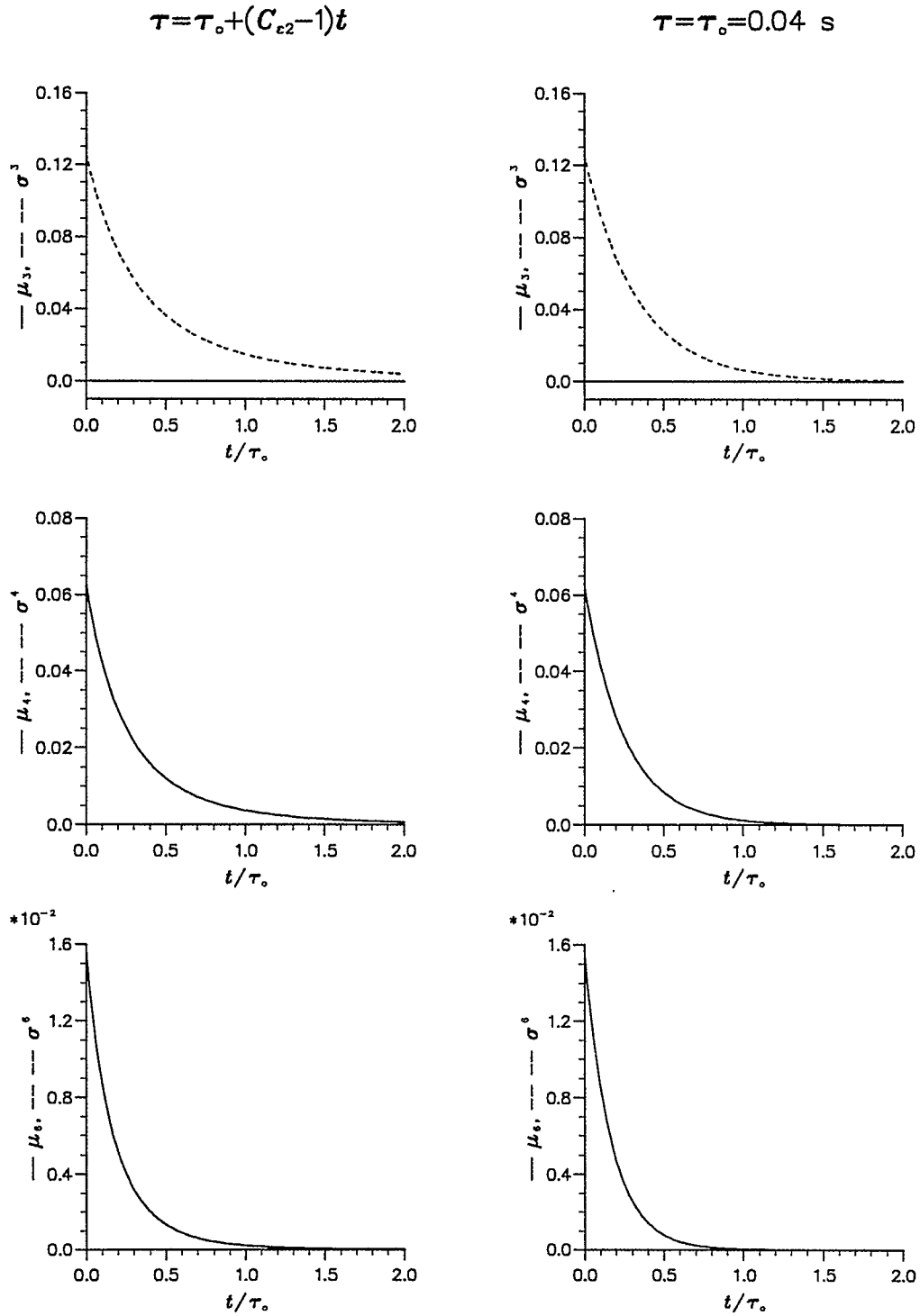


Figure 6.19: Passive scalar statistics in homogeneous, isotropic and inert turbulence. LMSE is employed with a double δ -function initial condition and with two characteristic turbulent time scales ($\equiv \frac{k}{\epsilon}$). The evolution of the individual moments are shown.

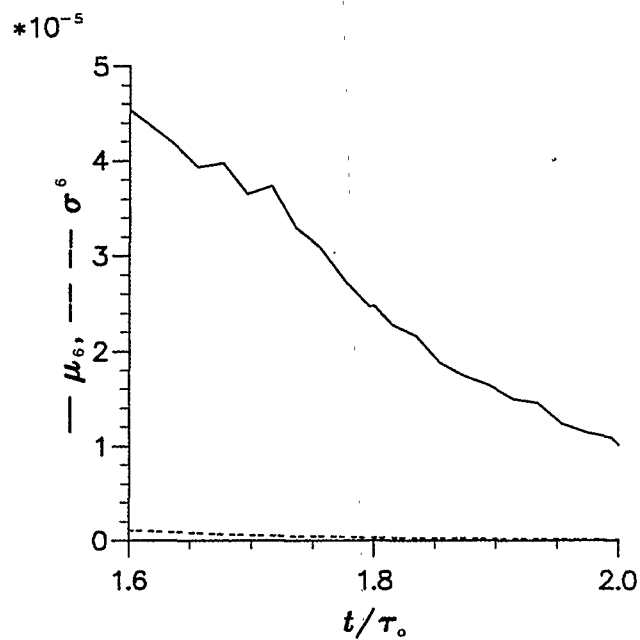


Figure 6.20: The same conditions as in figure 6.18 are employed. The behaviour of the individual moments in the final stages of mixing are expanded on to show why 'wiggles' appear in the plots of the normalised moments.

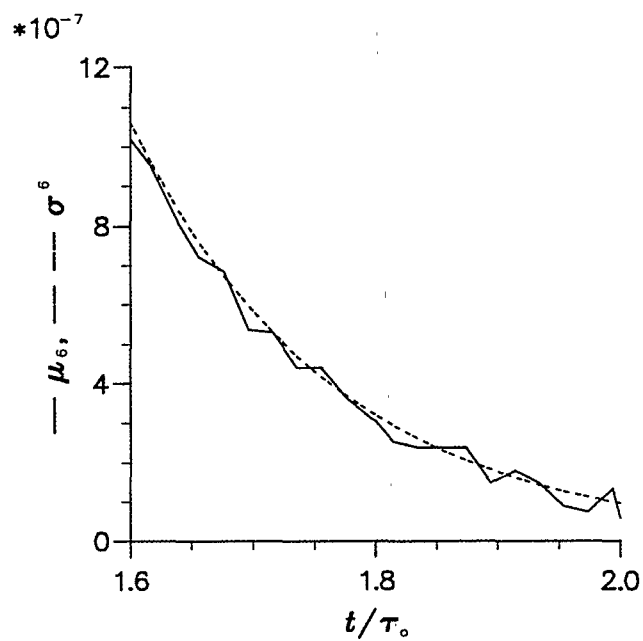


Figure 6.21: The same conditions as in figure 6.20 are employed, but with LMSE.

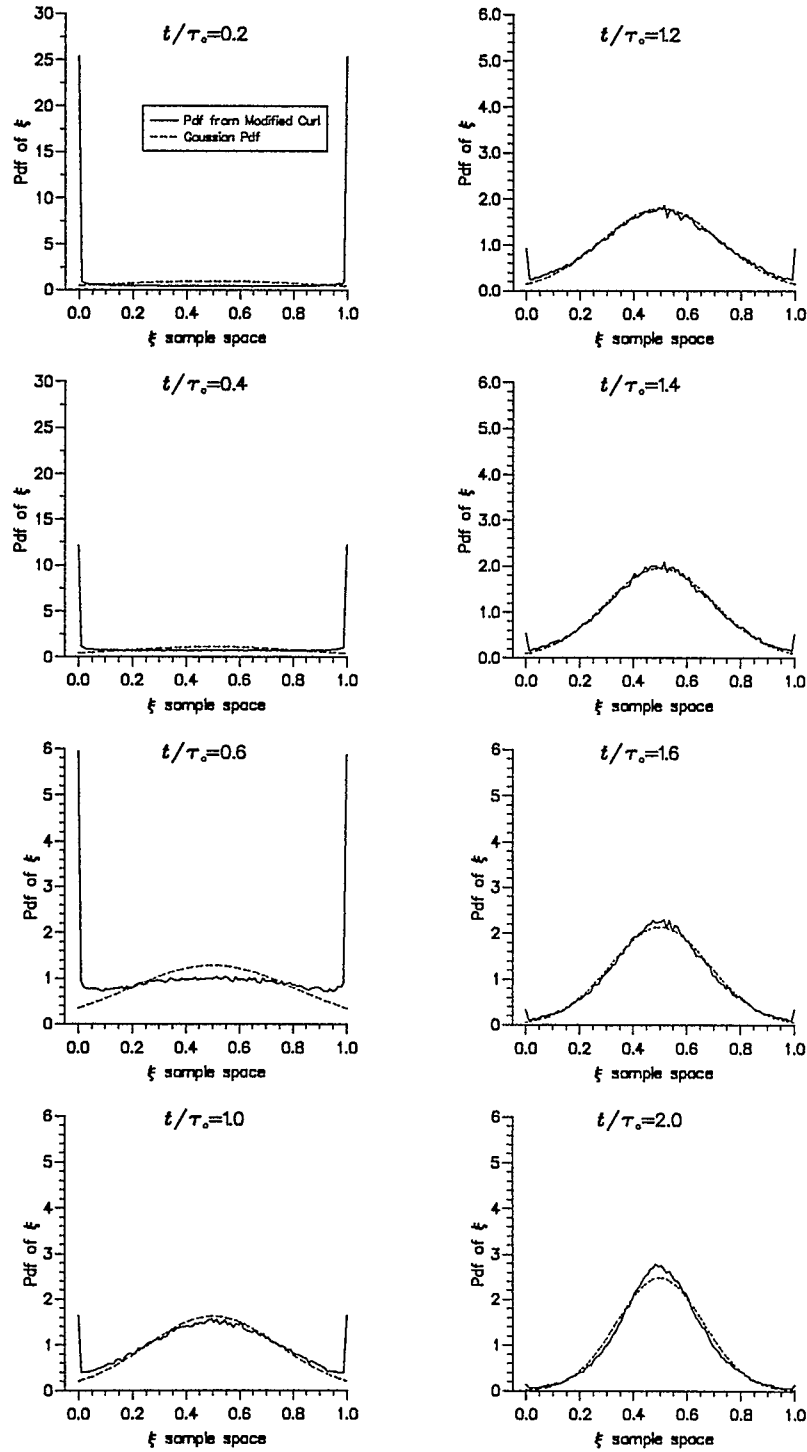


Figure 6.22: Evolution of the pdf for a passive scalar in homogeneous, isotropic and inert turbulence. Modified Curl is employed with a double δ -function initial condition and with $\tau(\equiv \frac{k}{\varepsilon}) = \tau_o + (C_{\varepsilon_2} - 1)t$, where $\tau_o = 0.04s$.

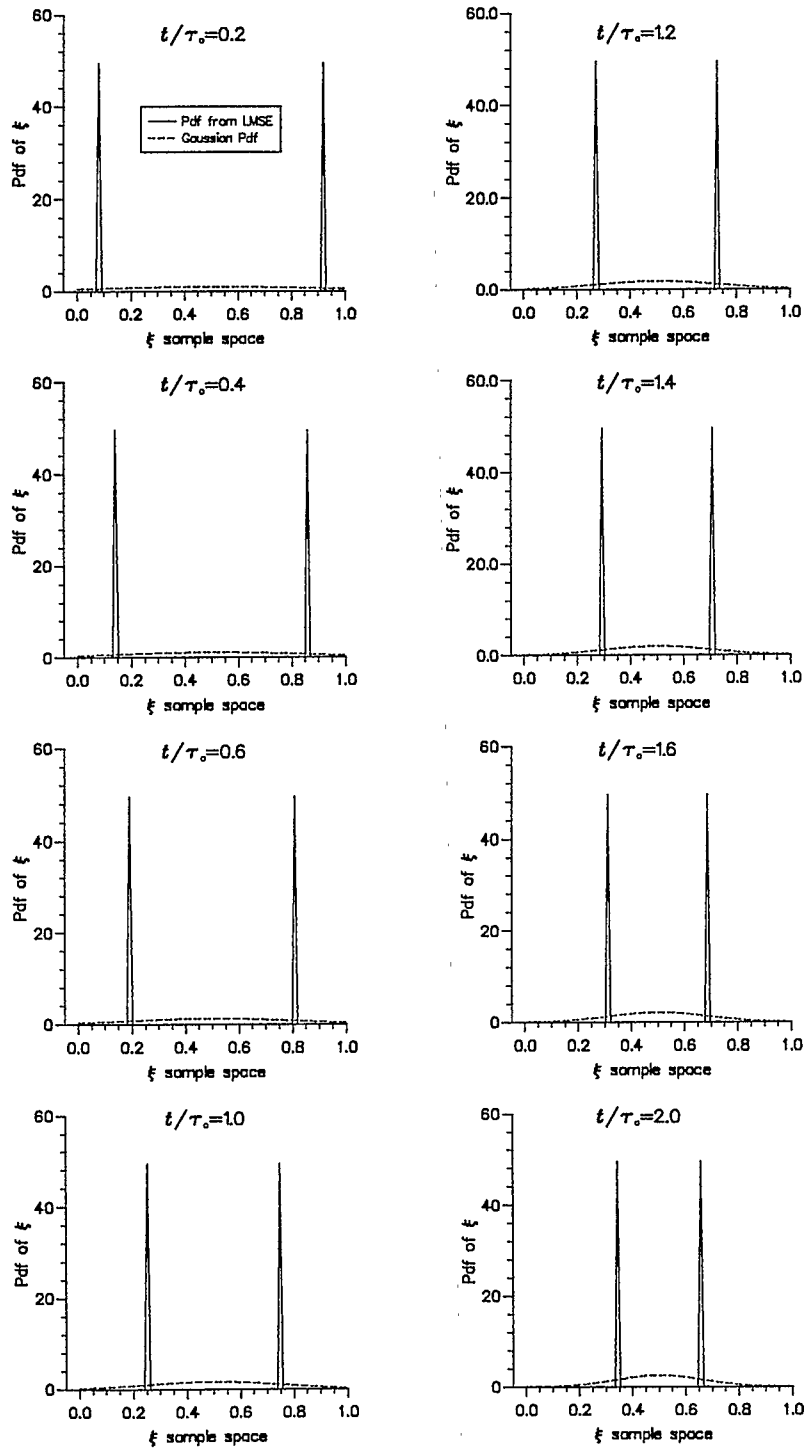


Figure 6.23: Evolution of the pdf for a passive scalar in homogeneous, isotropic and inert turbulence. LMSE is employed with a double δ -function initial condition and with $\tau(\equiv \frac{k}{\epsilon}) = \tau_o + (C_{\epsilon_2} - 1)t$, where $\tau_o = 0.04s$.

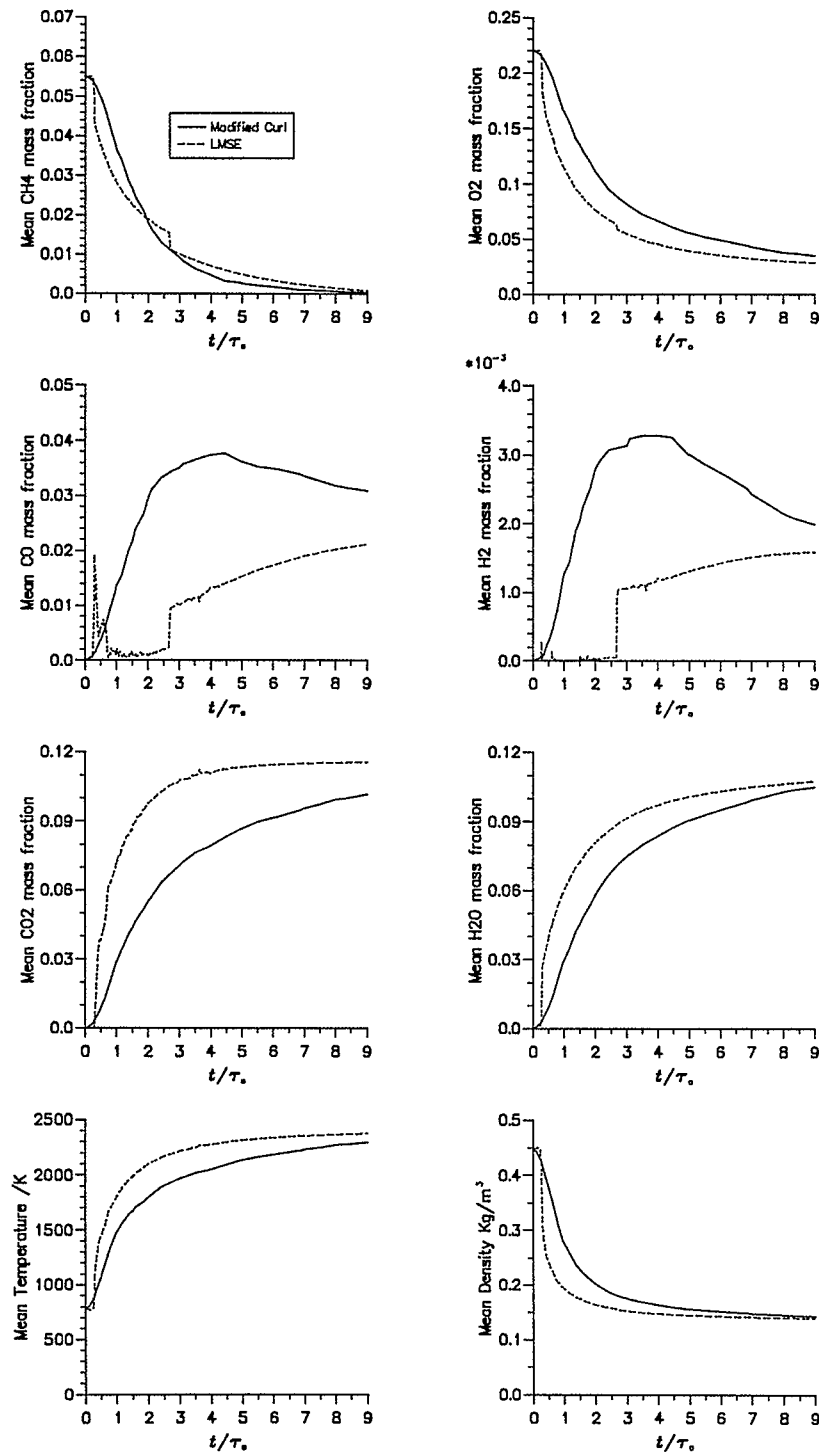


Figure 6.24: Predictions of the density-weighted mean reactive scalar properties in homogeneous, isotropic turbulence. Modified Curl and LMSE are compared starting with a double δ -function condition and $\tau(\equiv \frac{k}{\varepsilon}) = \tau_o + (C_{\varepsilon_2} - 1)t$, where $\tau_o = 0.04s$. The reduced scheme of Jones and Lindstedt [46] is used *without* the interpolation table. $\tilde{\xi} = \xi_{st} = 0.05$ (for CH₄).

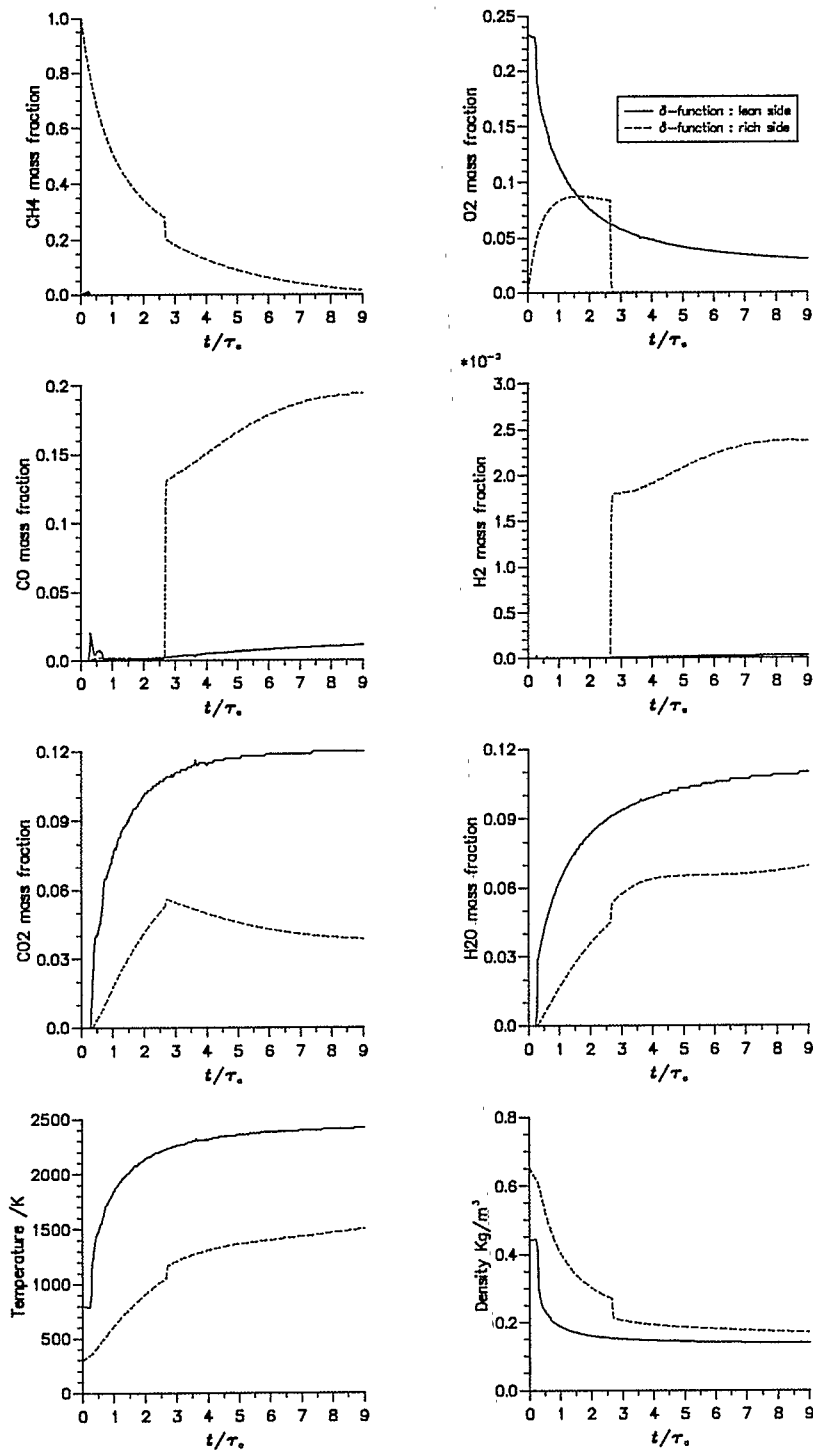


Figure 6.25: Predictions of the density-weighted mean reactive scalar properties in homogeneous, isotropic turbulence. The evolution of the initial δ -functions is monitored for the case of LMSE to explain the behaviour of this mixing model in figure 6.24).

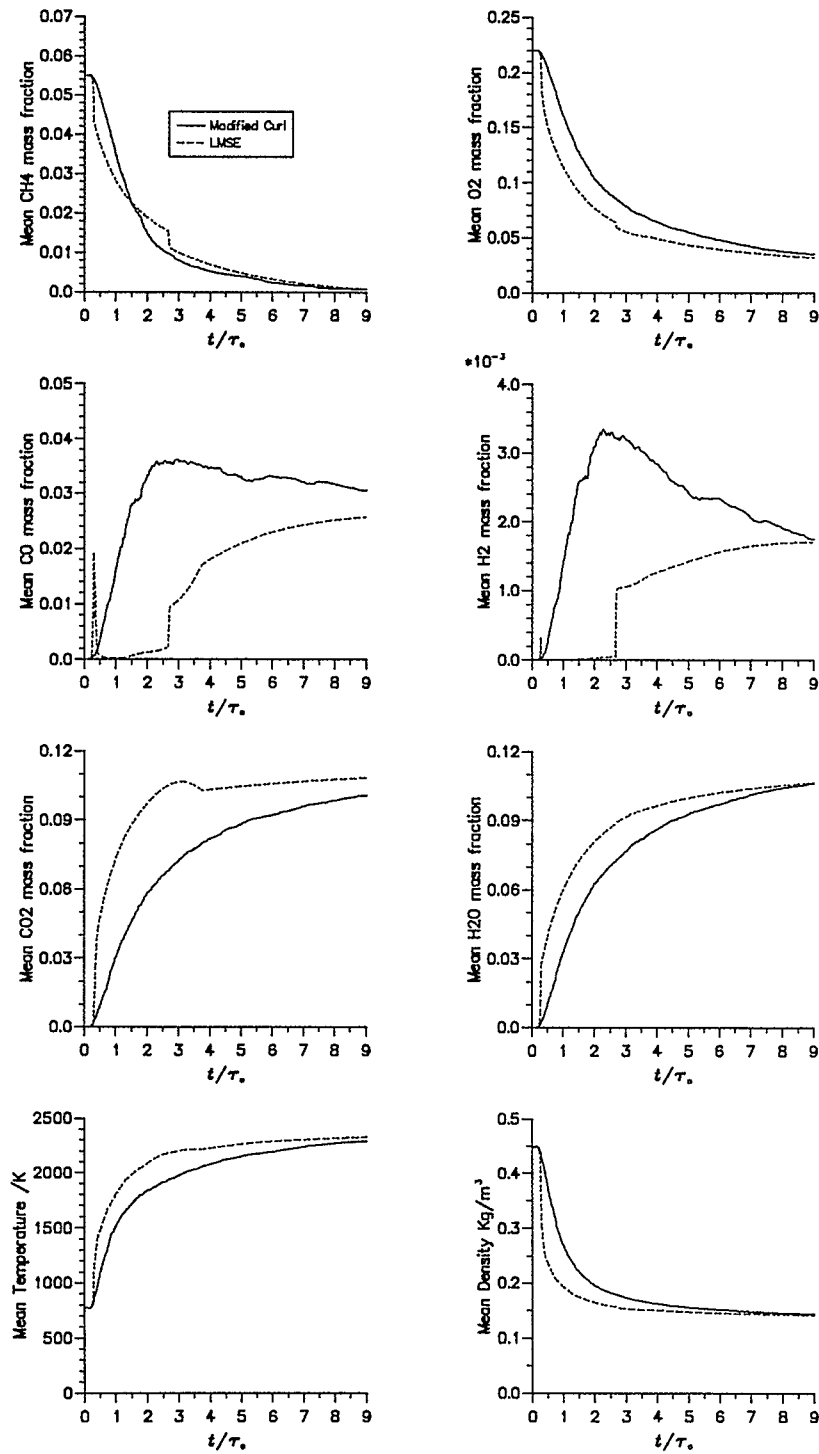


Figure 6.26: Predictions of the density-weighted mean reactive scalar properties in homogeneous, isotropic turbulence. Modified Curl and LMSE are compared starting with a double δ -function condition and $\tau(\equiv \frac{k}{\epsilon}) = \tau_o + (C_{\epsilon_2} - 1)t$, where $\tau_o = 0.04s$. The reduced scheme of Jones and Lindstedt [46] is used with the interpolation table. $\tilde{\xi} = \xi_{st} = 0.05$ (for CH₄).

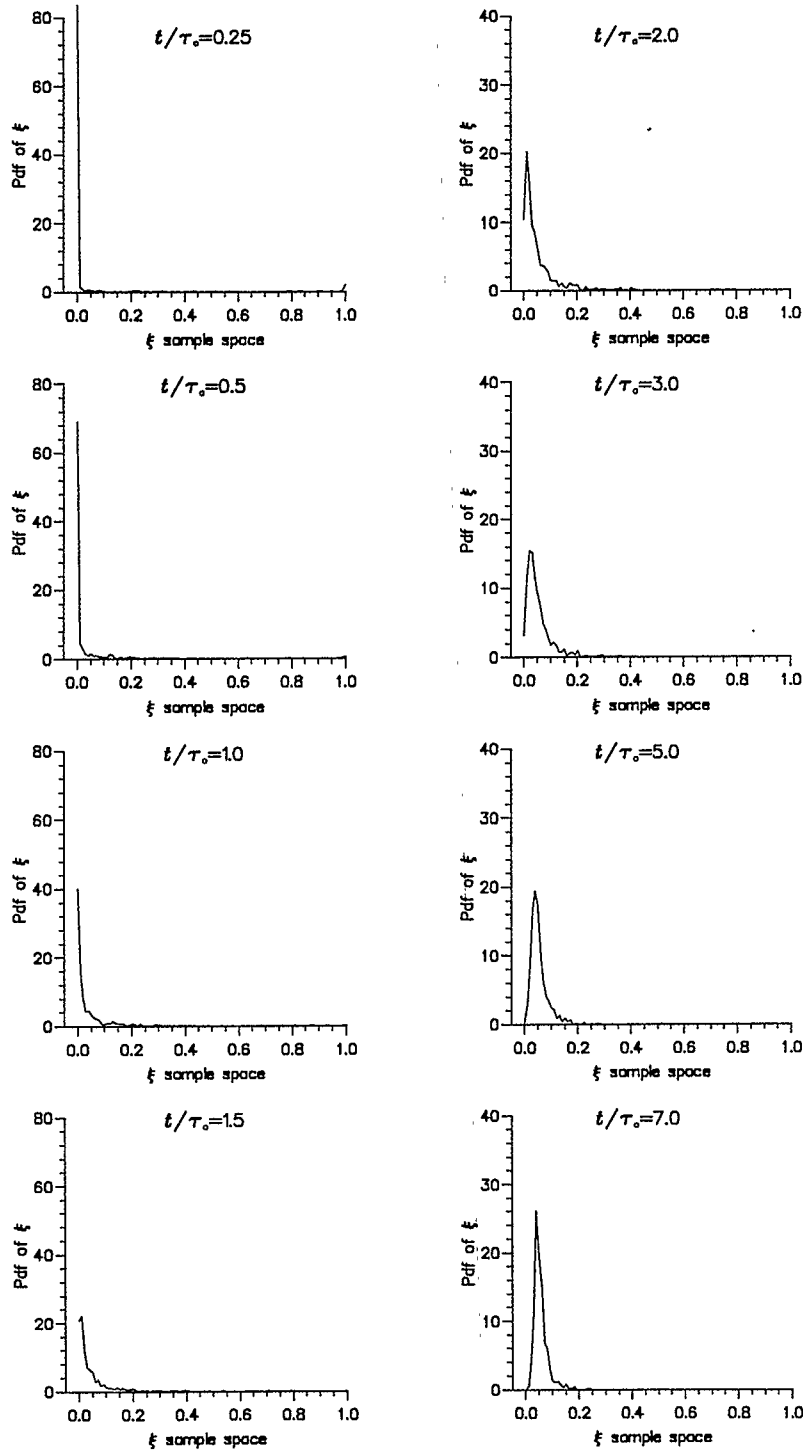


Figure 6.27: Evolution of $\tilde{P}(\tilde{\xi}; t)$ in homogeneous, isotropic turbulence. Modified Curl is employed with a double δ -function initial condition and with $\tau(\equiv \frac{k}{\epsilon}) = \tau_o + (C_{\epsilon_2} - 1)t$, where $\tau_o = 0.04s$. The reduced scheme of Jones and Lindstedt [46] is used *without* the interpolation table. $\tilde{\xi} = \xi_{st} = 0.05$ (for CH₄).

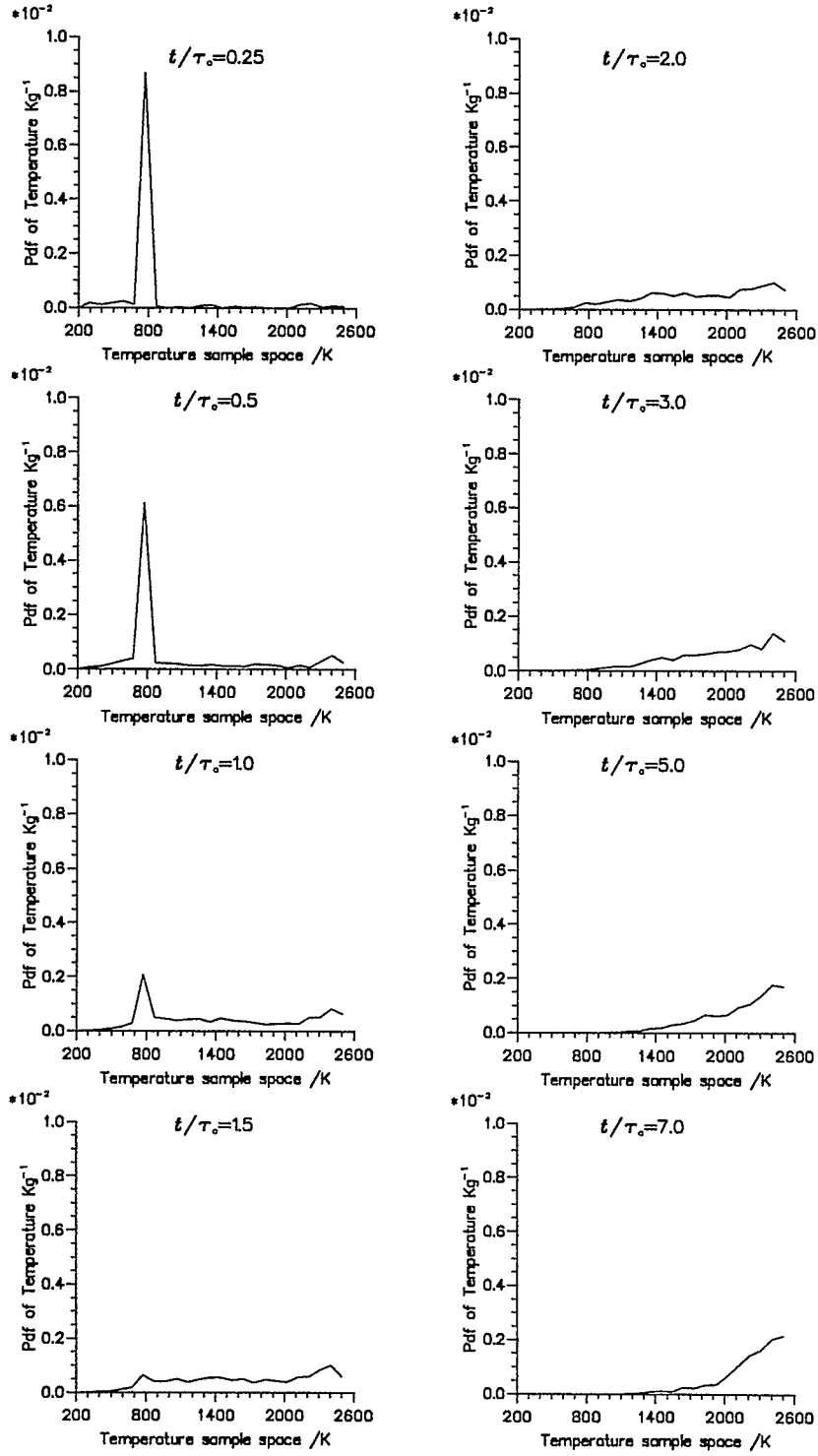


Figure 6.28: Evolution of $\tilde{P}(\hat{T}; t)$ in homogeneous, isotropic turbulence. Modified Curl is employed with a double δ -function initial condition and with $\tau(\equiv \frac{k}{\varepsilon}) = \tau_o + (C_{\varepsilon_2} - 1) t$, where $\tau_o = 0.04s$. The reduced scheme of Jones and Lindstedt [46] is used *without* the interpolation table. $\tilde{\xi} = \xi_{st} = 0.05$ (for CH_4).

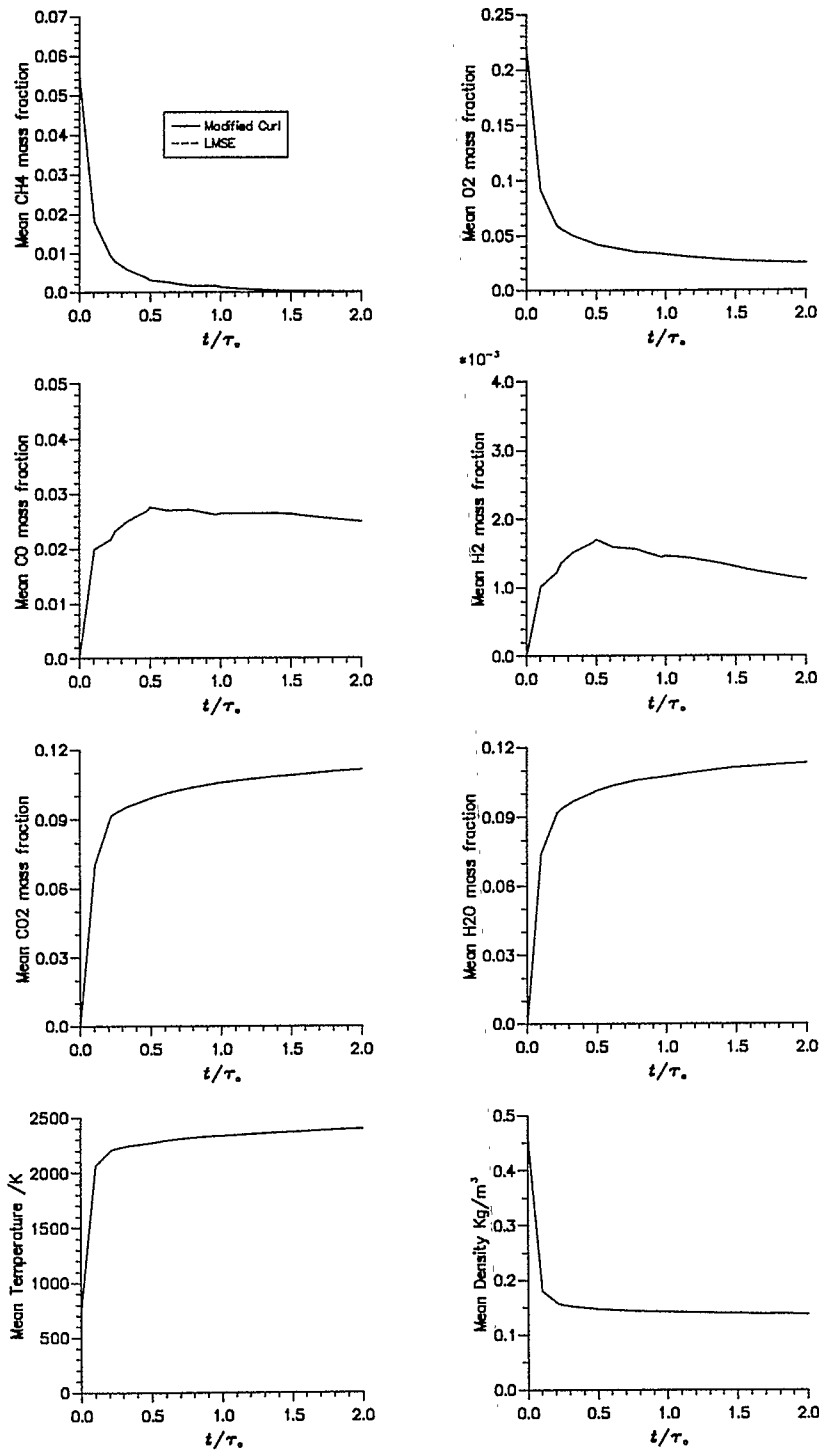


Figure 6.29: Predictions of the density-weighted mean reactive scalar properties in homogeneous, isotropic turbulence. Modified Curl and LMSE are compared starting with a continuous distribution taken from figure 6.27 at $t/\tau_o = 5$, where $\tau_o = 0.04$ s. The reduced scheme of Jones and Lindstedt [46] is used *without* the interpolation table. $\xi = \xi_{st} = 0.05$ (for CH₄).

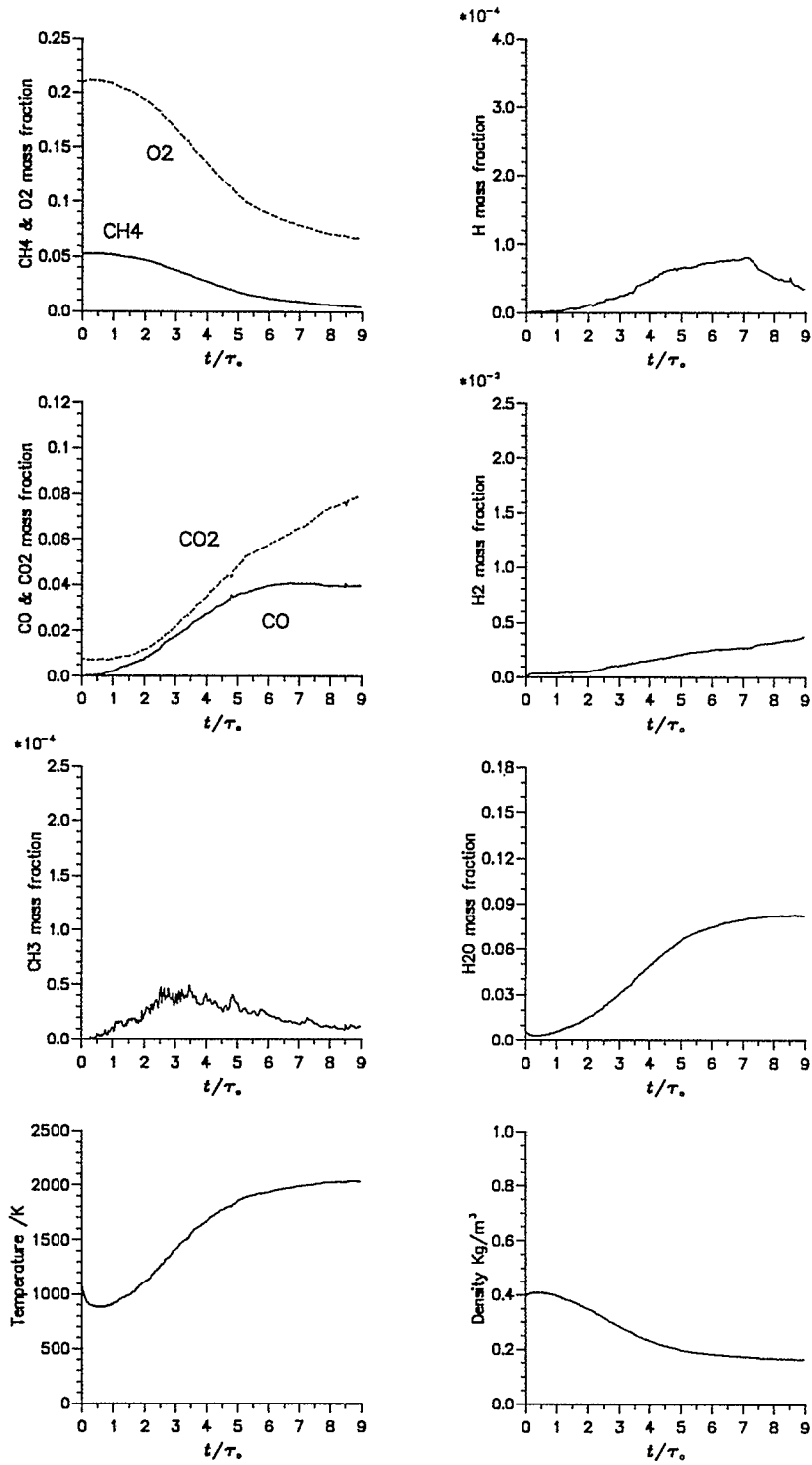


Figure 6.30: Predictions of the density-weighted mean reactive scalar properties in homogeneous, isotropic turbulence. Modified Curl is employed with a triple δ -function initial condition and $\tau(\equiv \frac{k}{\epsilon}) = \tau_0 + (C_{\epsilon_2} - 1)t$, where $\tau_0 = 0.04s$. The reduced scheme of Chelliah et al. [10] is used *without* the interpolation table. $\tilde{\xi} = \xi_{st} = 0.05$ (for CH₄).

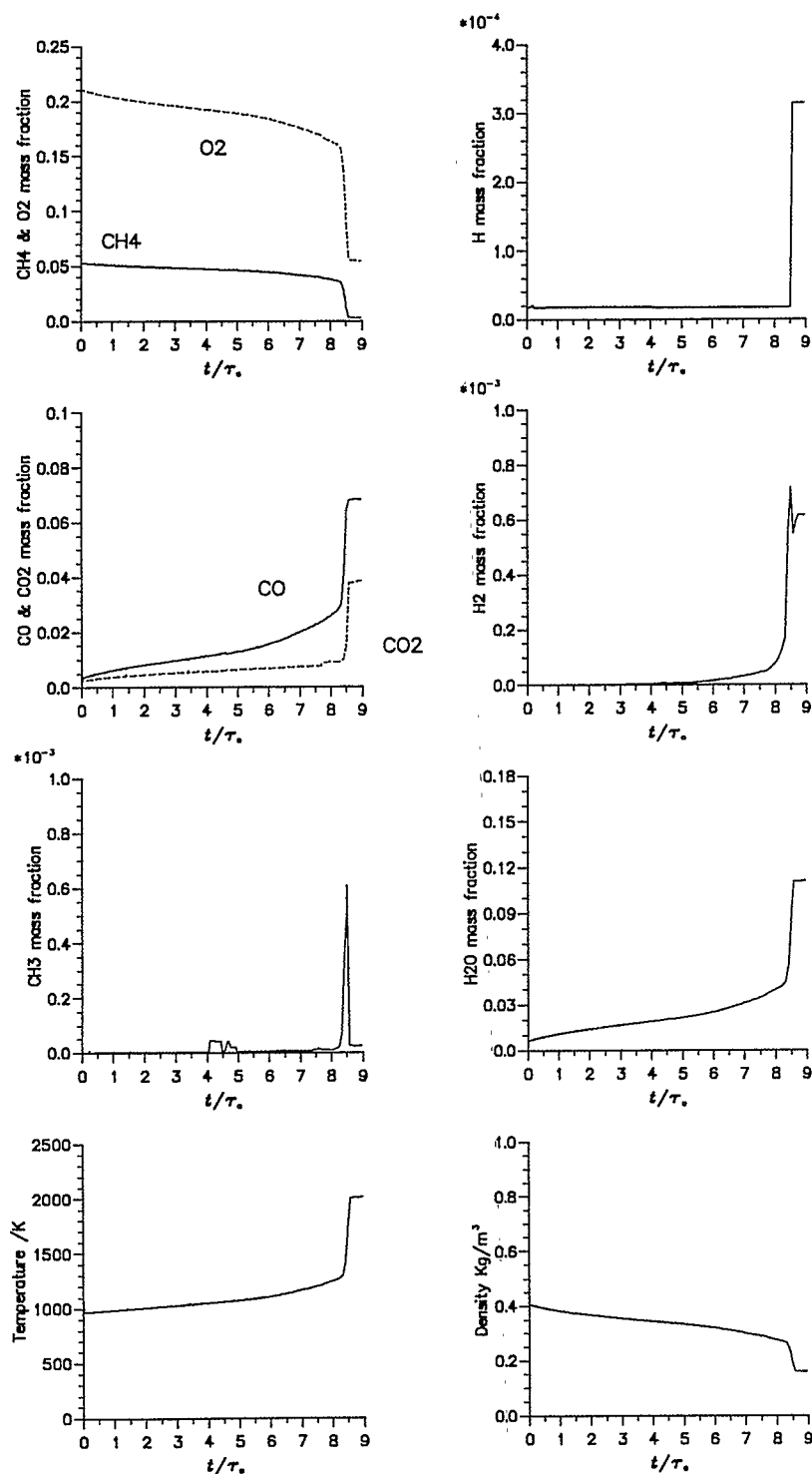


Figure 6.31: Predictions of the density-weighted mean reactive scalar properties in homogeneous, isotropic turbulence. LMSE is employed with a triple δ -function initial condition and $\tau(\equiv \frac{k}{\epsilon}) = \tau_0 + (C_{\epsilon_2} - 1)t$, where $\tau_0 = 0.04s$. The reduced scheme of Chelliah et al. [10] is used *without* the interpolation table. $\xi = \xi_{st} = 0.05$ (for CH₄).

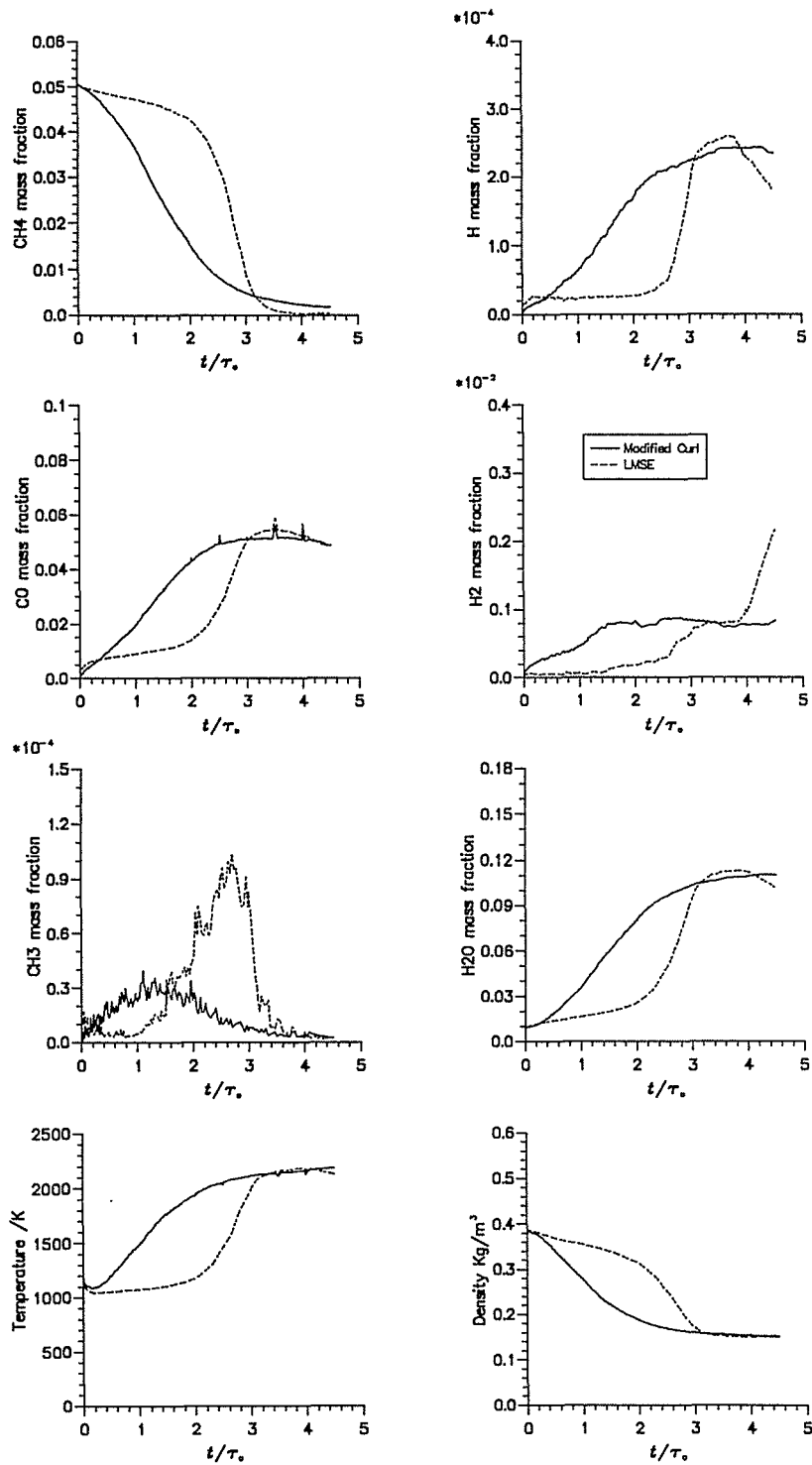


Figure 6.32: Predictions of the density-weighted mean reactive scalar properties in homogeneous, isotropic turbulence. Modified Curl and LMSE are compared starting with a continuous distribution taken from figure 6.27 at $t/\tau_o = 5$, where $\tau_o = 0.04\text{s}$. The reduced scheme of Chelliah et al. [10] is used *without* the interpolation table. $\tilde{\xi} = \xi_{st} = 0.05$ (for CH_4).

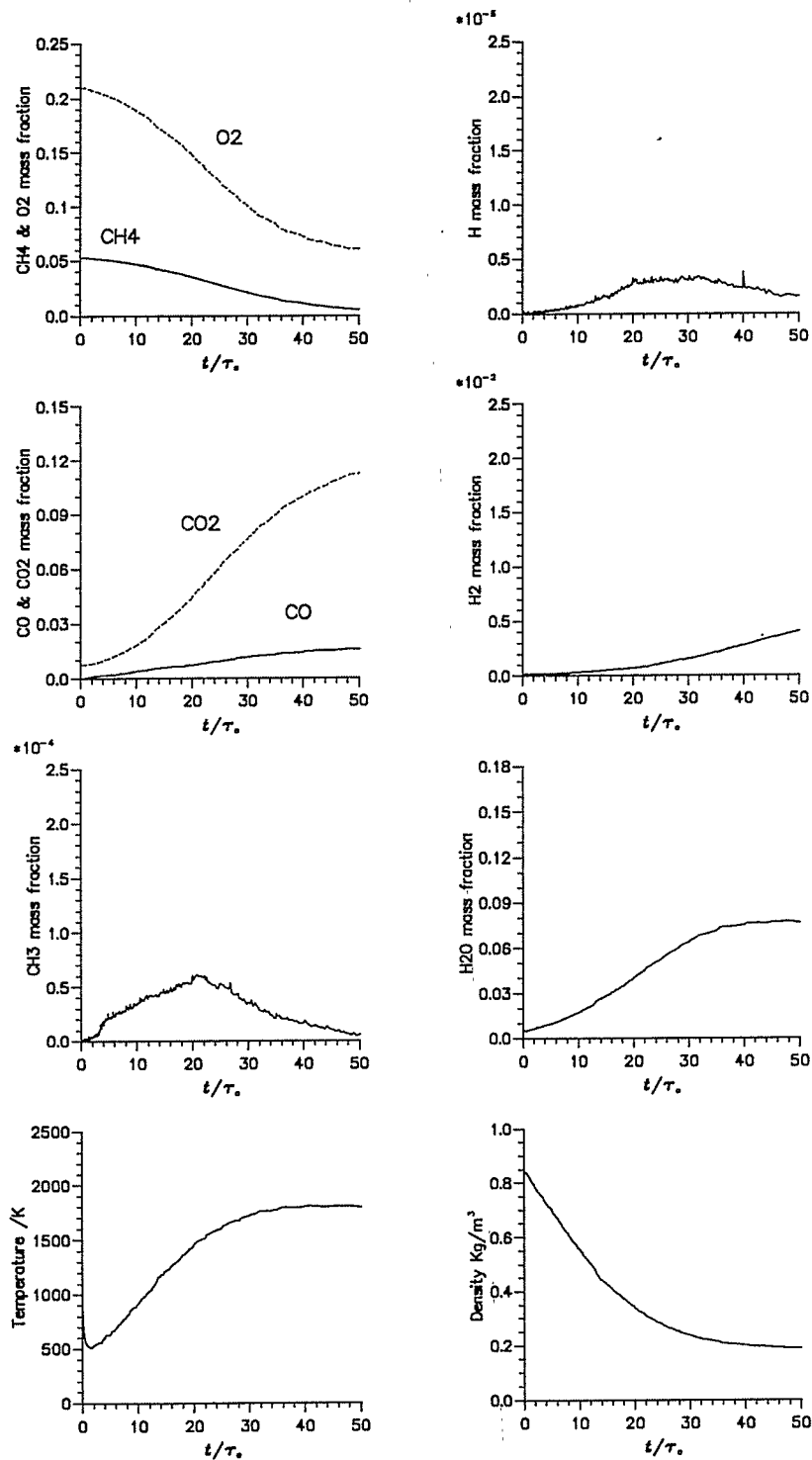


Figure 6.33: Predictions of the density-weighted mean reactive scalar properties in homogeneous, isotropic turbulence. Same conditions are used as in figure 6.30 except for the air temperature which is now 298K instead of 800K. LSODE is employed.

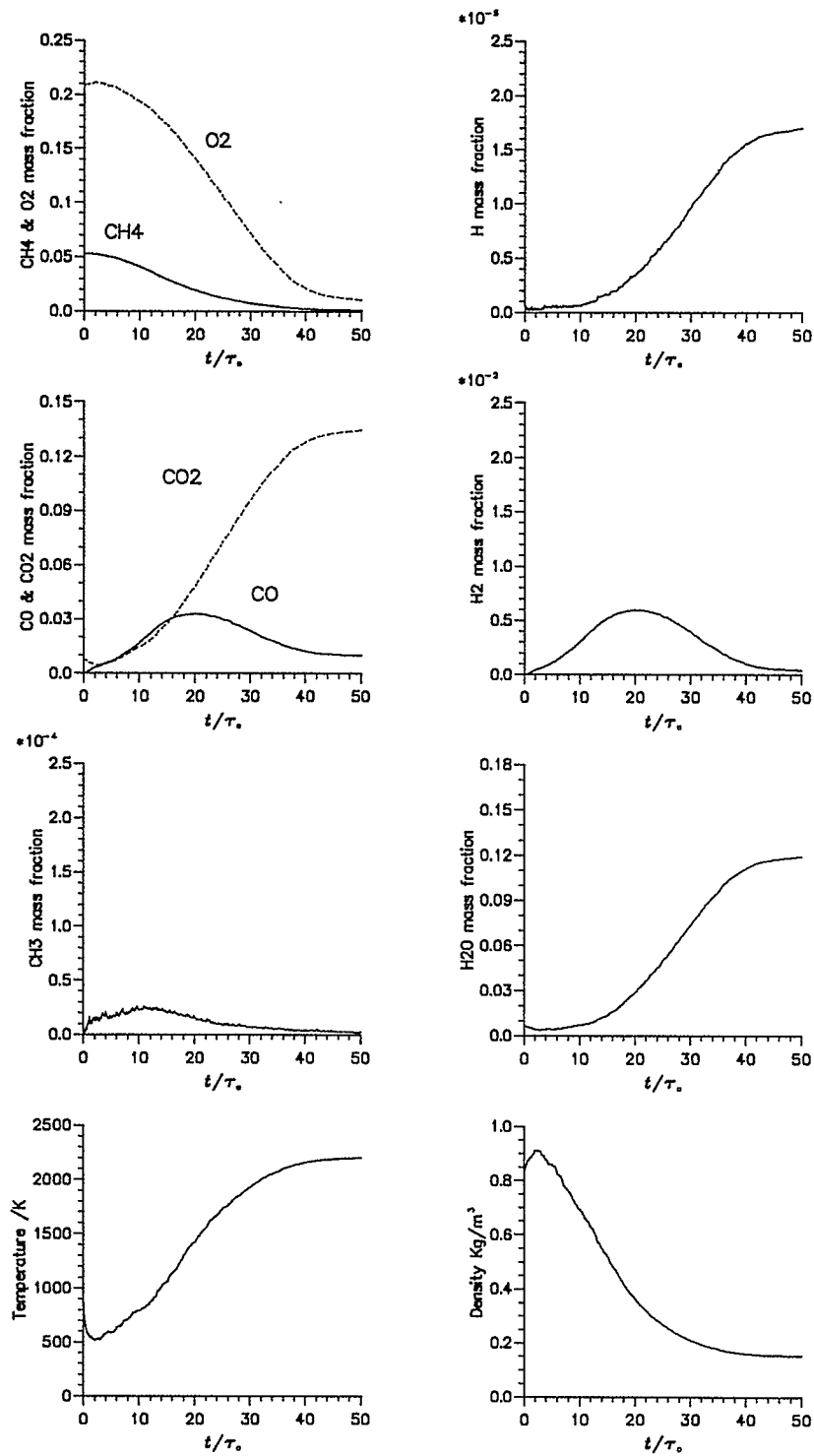


Figure 6.34: Predictions of the density-weighted mean reactive scalar properties in homogeneous, isotropic turbulence. Same conditions are used as in figure 6.30 except for the air temperature which is now 298K instead of 800K. The interpolation table is employed.

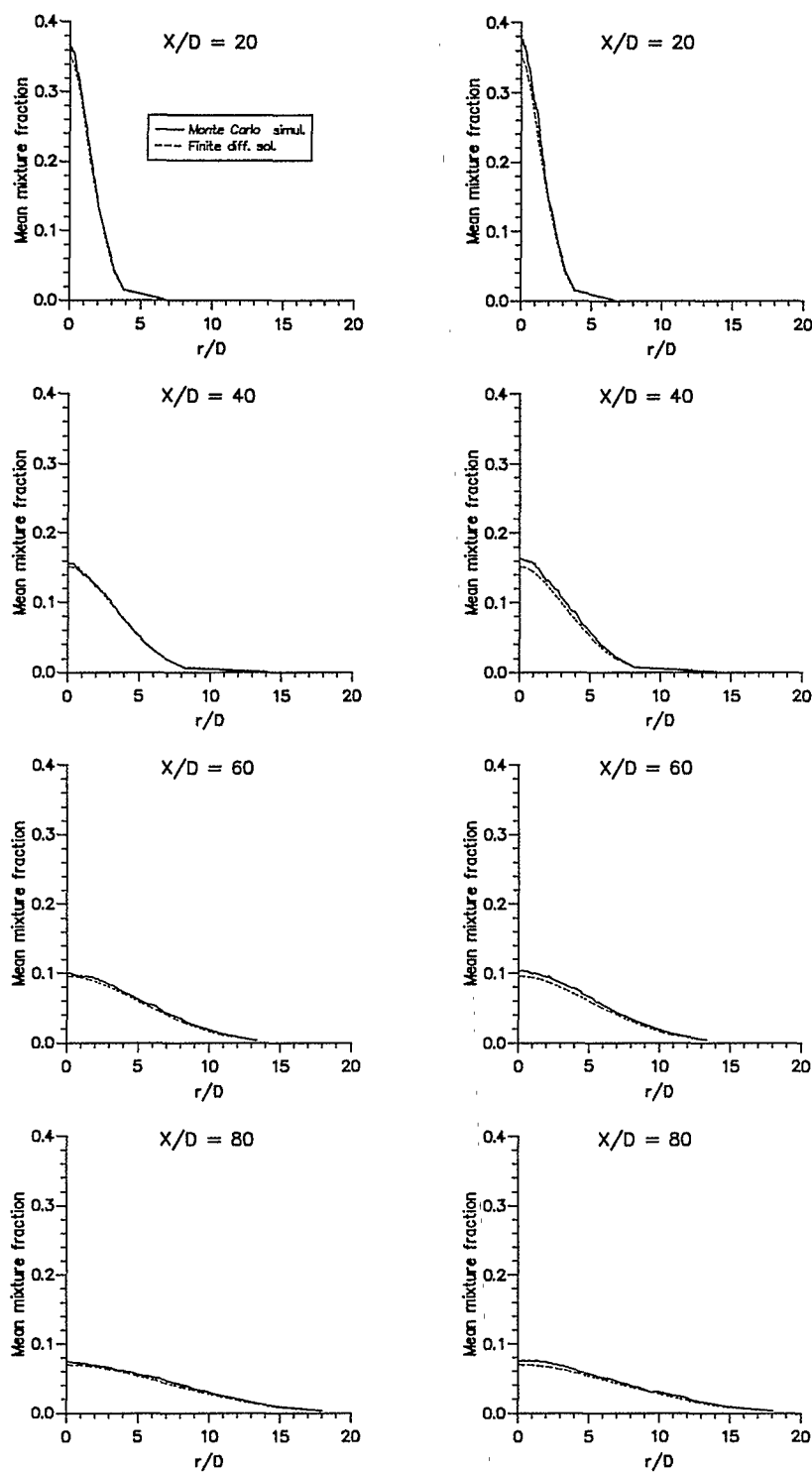


Figure 6.35: Predictions of $\bar{\xi}$ in a non-reacting, constant density air-jet. The jet pipe diameter and mean velocity are 6mm and 19.9 m s^{-1} respectively. A comparison of the finite-difference and Monte Carlo simulation is shown, using the $k-\varepsilon$ turbulence model. The plots on the left employ modified Curl (for the Monte Carlo simulation) and LMSE on the right.

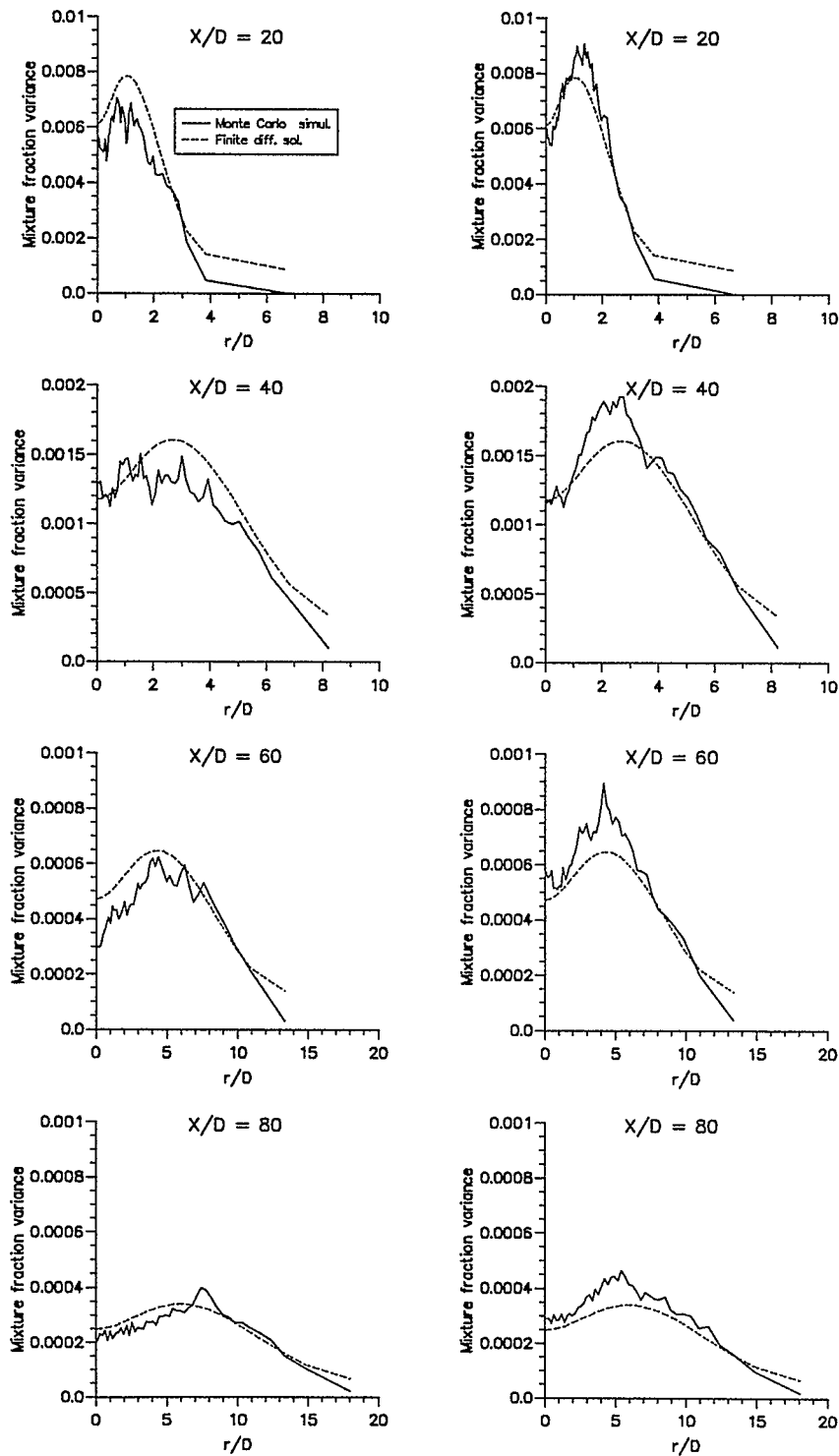


Figure 6.36: Predictions of $\overline{\xi'^2}$ in a non-reacting, constant density air-jet. The jet pipe diameter and mean velocity are 6mm and 19.9 m s^{-1} respectively. A comparison of the finite-difference and Monte Carlo simulation is shown, using the $k-\varepsilon$ turbulence model. The plots on the left employ modified Curl (for the Monte Carlo simulation) and LMSE on the right.

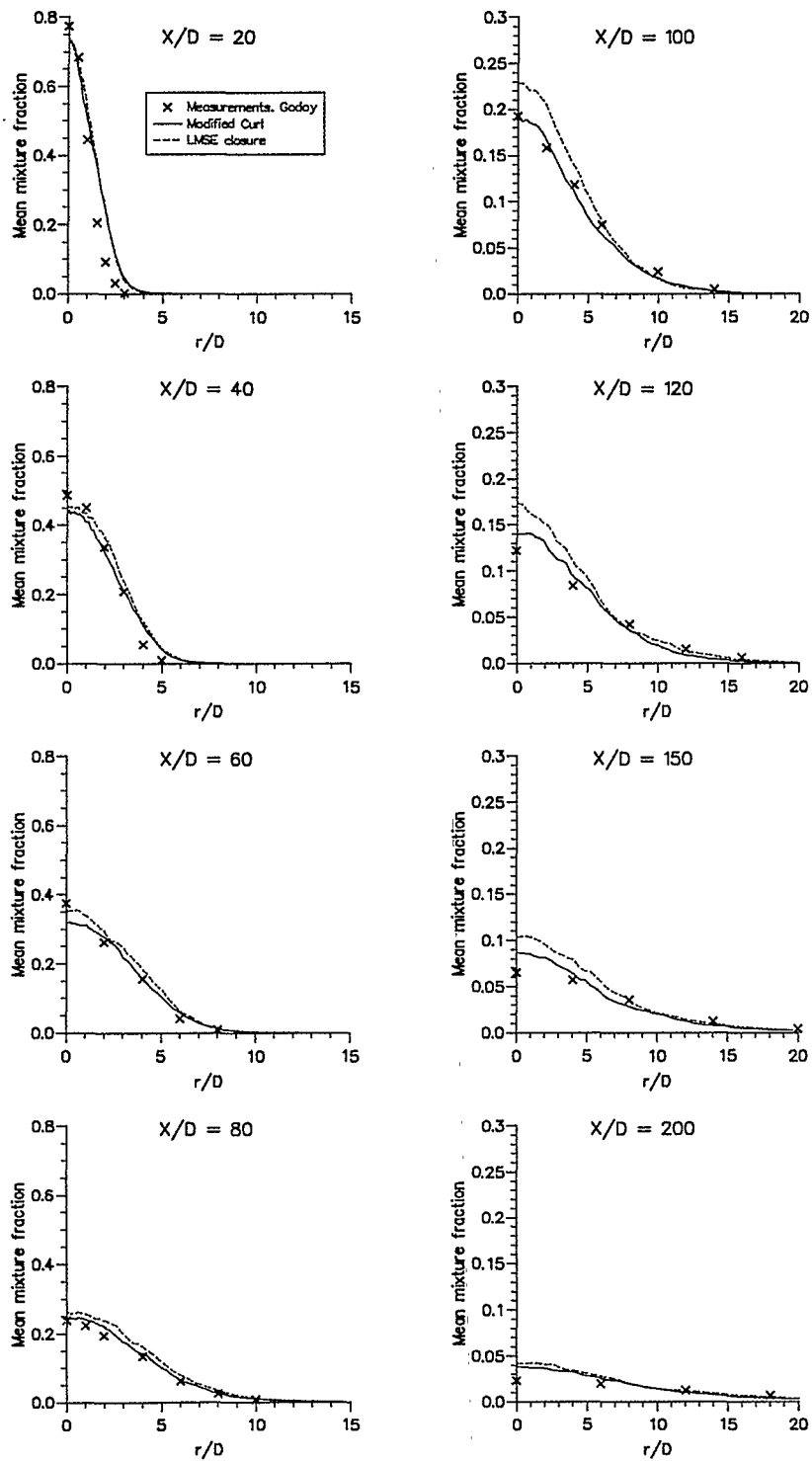


Figure 6.37: Radial profiles of $\bar{\xi}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\varepsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed.

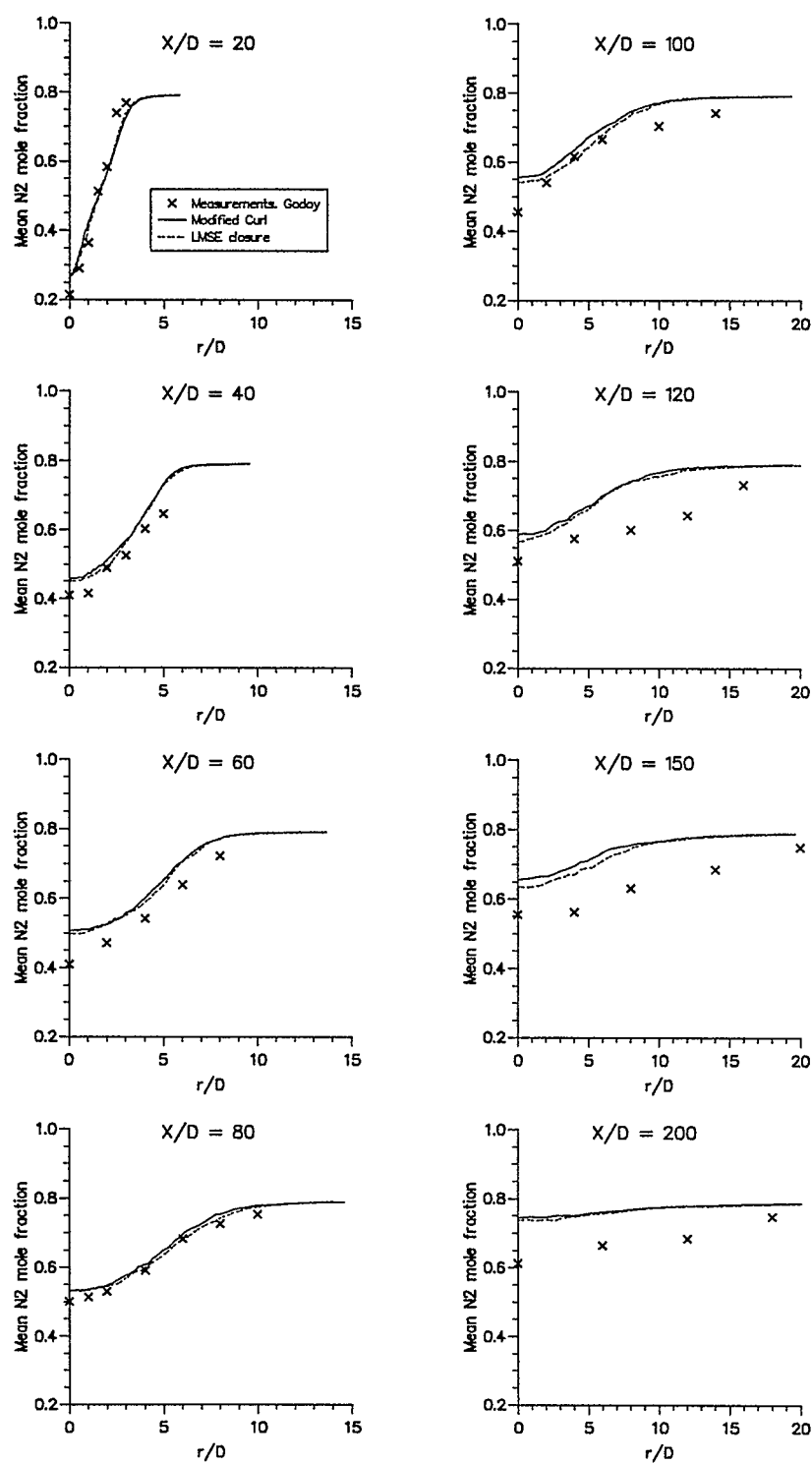


Figure 6.38: Radial profiles of $\tilde{X}_{N_2}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\epsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed.

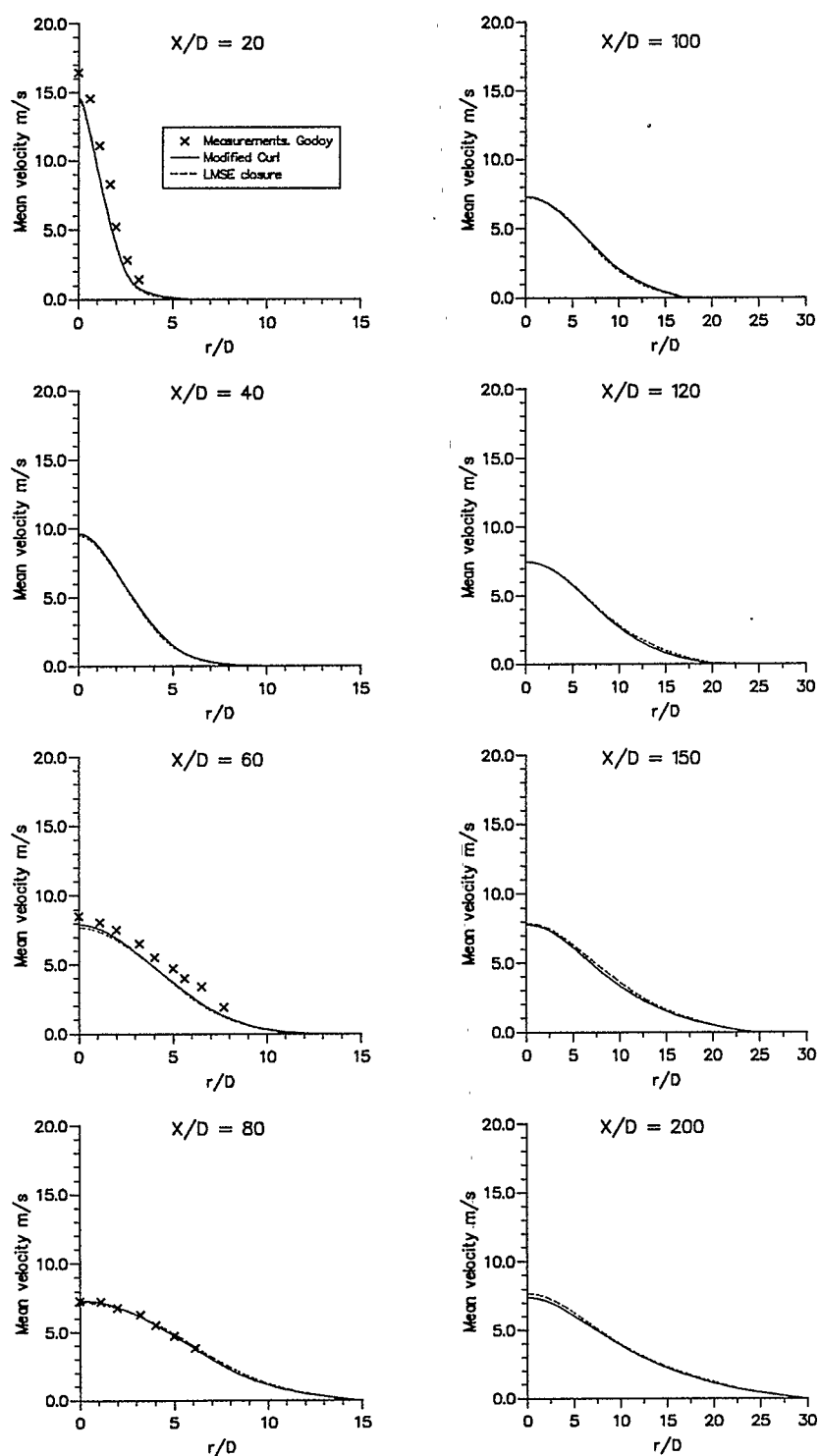


Figure 6.39: Radial profiles of $\bar{u}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\varepsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed.

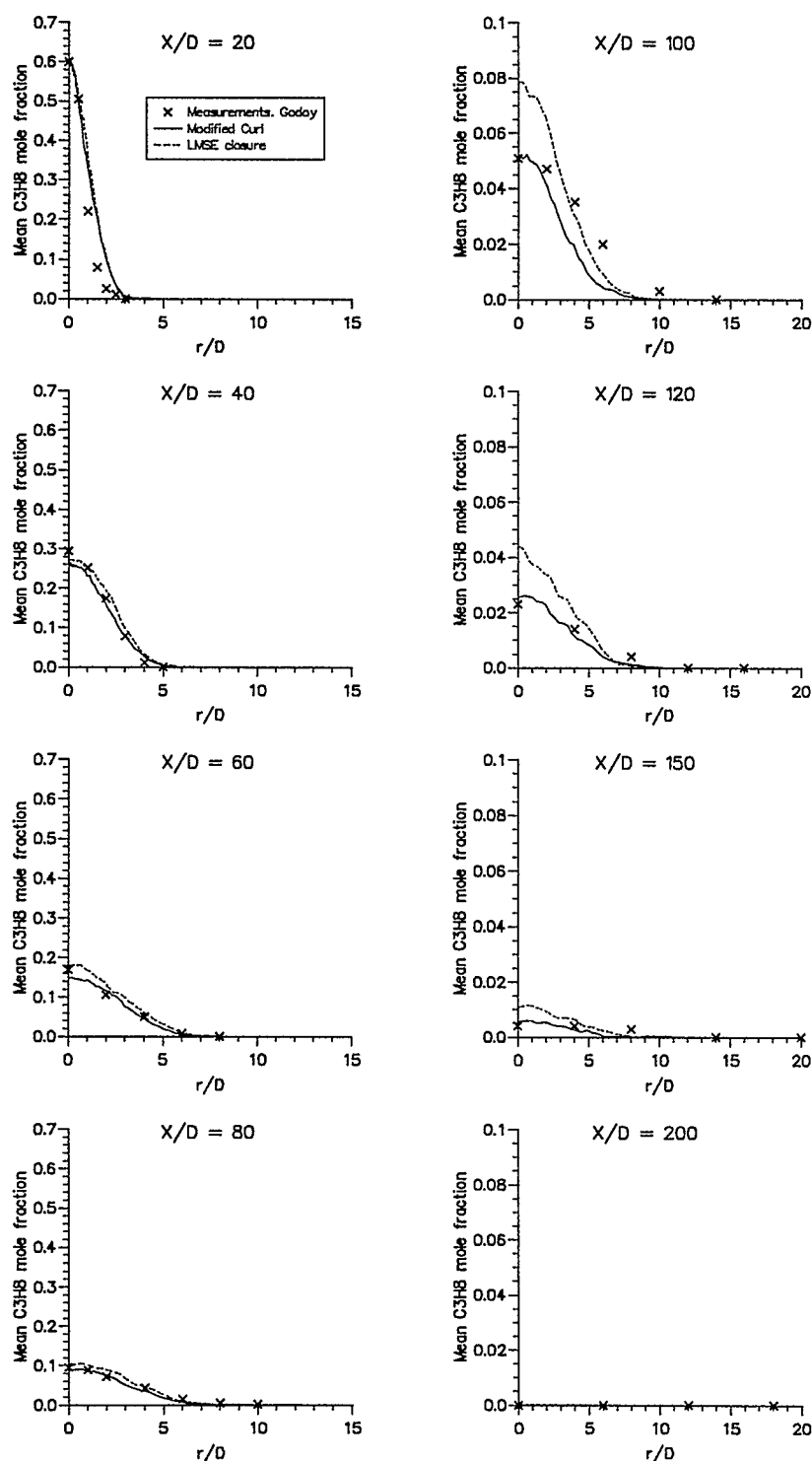


Figure 6.40: Radial profiles of $\tilde{X}_{\text{C}_3\text{H}_8}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\epsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed.

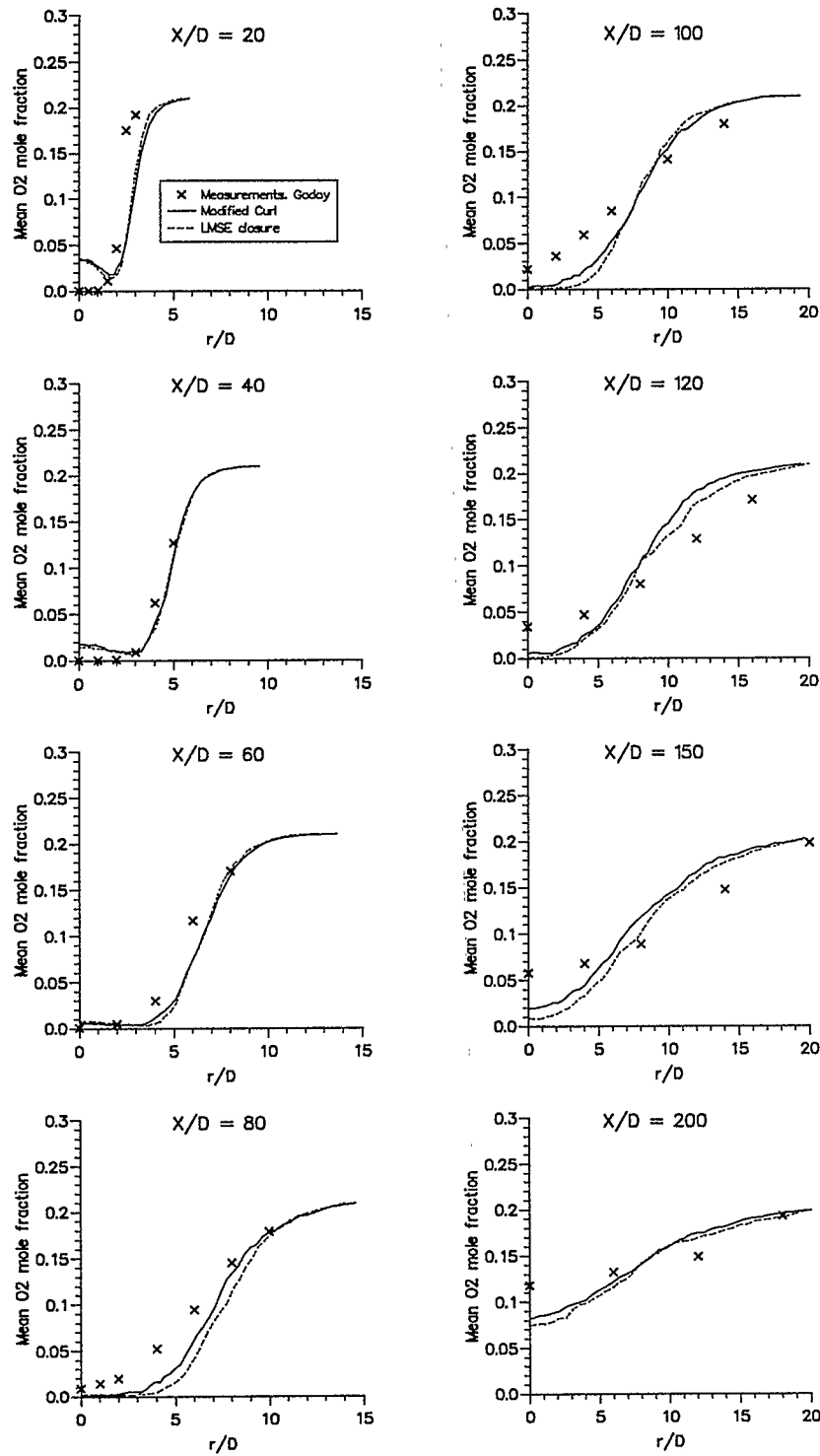


Figure 6.41: Radial profiles of $\bar{X}_{O_2}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\epsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed.

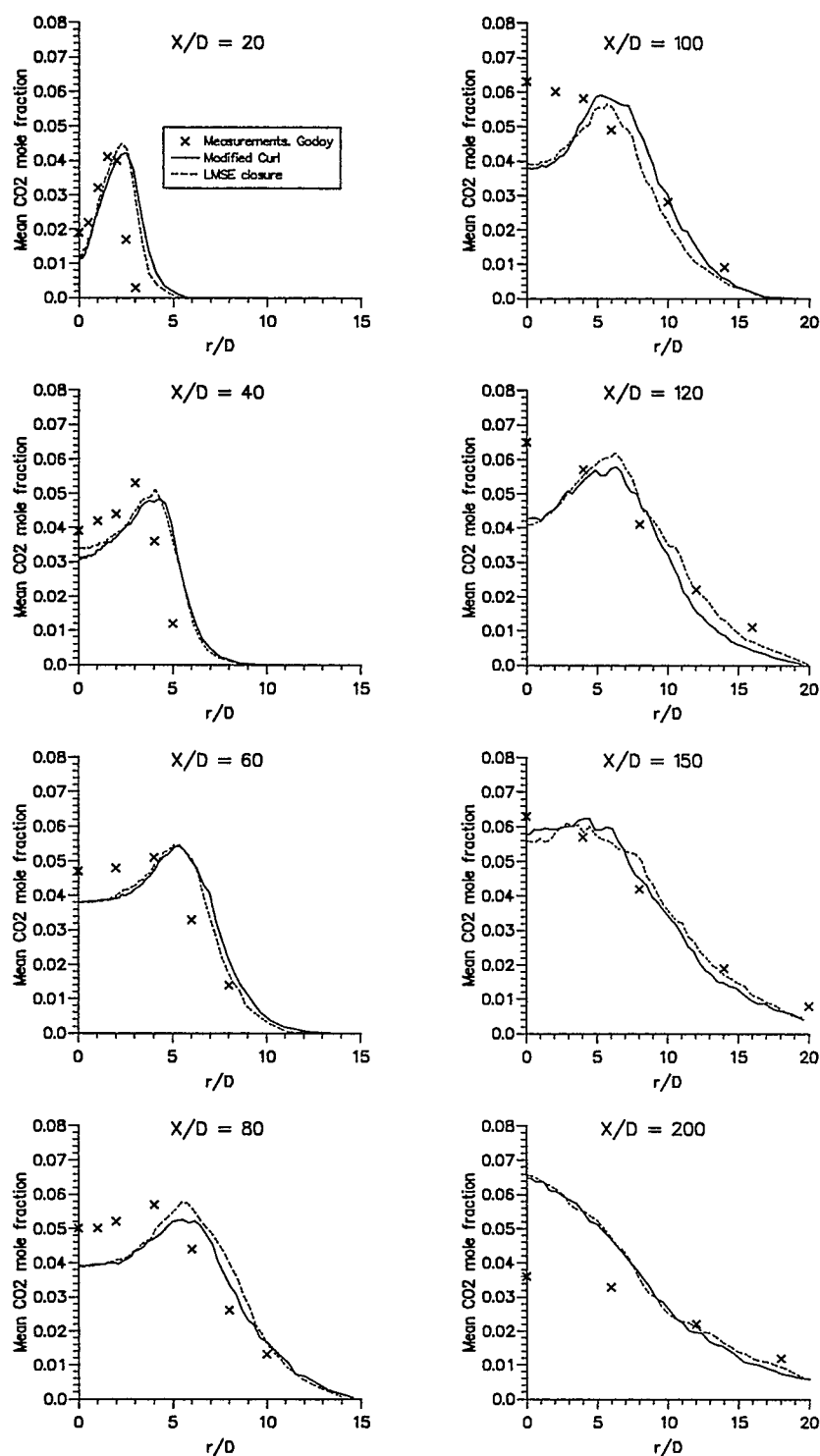


Figure 6.42: Radial profiles of $\bar{X}_{\text{CO}_2}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\varepsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed.

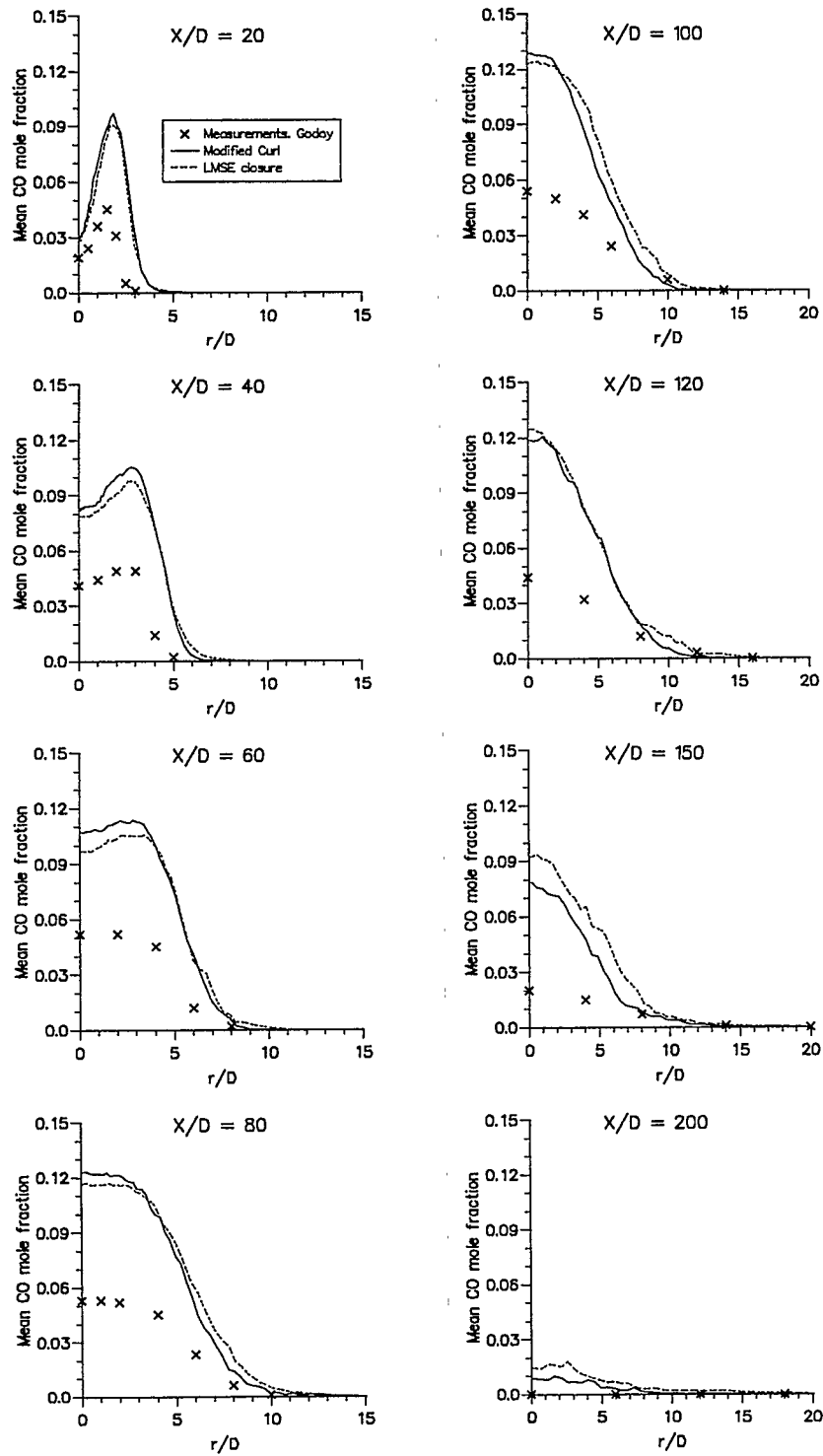


Figure 6.43: Radial profiles of $\bar{X}_{CO}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\varepsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed.

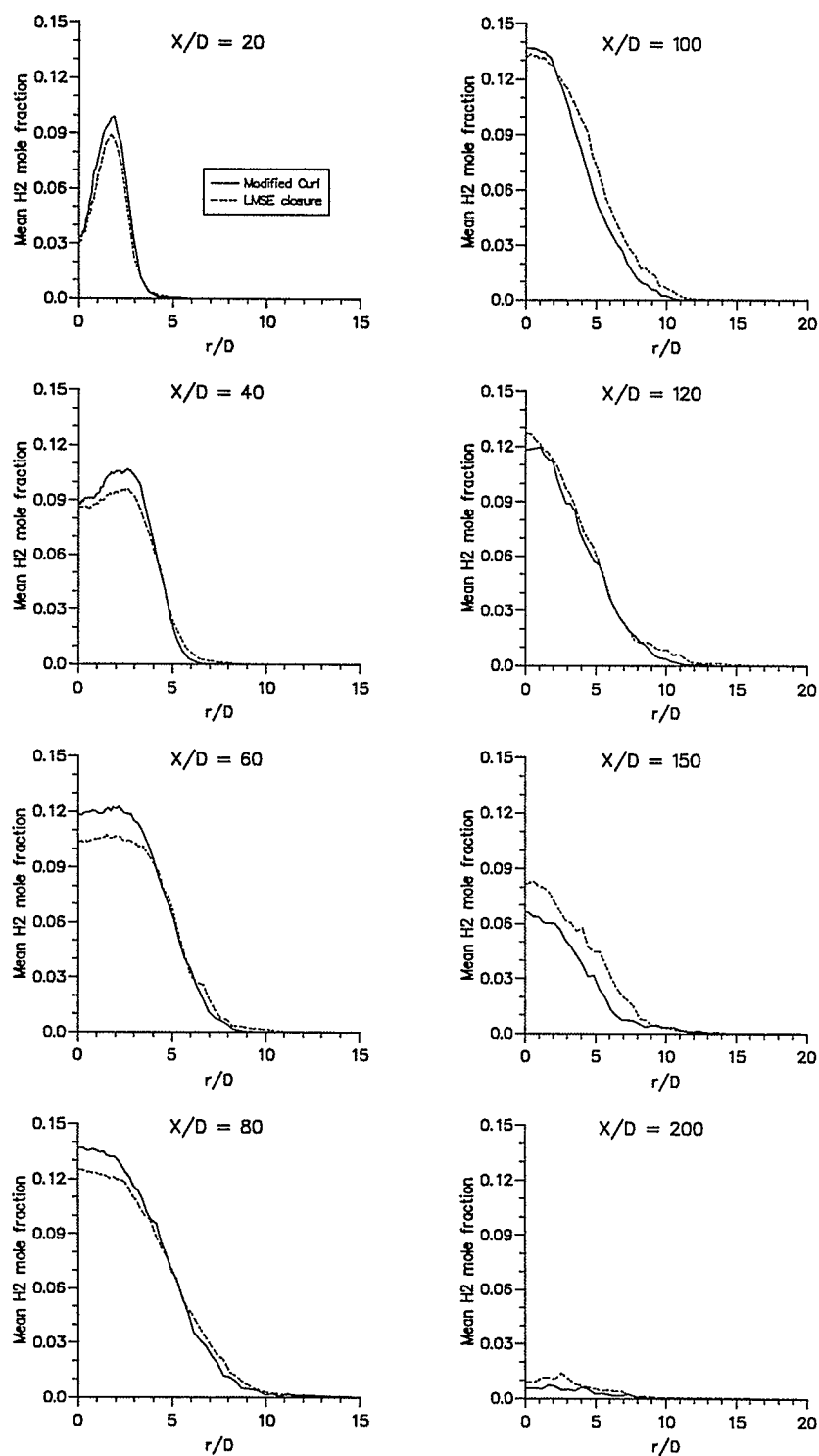


Figure 6.44: Radial profiles of $\tilde{X}_{H_2}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\varepsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed.

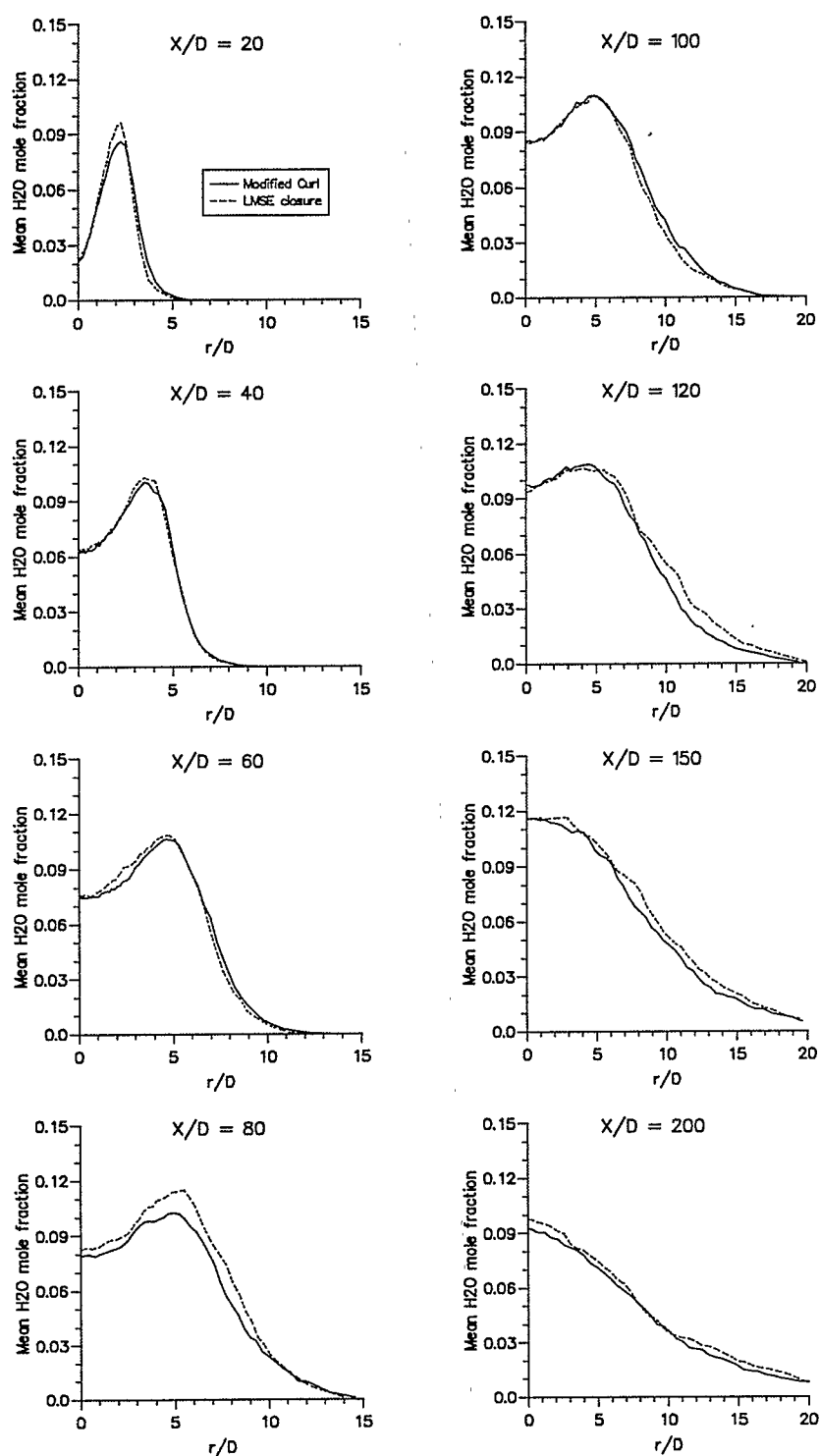


Figure 6.45: Radial profiles of $\bar{X}_{\text{H}_2\text{O}}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\varepsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed.

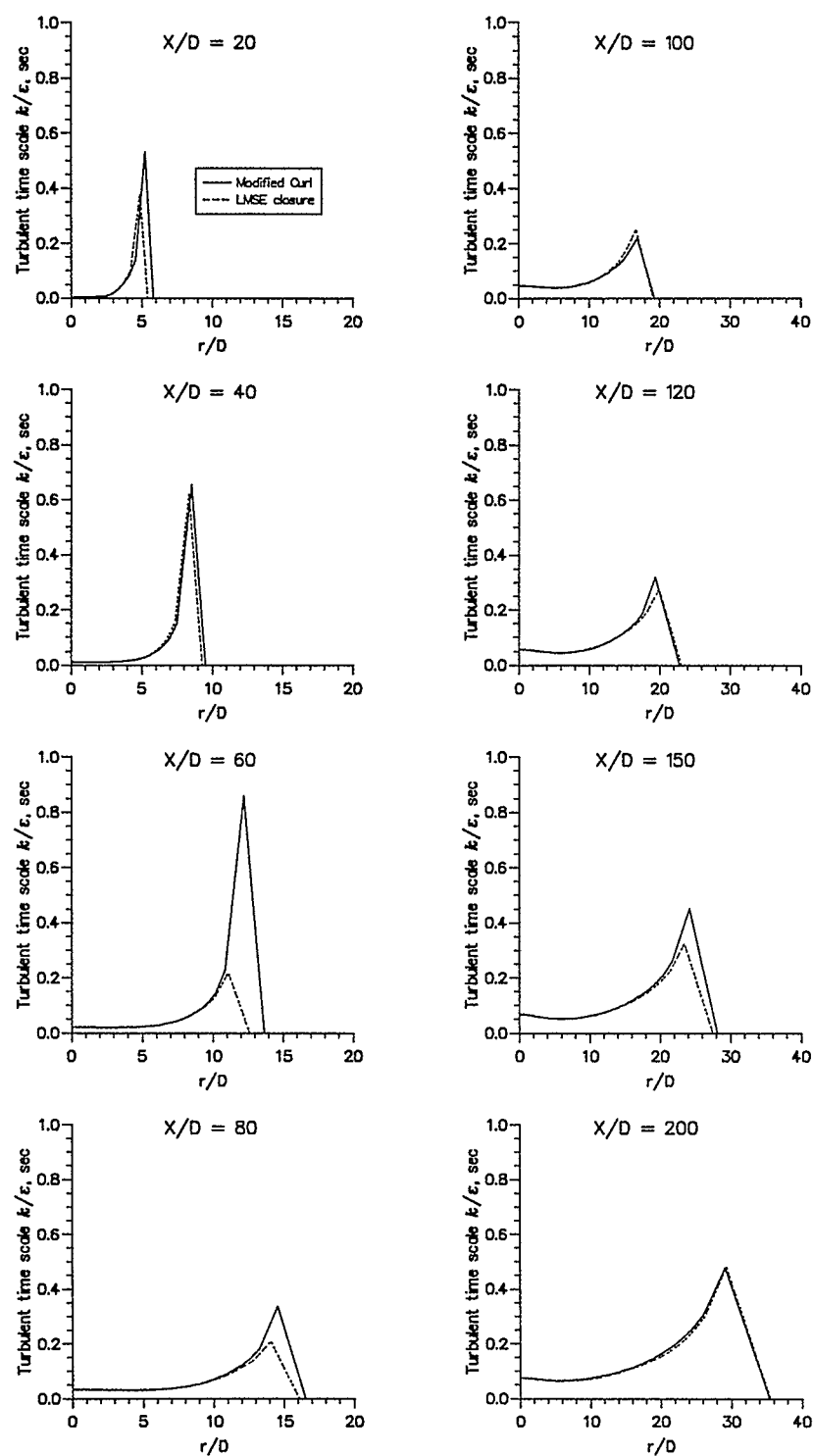


Figure 6.46: Radial profiles of $\frac{k}{\epsilon}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\epsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed.

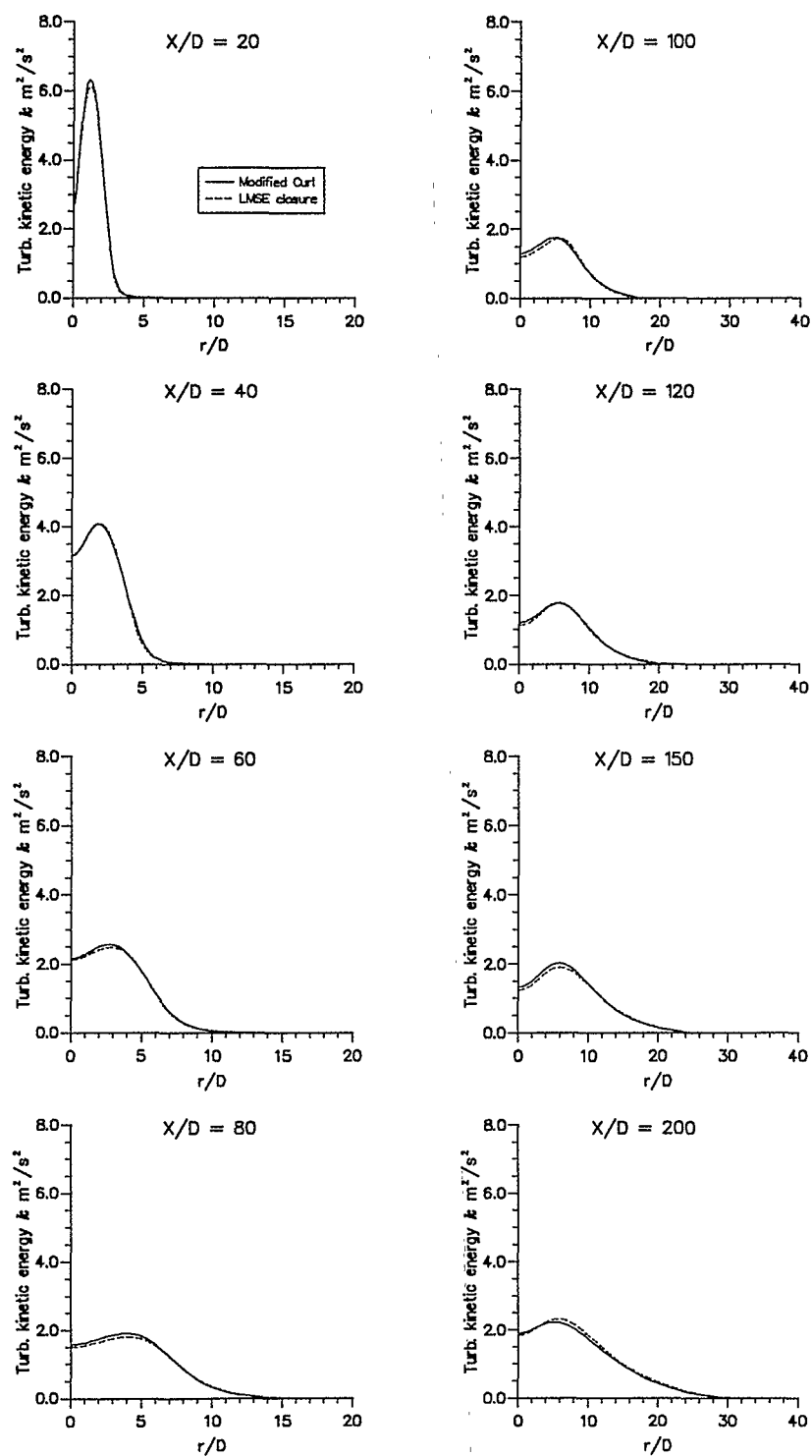


Figure 6.47: Radial profiles of the turbulence kinetic energy (k) for the flame of Godoy [31] at $Re = 24000$. The k - ε turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed.

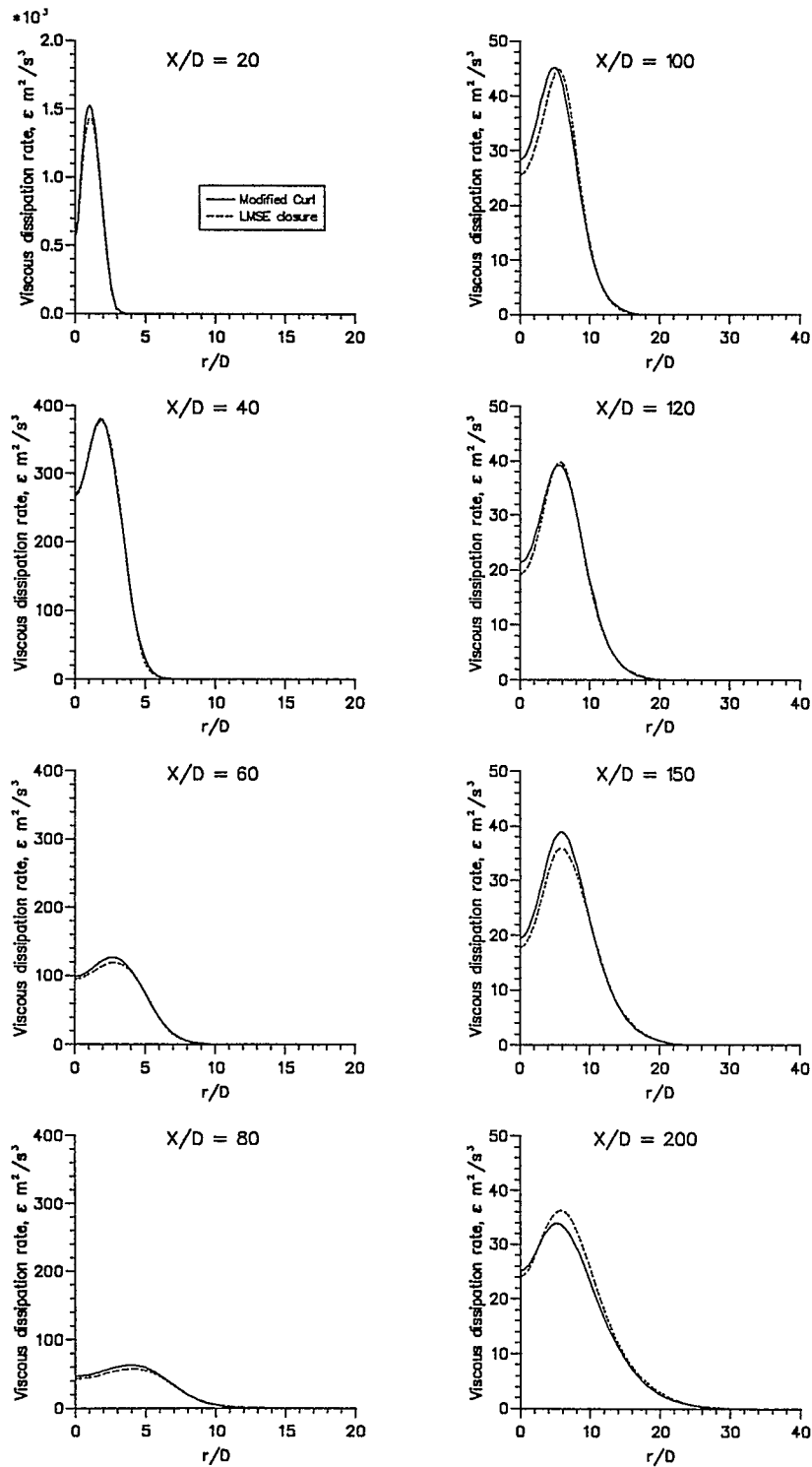


Figure 6.48: Radial profiles of the viscous dissipation rate of the turbulence kinetic energy (ε) for the flame of Godoy [31] at $Re = 24000$. The k - ε turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed.

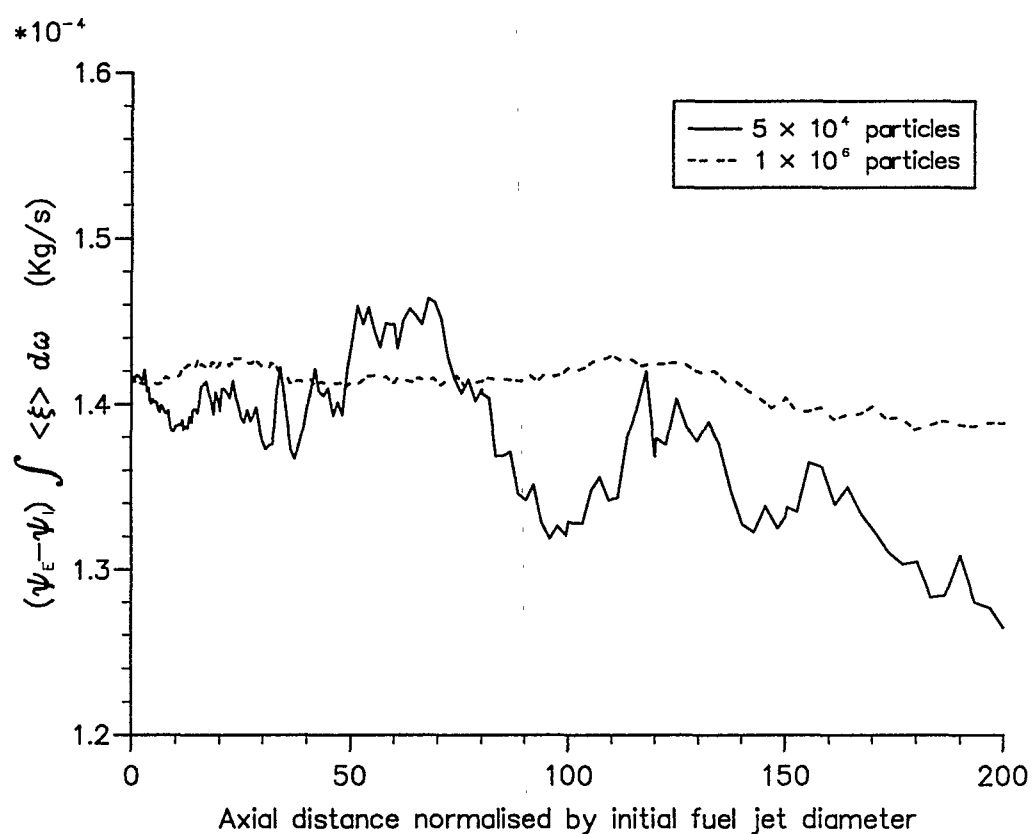


Figure 6.49: This figure shows the axial evolution of the integral of the mixture fraction convection as defined in equation (6.17), for the flame of Godoy [31] at $Re = 24000$. The comparison of the integral with 5×10^4 and 1×10^6 particles is shown. The $k-\varepsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. The result is only shown for modified Curl.

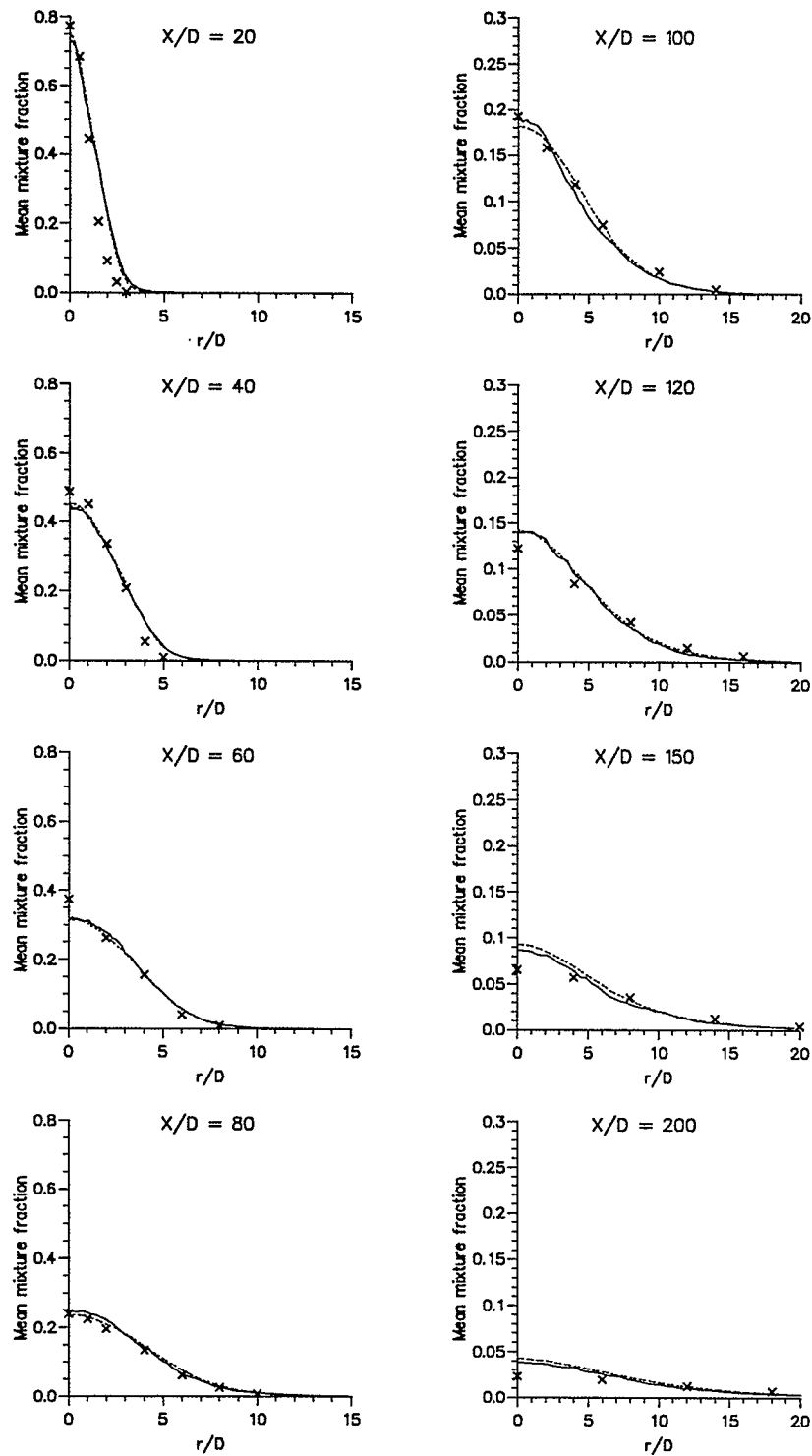


Figure 6.50: Radial profiles of $\tilde{\xi}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\epsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. Calculations are reported for modified Curl using 5×10^4 (solid line) and 1×10^6 particles (dashed line).

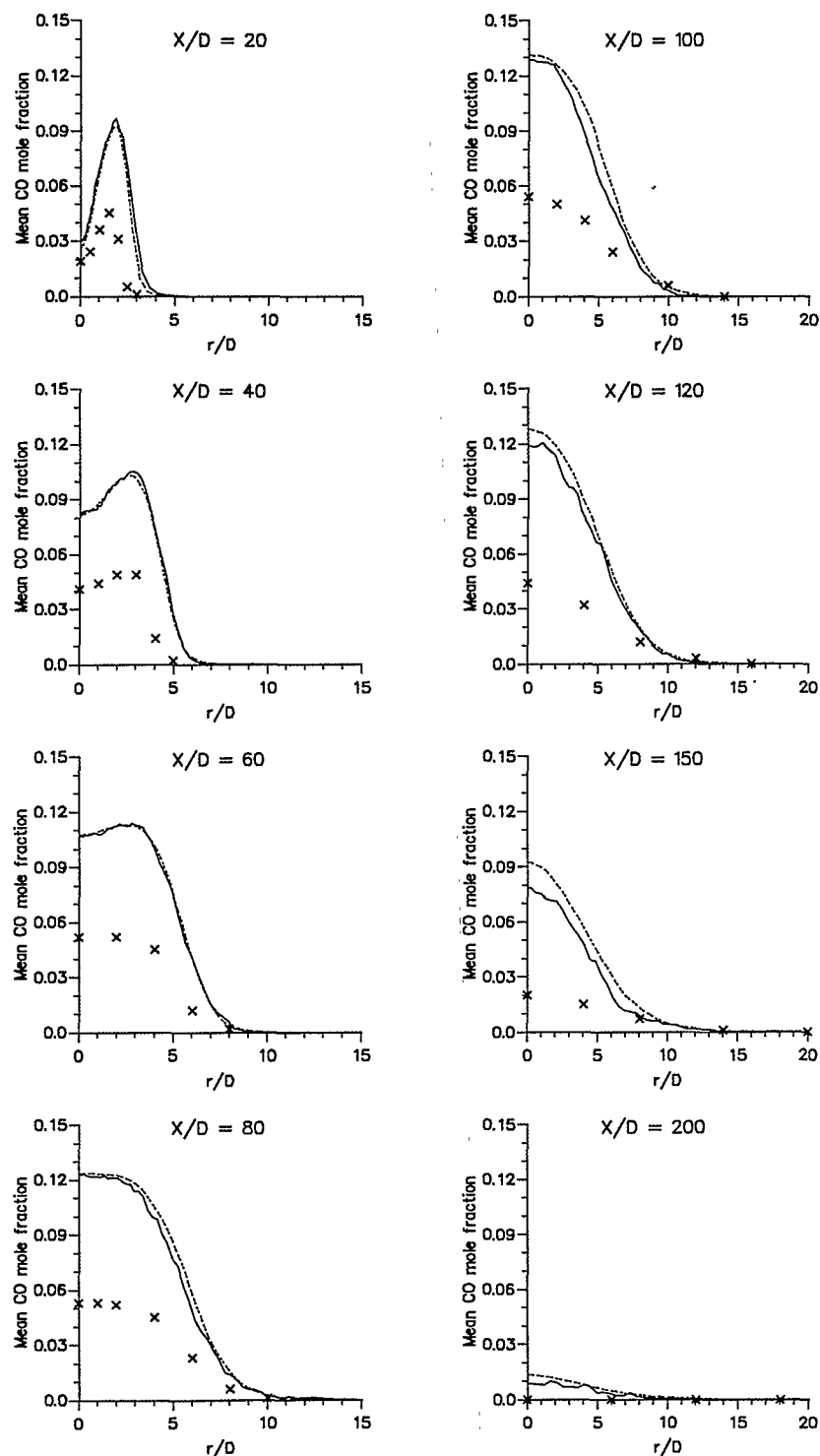


Figure 6.51: Radial profiles of $\bar{X}_{CO}$ for the flame of Godoy [31] at $Re = 24000$. The $k-\varepsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed. Calculations are reported for modified Curl using 5×10^4 (solid line) and 1×10^6 particles (dashed line).

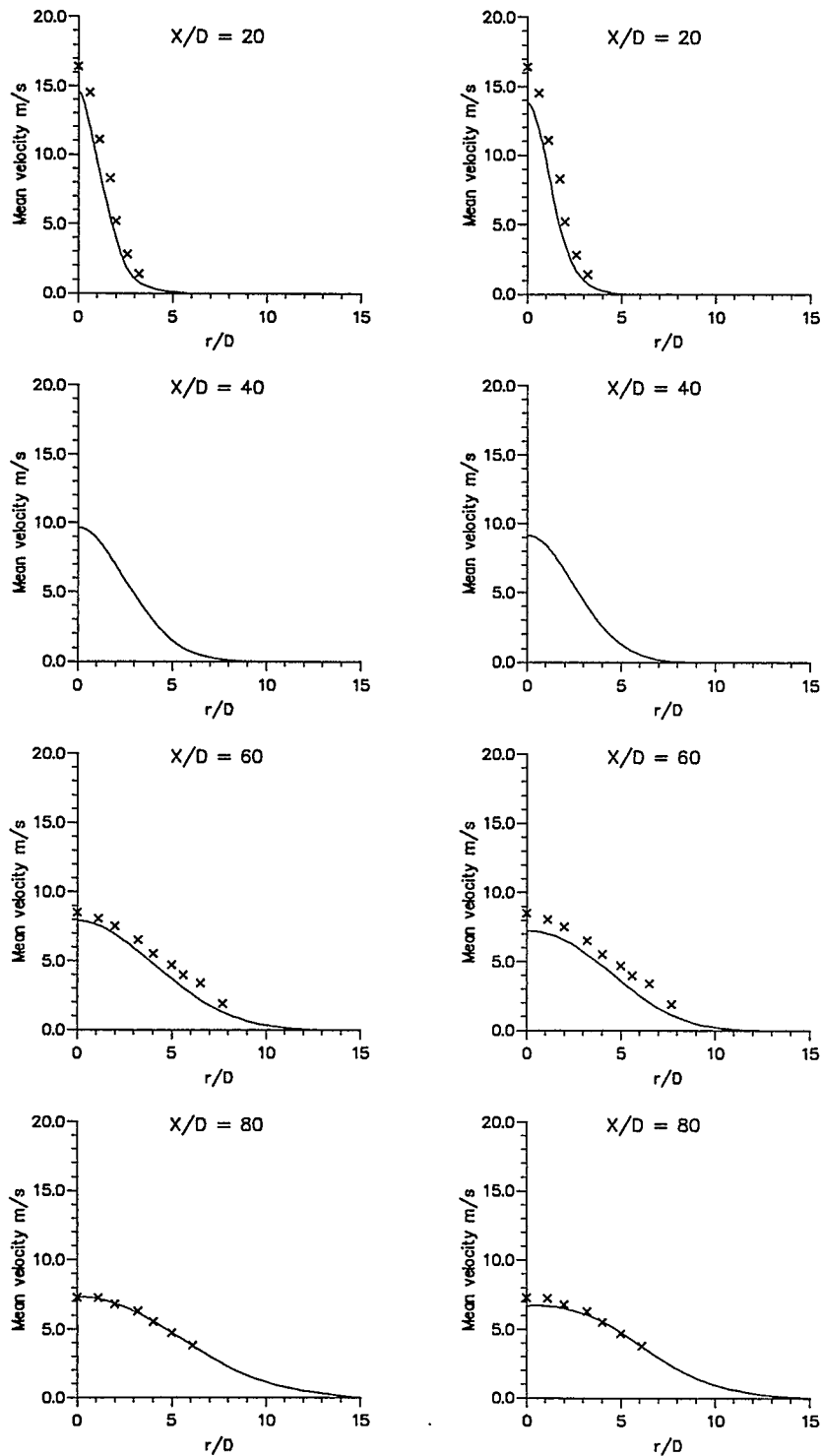


Figure 6.52: Radial profiles of $\bar{u}$ for the flame of Godoy [31] at $Re = 24000$. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. A comparison is made of the $k-\epsilon$ and Reynolds stress turbulence models on the left and right hand sides, respectively.

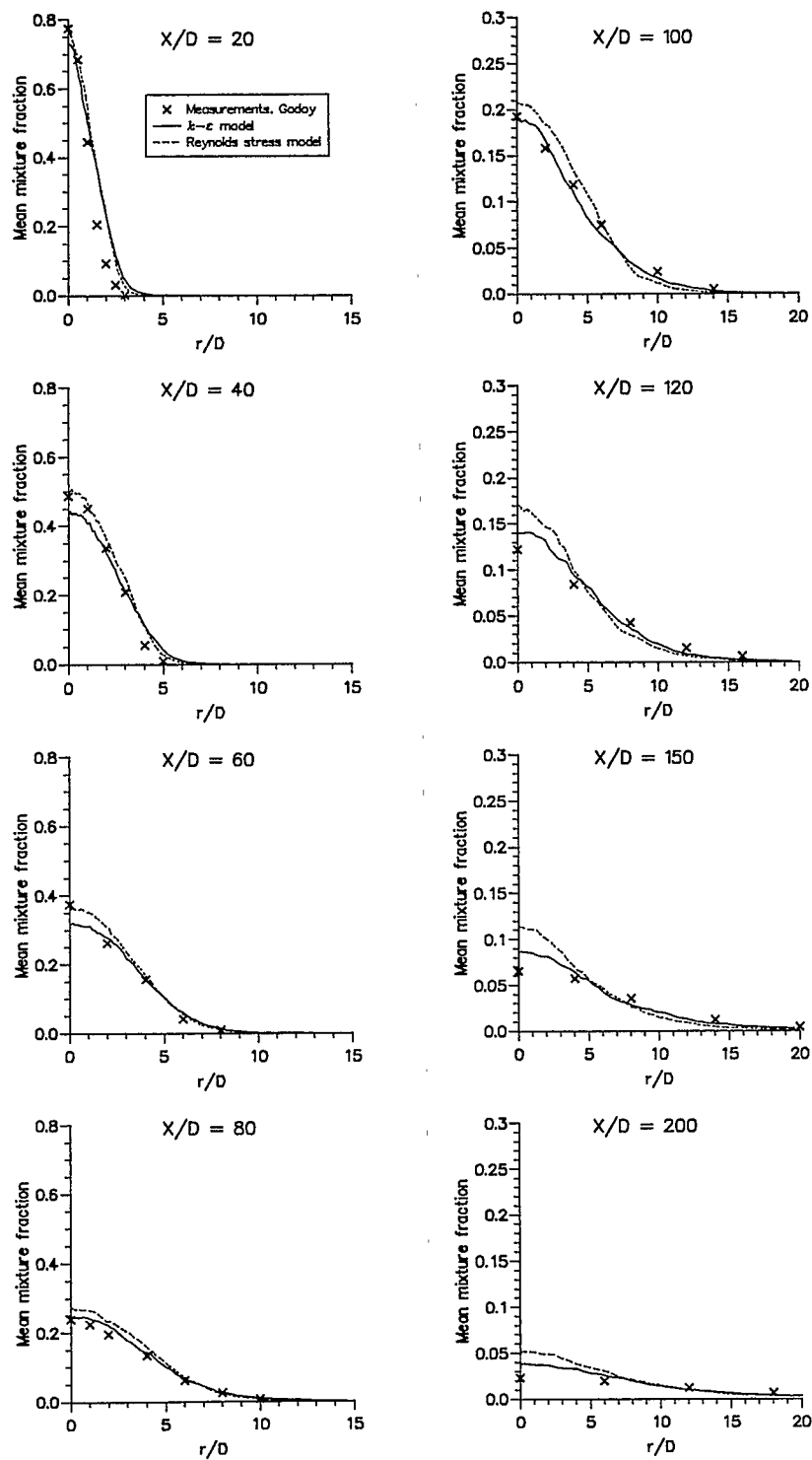


Figure 6.53: Radial profiles of $\tilde{\xi}$ for the flame of Godoy [31] at $Re = 24000$. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. A comparison is made of the k - ε and Reynolds stress turbulence models.

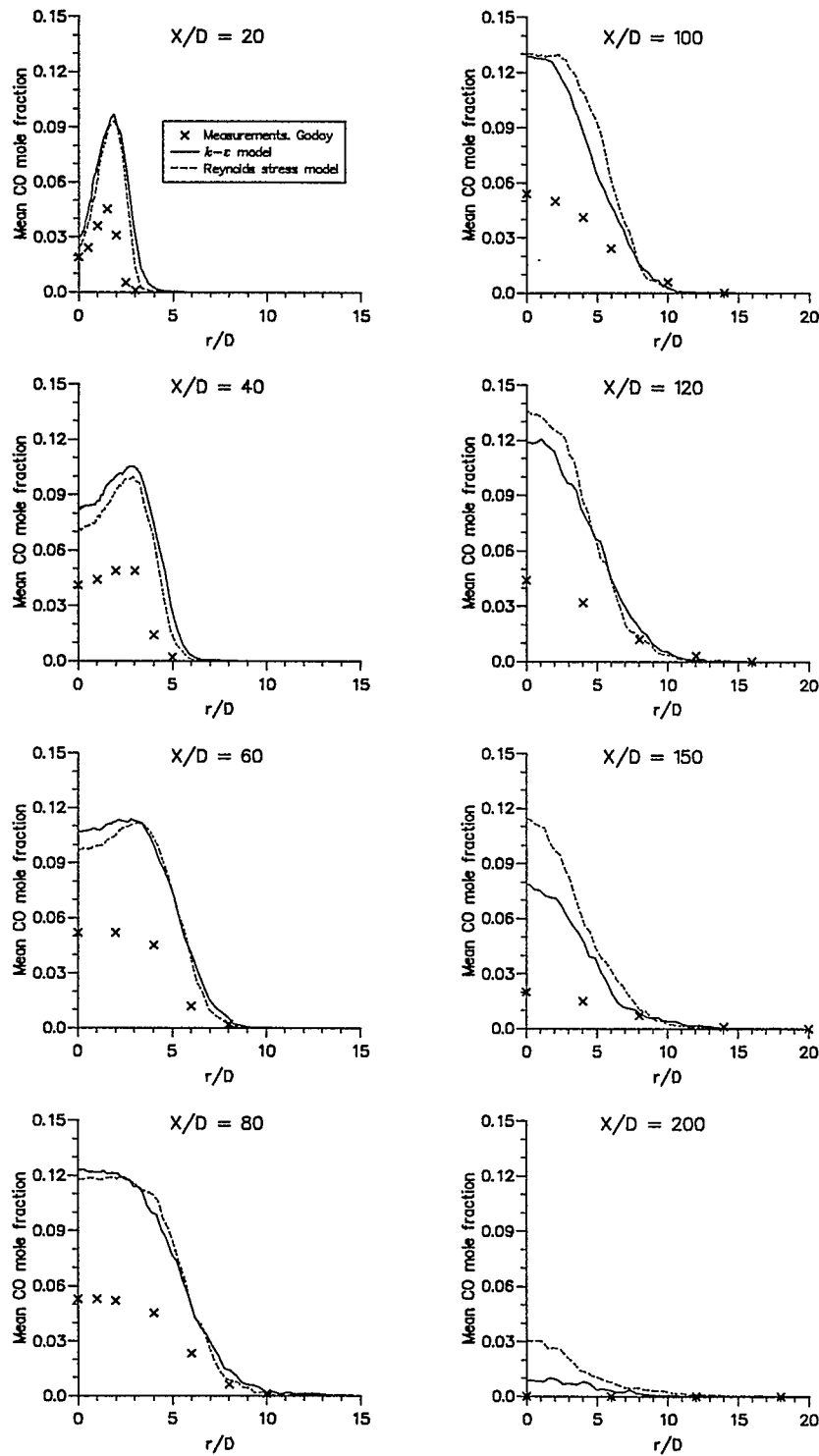


Figure 6.54: Radial profiles of $\bar{X}_{CO}$ for the flame of Godoy [31] at $Re = 24000$. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. A comparison is made of the $k-\epsilon$ and Reynolds stress turbulence models.

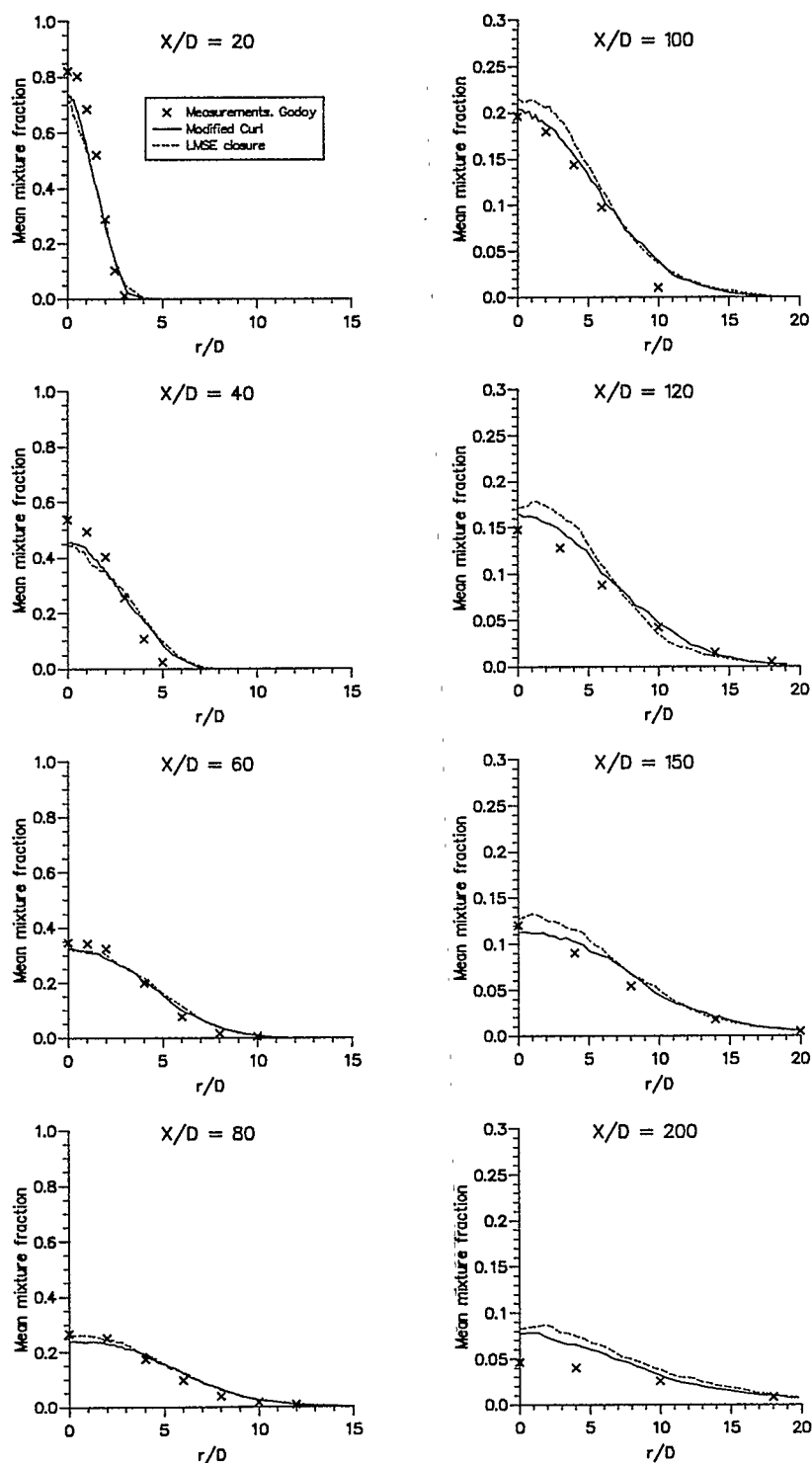


Figure 6.55: Radial profiles of $\tilde{\xi}$ for the flame of Godoy [31] at $Re = 42700$. The $k-\varepsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed.

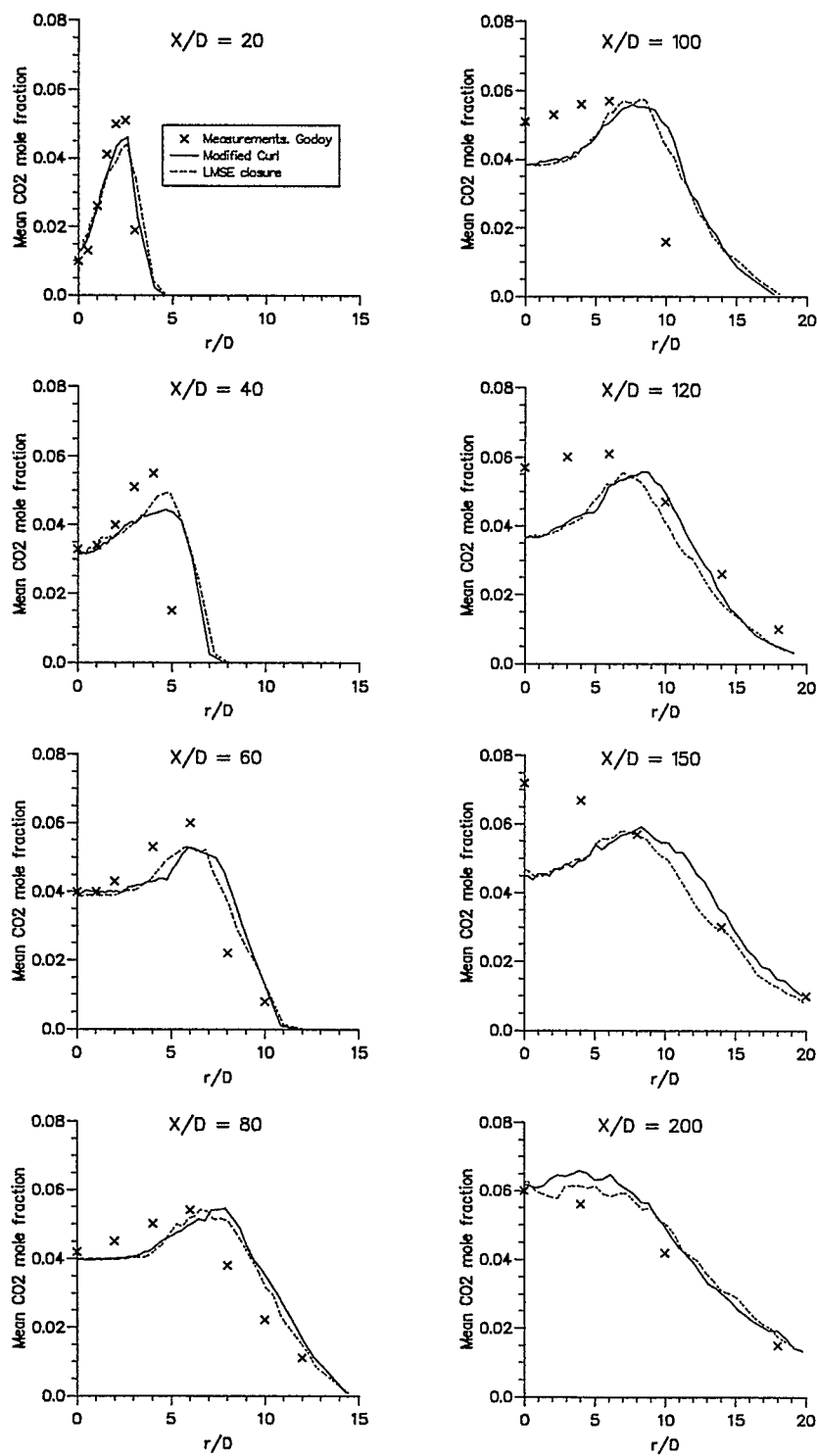


Figure 6.56: Radial profiles of $\bar{X}_{\text{CO}_2}$ for the flame of Godoy [31] at $Re = 42700$. The $k-\epsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed.

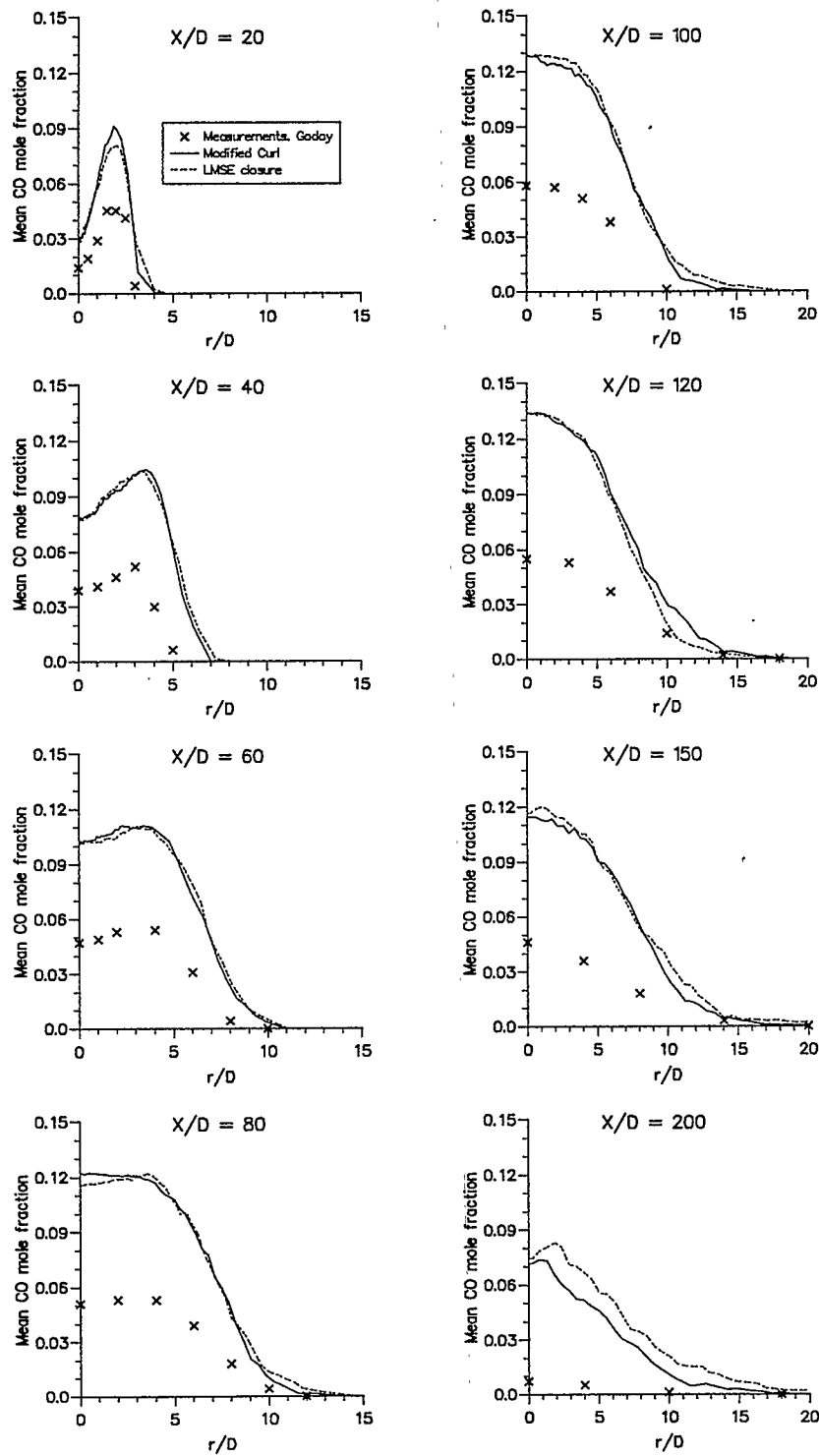


Figure 6.57: Radial profiles of $\bar{X}_{CO}$ for the flame of Godoy [31] at $Re = 42700$. The $k-\varepsilon$ turbulence model and the reduced scheme of Jones and Lindstedt [46] are employed.

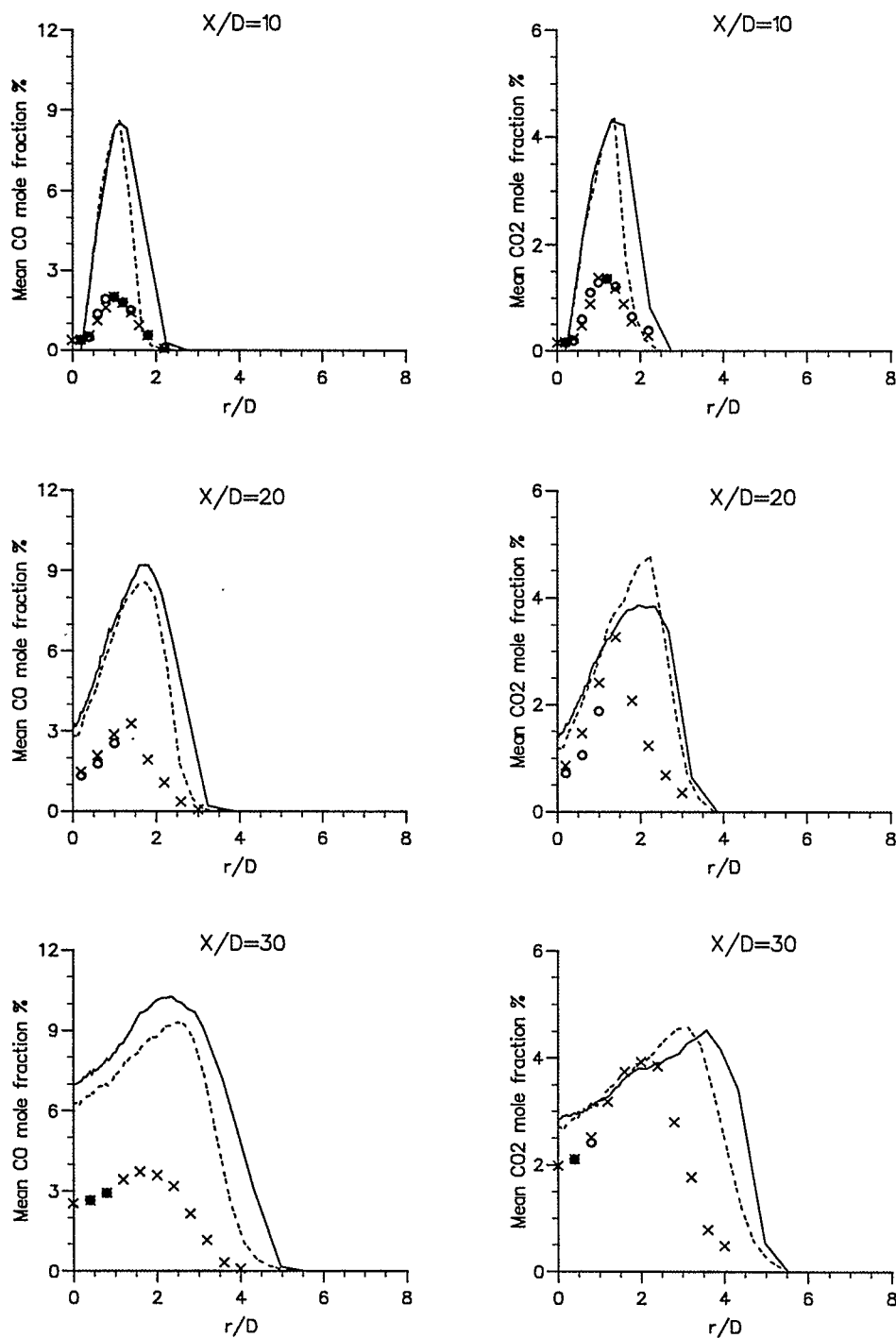


Figure 6.58: Predictions of $\bar{X}_{CO}$ and $\bar{X}_{CO_2}$ for the flame of Heitor et al. [37], burner 2 at $Re = 37000$. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. The $k-\epsilon$ (solid line) and Reynolds stress (dotted line) turbulence models are used. Crosses and circles refer to the measurements at diametrically opposing locations.

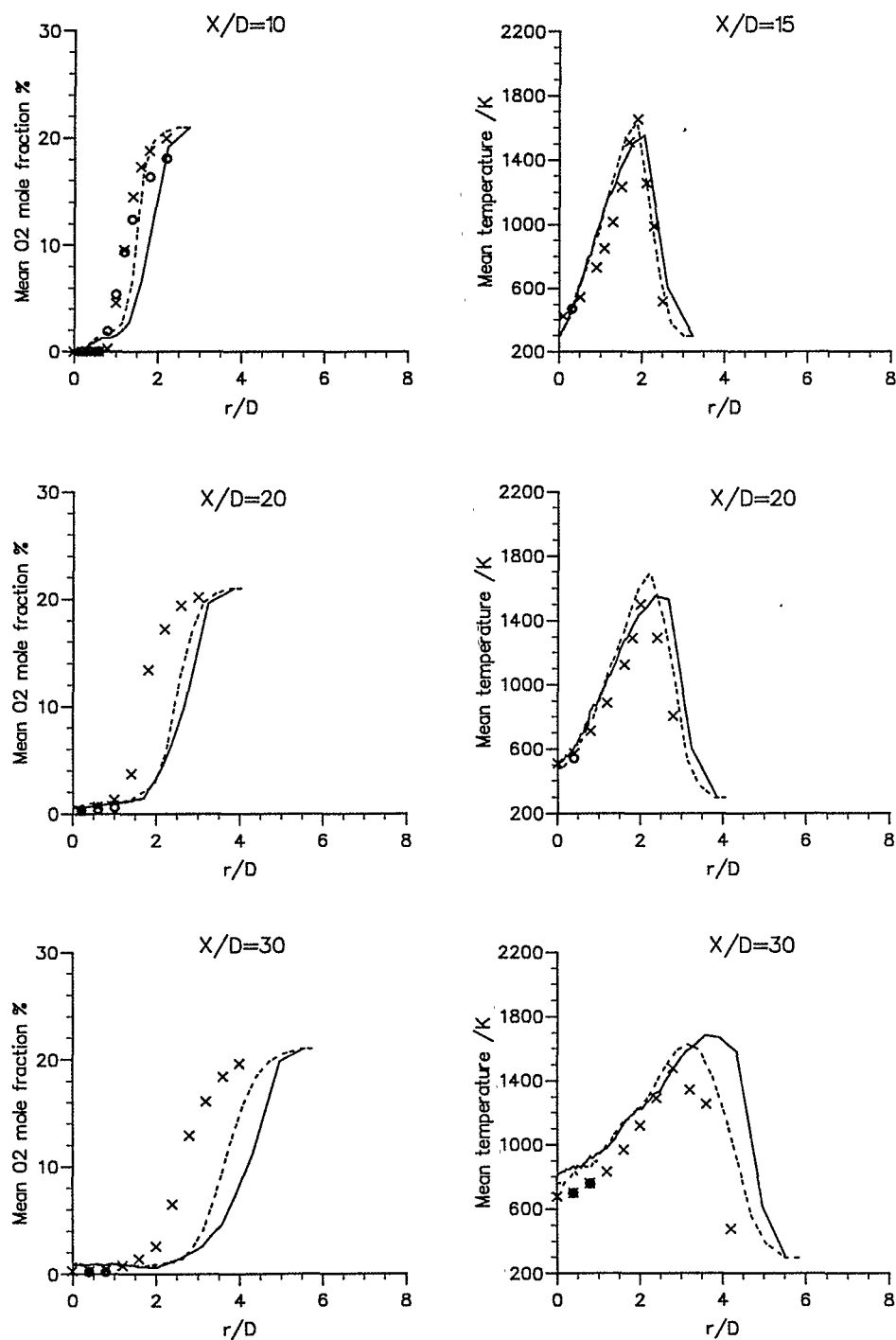


Figure 6.59: Predictions of $\bar{X}_{O_2}$ and $\bar{T}$ for the flame of Heitor et al. [37], burner 2 at $Re = 37000$. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. The $k-\epsilon$ (solid line) and Reynolds stress (dotted line) turbulence models are used. Crosses and circles refer to the measurements at diametrically opposing locations.

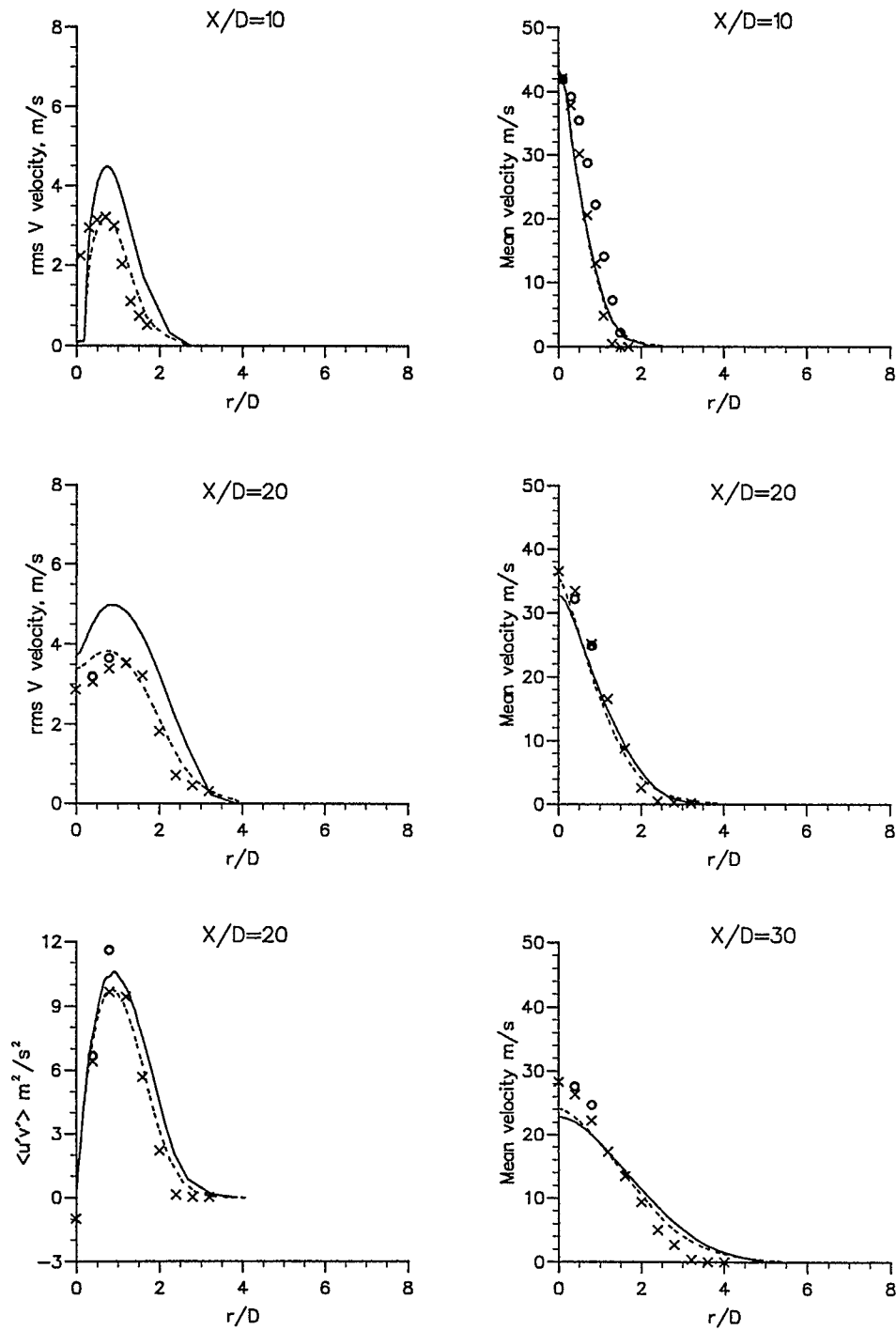


Figure 6.60: Predictions of $(\widetilde{v'^2})^{1/2}$, $\widetilde{u''v''}$ and $\widetilde{u}$ for the flame of Heitor et al. [37], burner 2 at $Re = 37000$. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. The $k-\epsilon$ (solid line) and Reynolds stress (dotted line) turbulence models are used. Crosses and circles refer to the measurements at diametrically opposing locations.

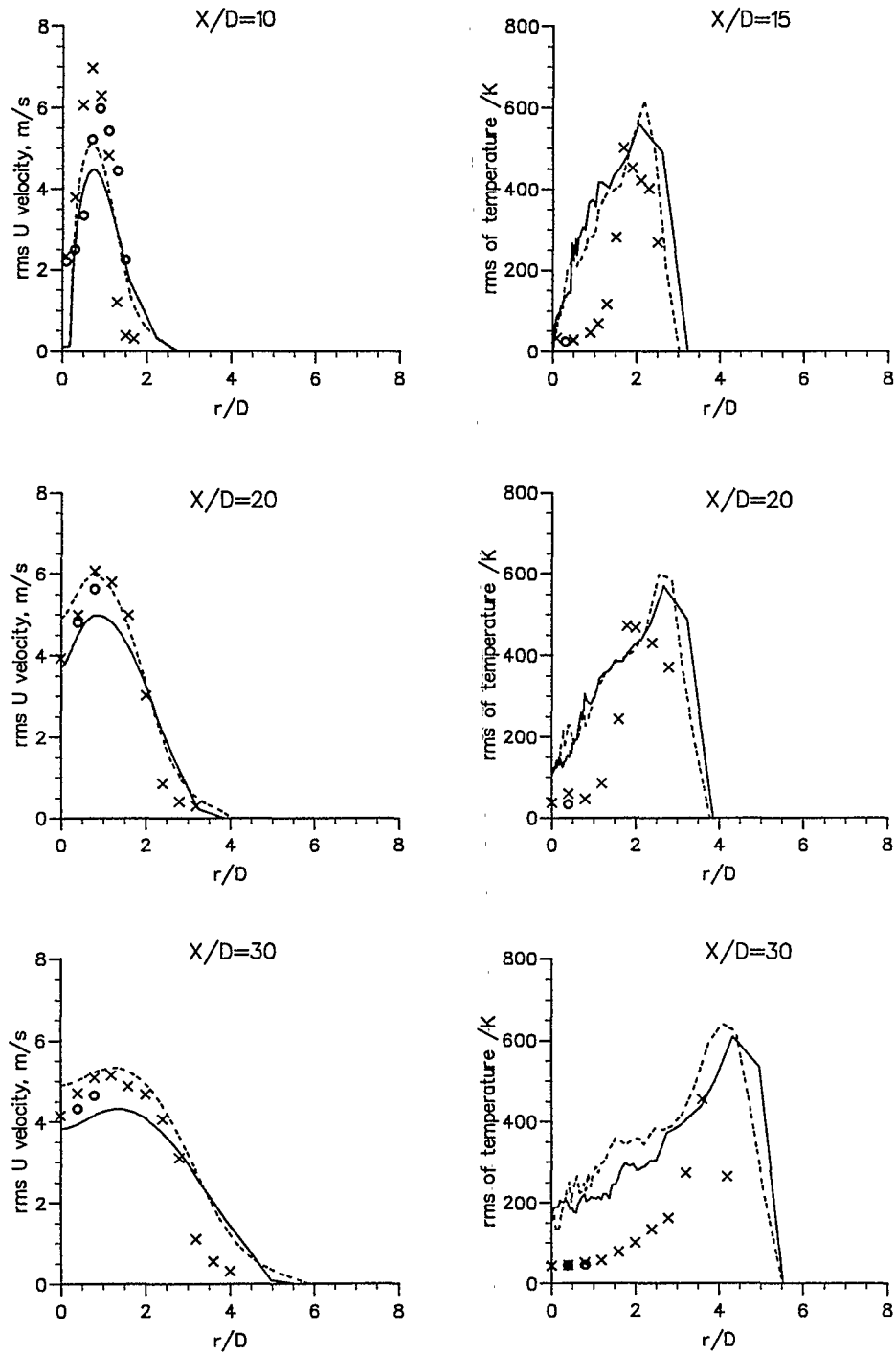


Figure 6.61: Predictions of $(\overline{u'^2})^{1/2}$ and $(\overline{T'^2})^{1/2}$ for the flame of Heitor et al. [37], burner 2 at $Re = 37000$. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. The $k-\varepsilon$ (solid line) and Reynolds stress (dotted line) turbulence models are used. Crosses and circles refer to the measurements at diametrically opposing locations.

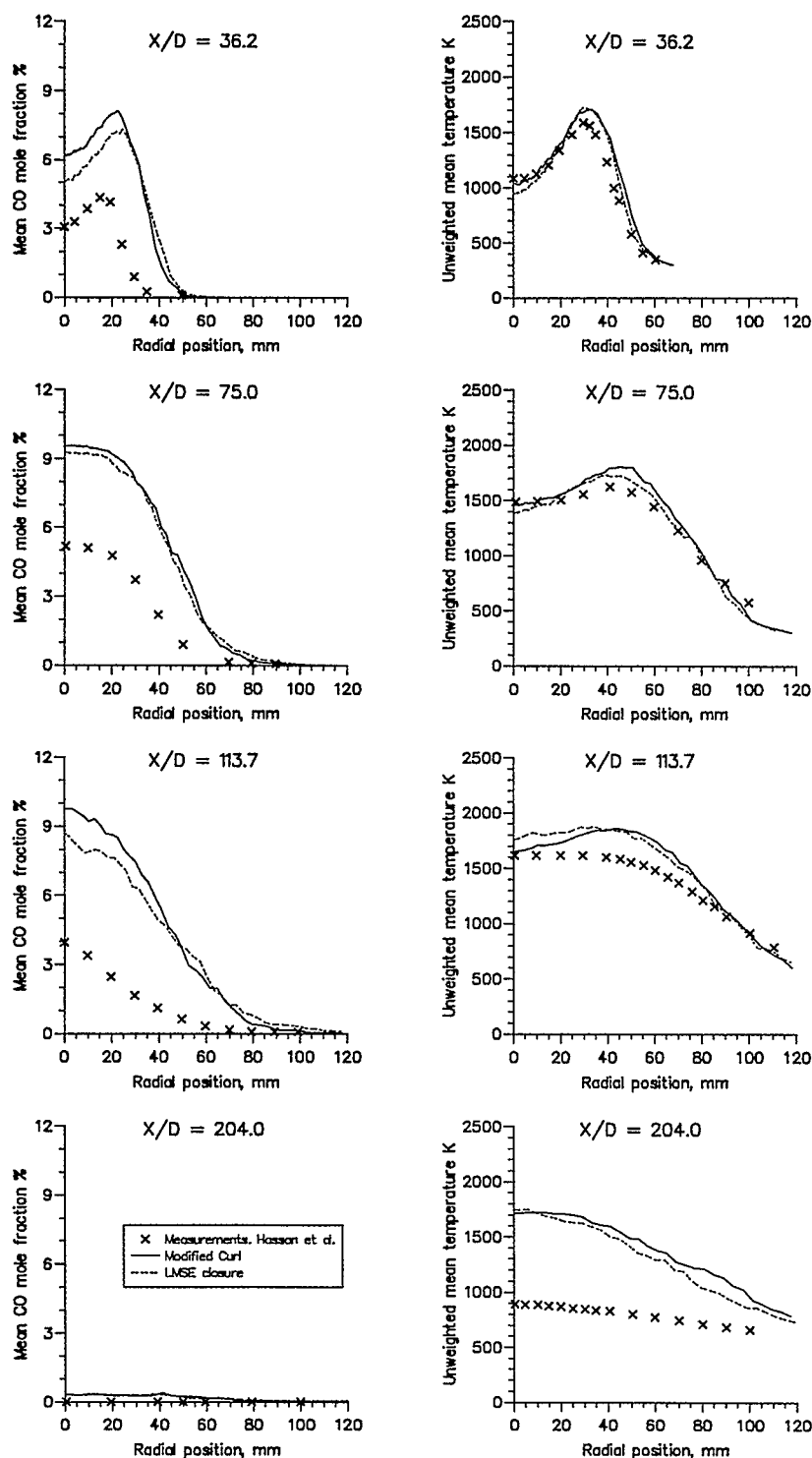


Figure 6.62: Predictions of $\bar{X}_{CO}$ and $\bar{T}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field.

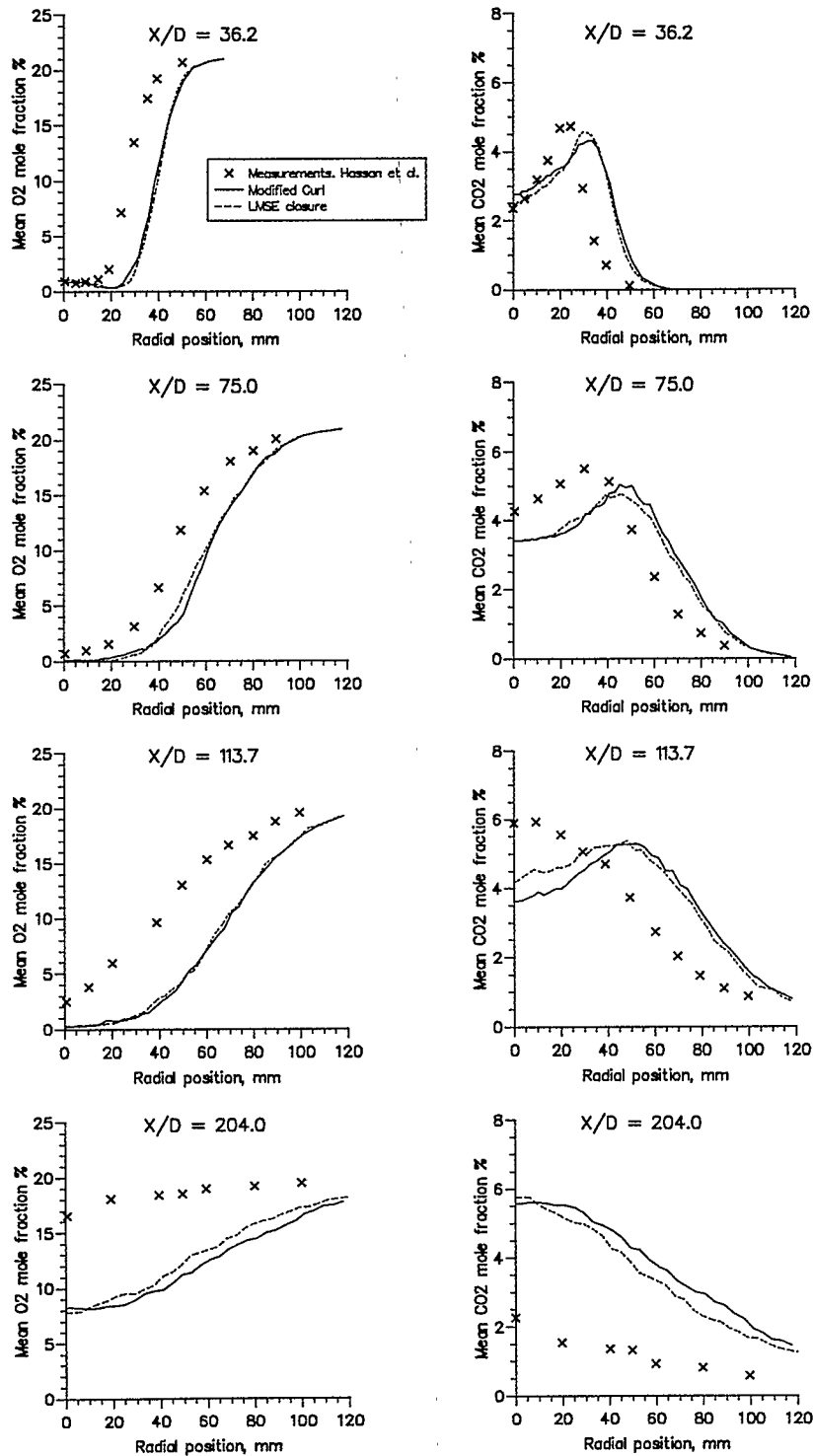


Figure 6.63: Predictions of $\bar{X}_{O_2}$ and $\bar{X}_{CO_2}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field.

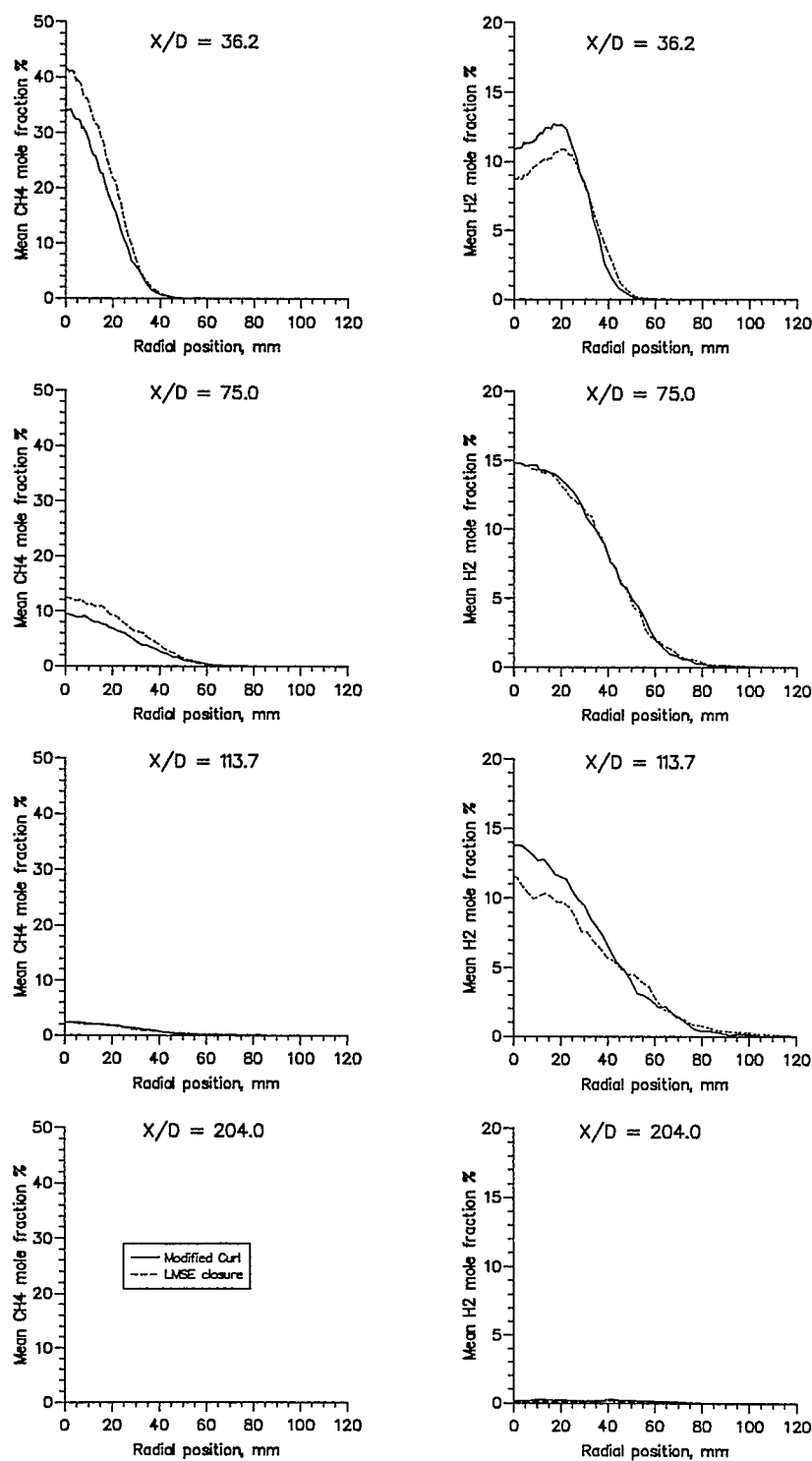


Figure 6.64: Predictions of $\bar{X}_{\text{CH}_4}$ and $\bar{X}_{\text{H}_2}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field.

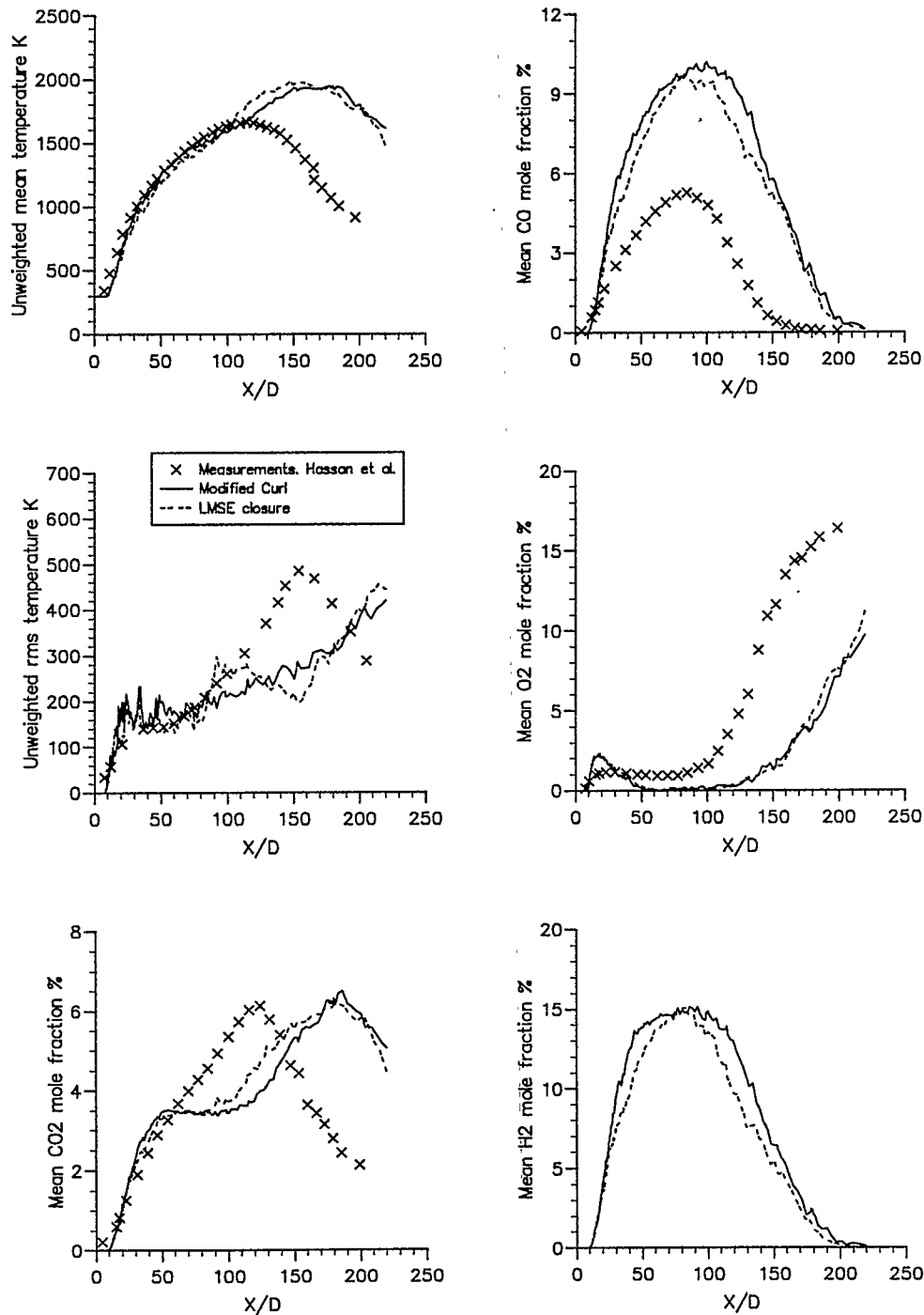


Figure 6.65: Axial evolution of $\bar{T}$, $(\overline{T'^2})^{1/2}$, $\bar{X}_{\text{CO}_2}$, $\bar{X}_{\text{CO}}$, $\bar{X}_{\text{O}_2}$ and $\bar{X}_{\text{H}_2}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field.

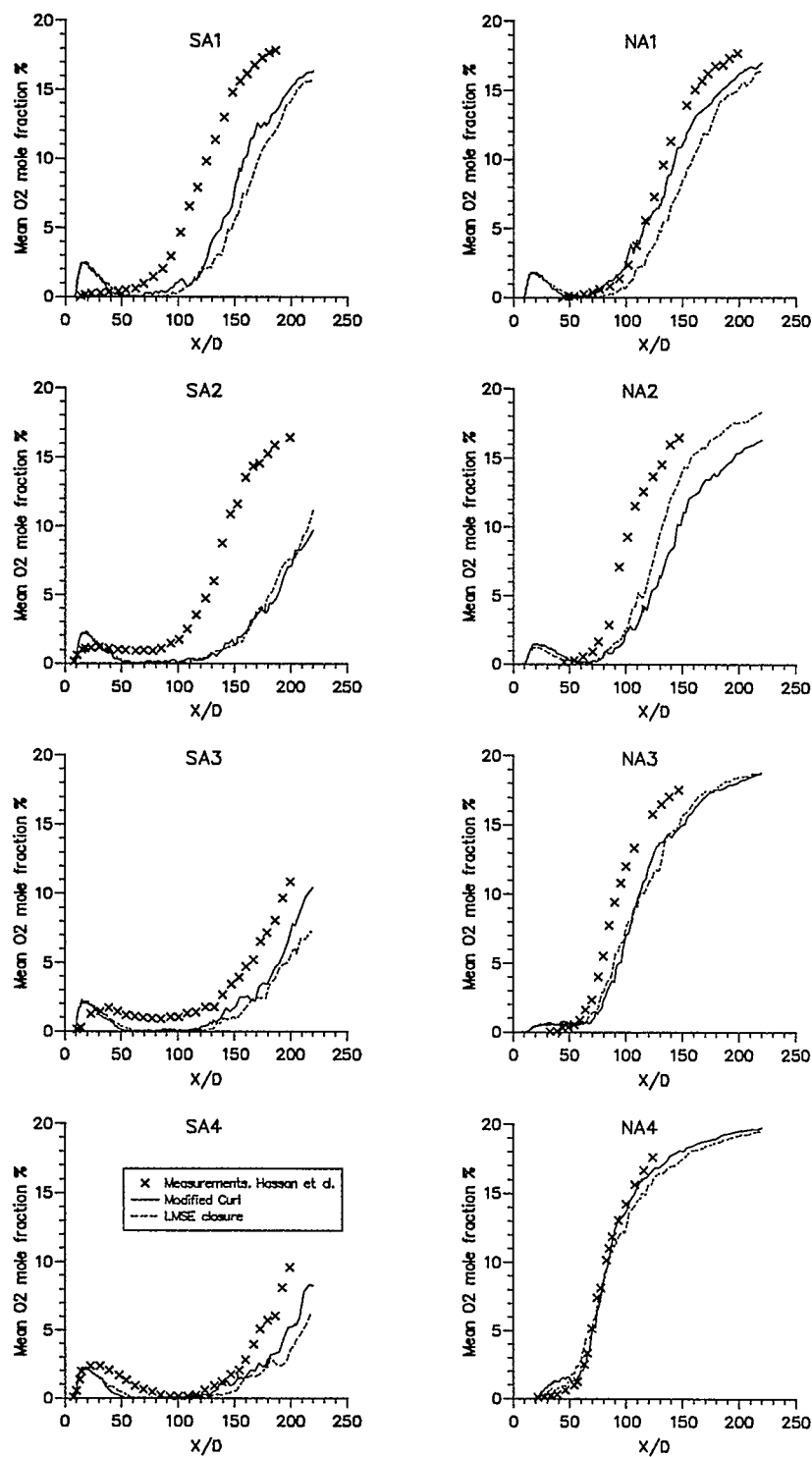


Figure 6.66: Axial evolution of $\bar{X}_{O_2}$ for the flame of Hassan et al. [35] in the burner stabilised configuration with and without N₂-dilution (SA and NA flames). The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. k - ε is used for the velocity field.

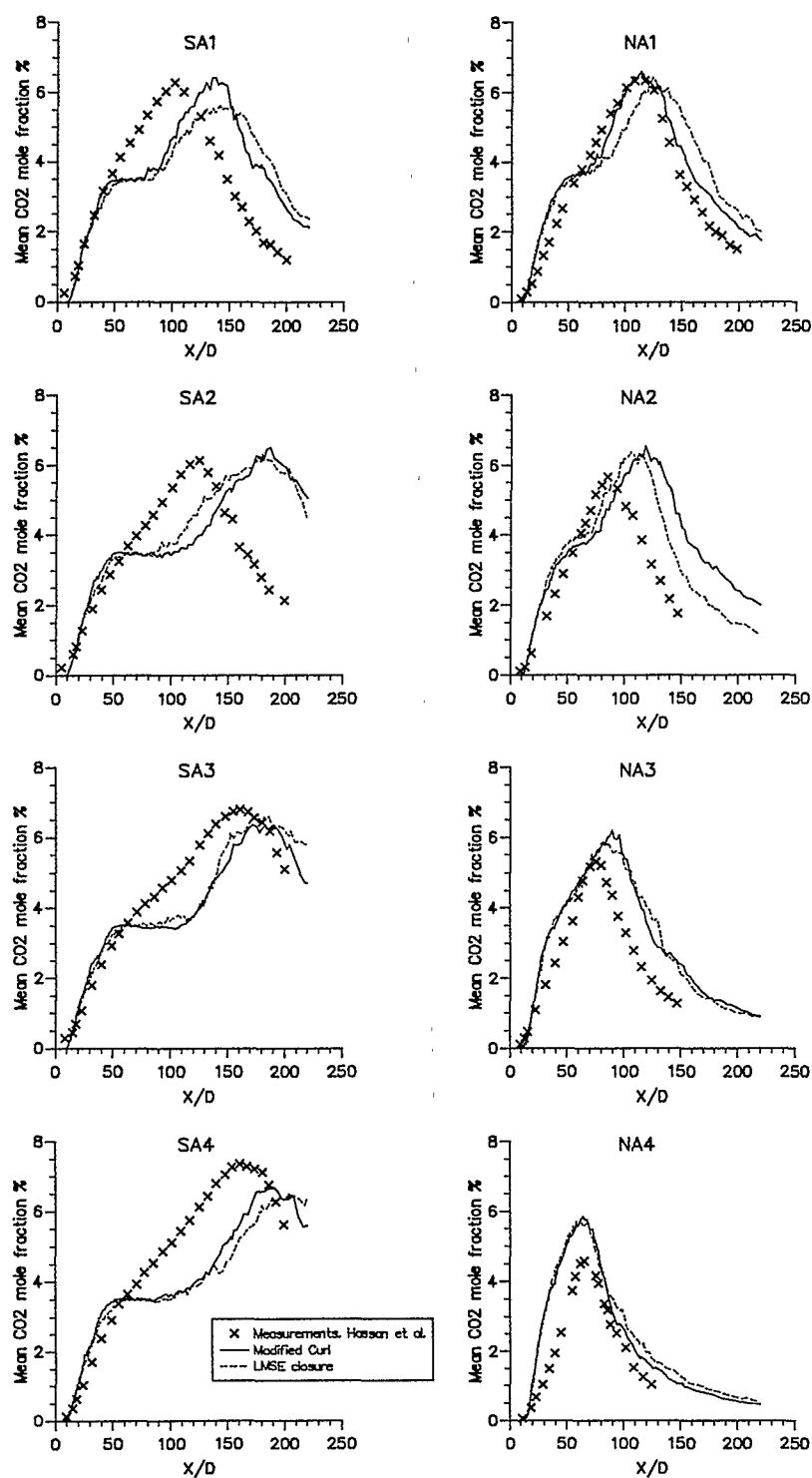


Figure 6.67: Axial evolution of $\bar{X}_{\text{CO}_2}$ for the flame of Hassan et al. [35] in the burner stabilised configuration with and without N_2 -dilution (SA and NA flames). The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field.

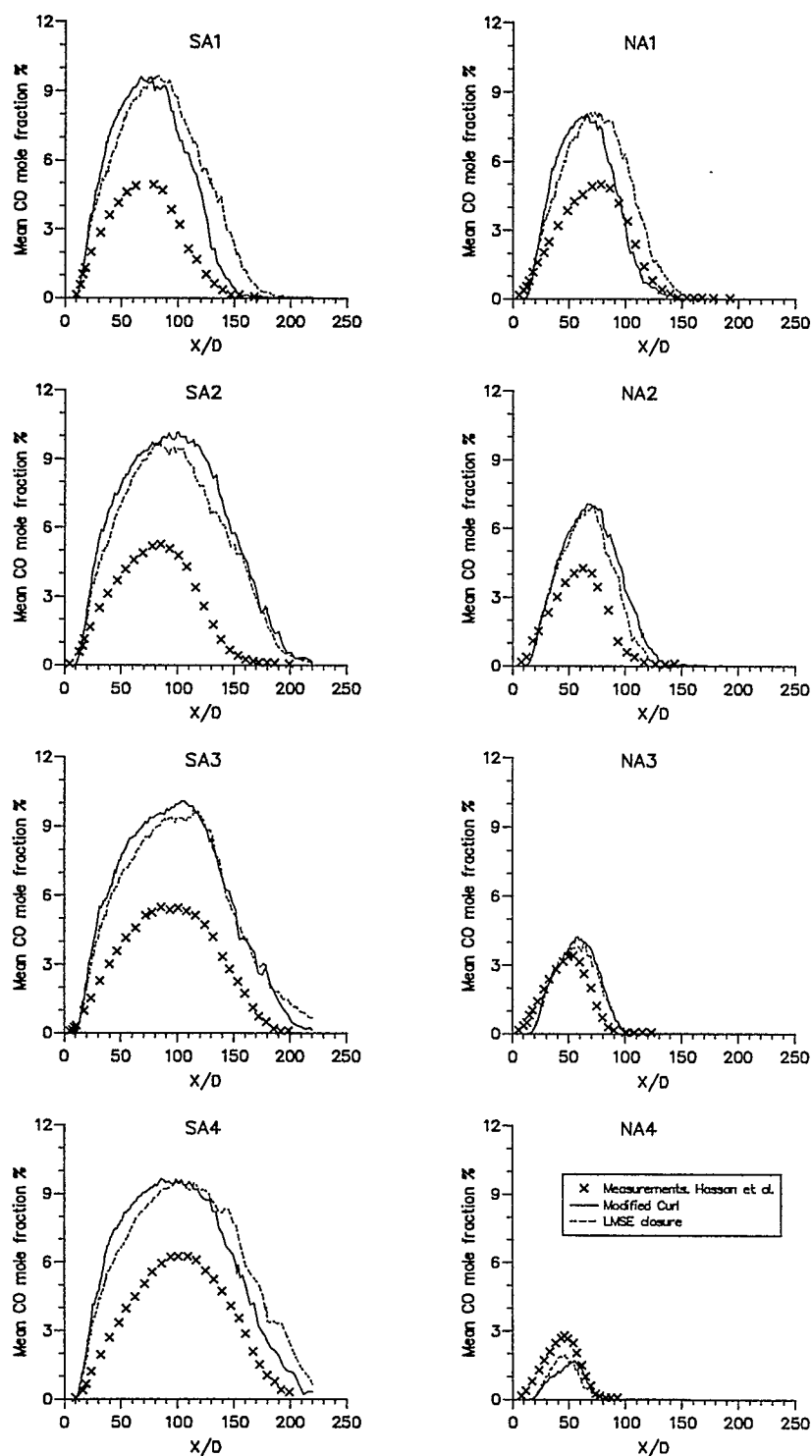


Figure 6.68: Axial evolution of $\bar{X}_{CO}$ for the flame of Hassan et al. [35] in the burner stabilised configuration with and without N_2 -dilution (SA and NA flames). The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field.

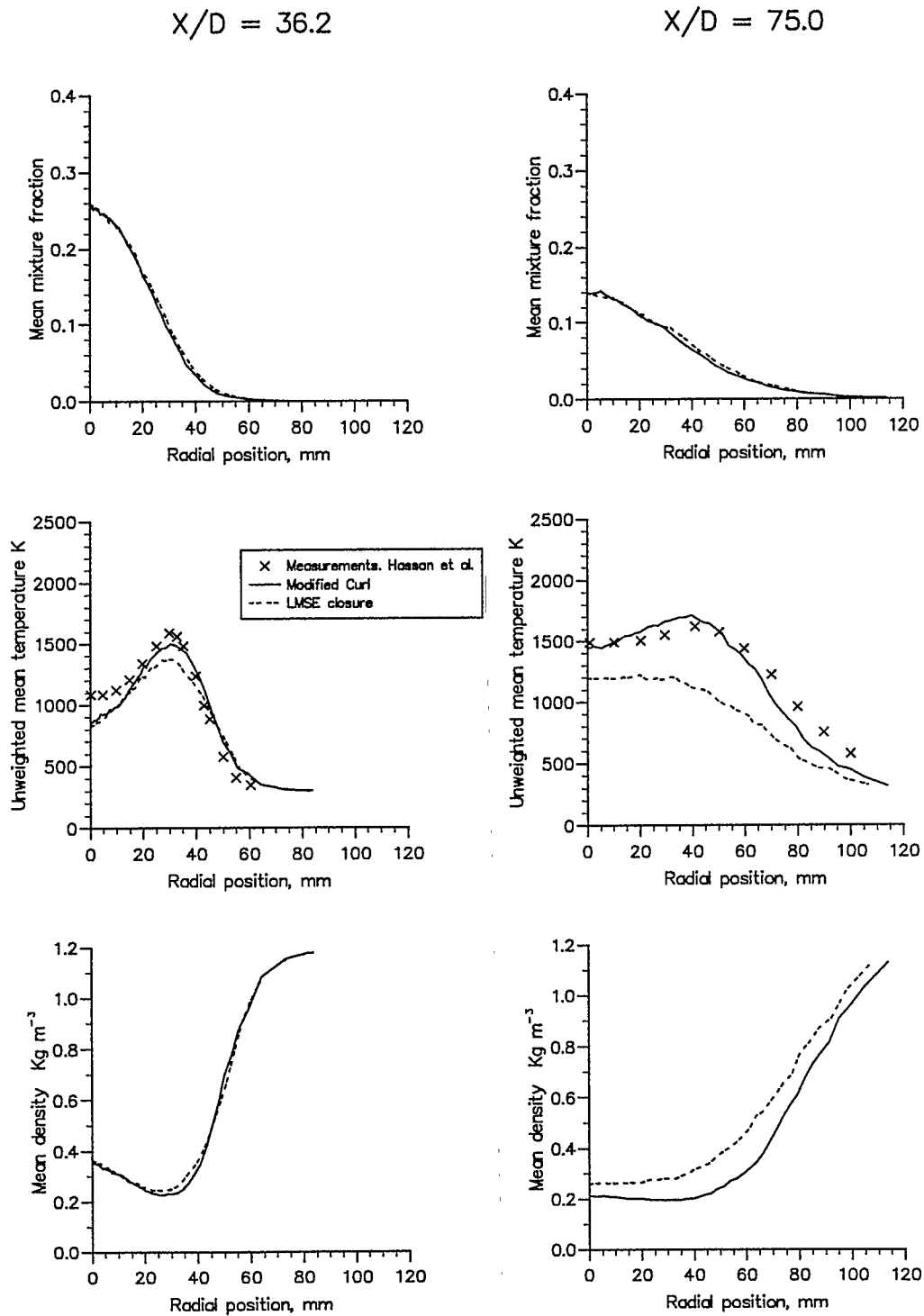


Figure 6.69: Radial profiles of $\tilde{\xi}$, $\bar{T}$ and $\bar{\rho}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Chelliah et al. [10] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. In this calculation the mixture is 'ignited' at $x/D = 15$

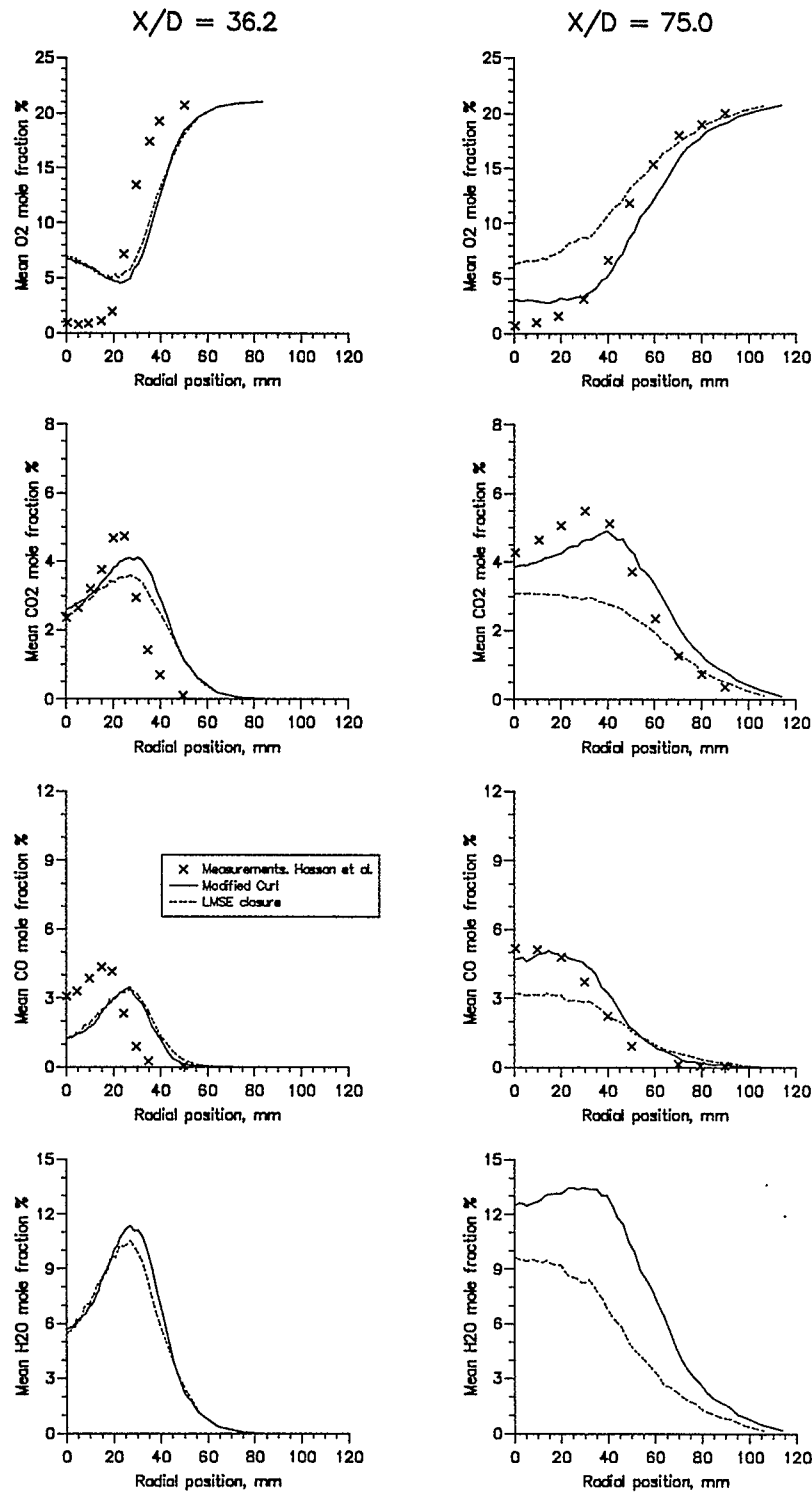


Figure 6.70: Radial profiles of $\bar{X}_{O_2}$, $\bar{X}_{CO_2}$, $\bar{X}_{CO}$ and $\bar{X}_{H_2O}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Chelliah et al. [10] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. In this calculation the mixture is 'ignited' at $x/D = 15$

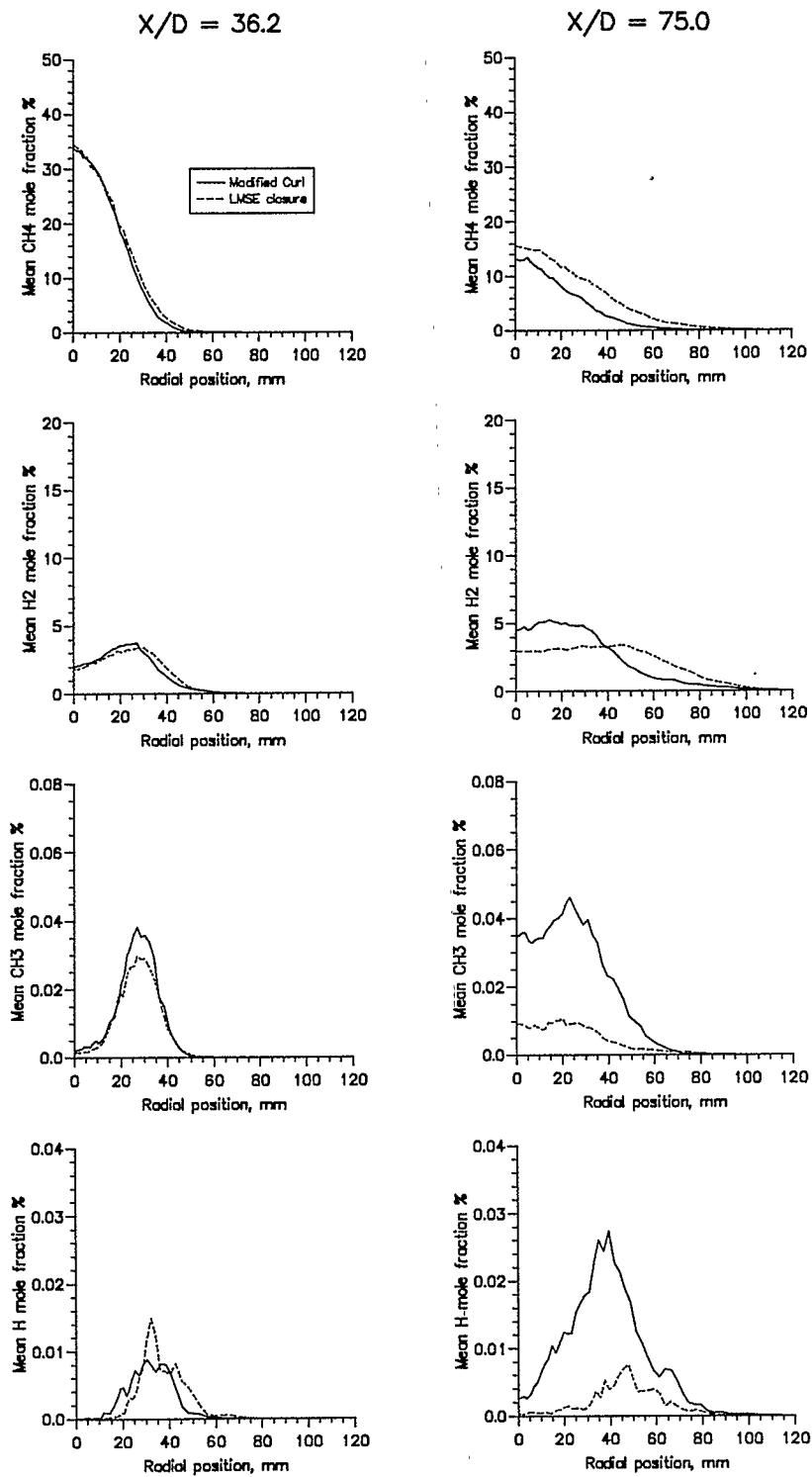


Figure 6.71: Radial profiles of $\tilde{X}_{CH_4}$, $\tilde{X}_{H_2}$, $\tilde{X}_{CH_3}$ and $\tilde{X}_H$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Chelliah et al. [10] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field. In this calculation the mixture is 'ignited' at $x/D = 15$

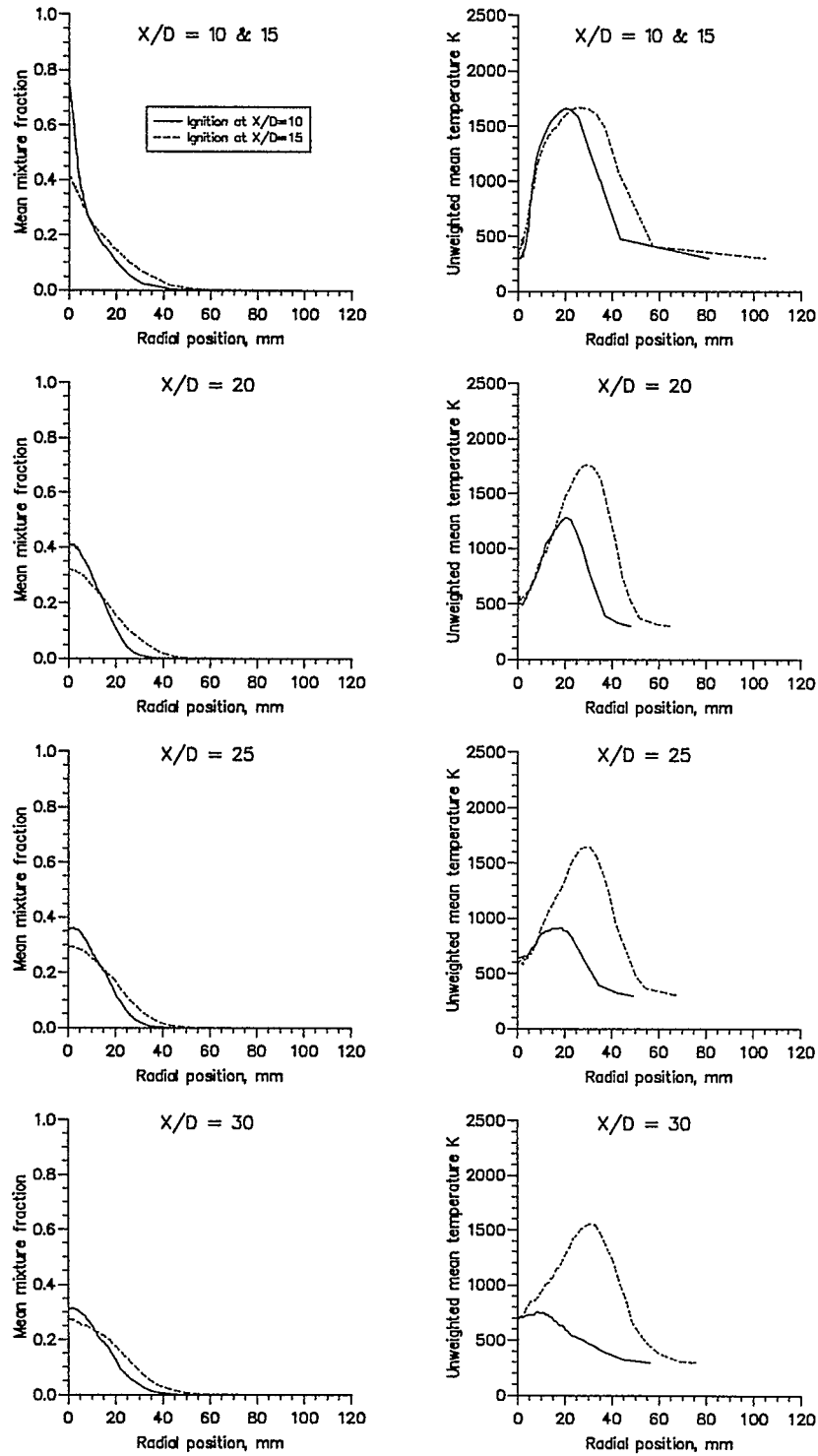


Figure 6.72: Radial profiles of $\bar{\xi}$ and $\bar{T}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Chelliah et al. [10] is employed, with modified Curl as the mixing model. $k-\varepsilon$ is used for the velocity field. In this figure, the effect of igniting the mixture at $x/D = 10$ and $x/D = 15$ is compared.

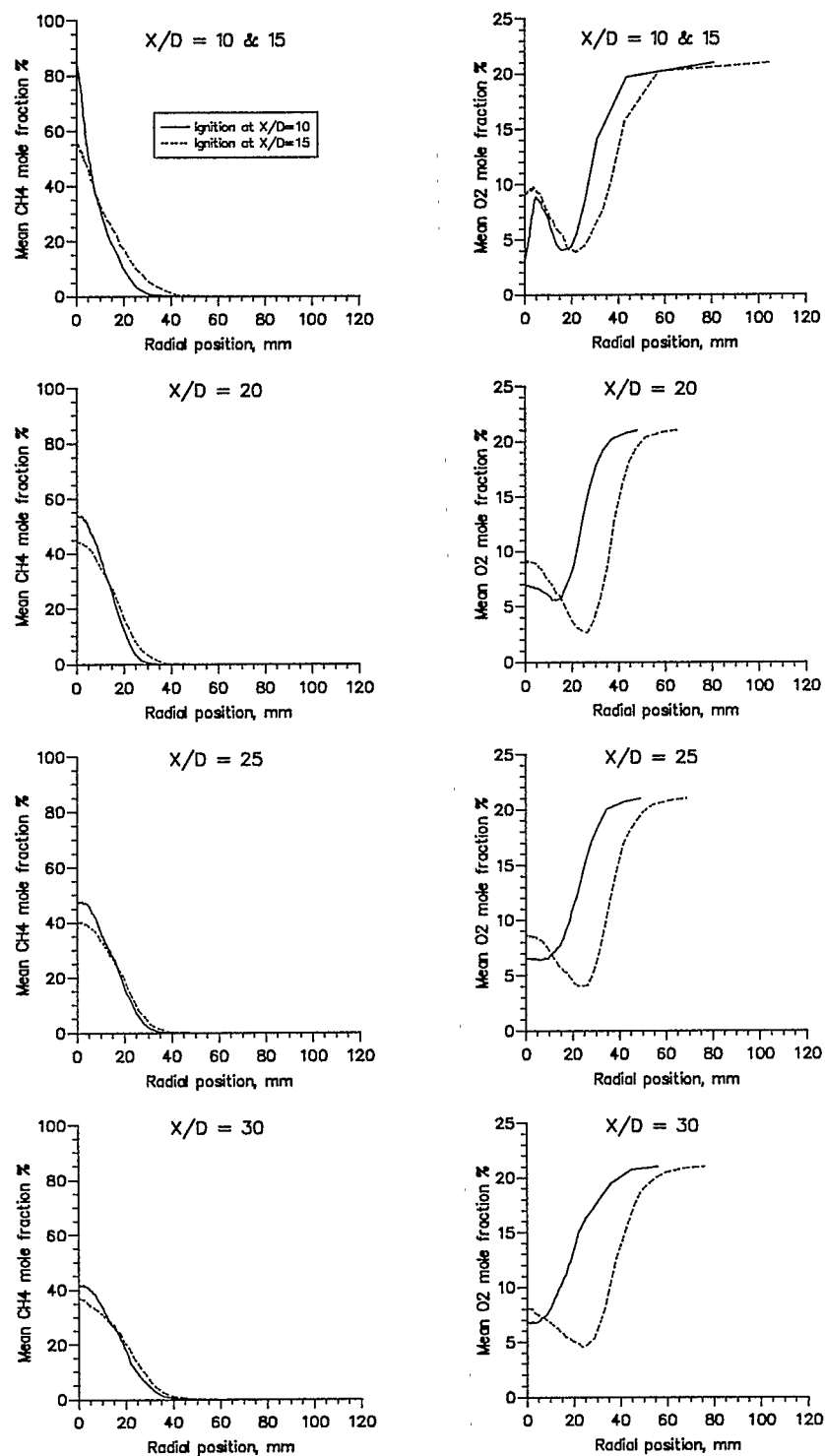


Figure 6.73: Radial profiles of $\bar{X}_{\text{CH}_4}$ and $\bar{X}_{\text{O}_2}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Chelliah et al. [10] is employed, with modified Curl as the mixing model. $k-\epsilon$ is used for the velocity field. In this figure, the effect of igniting the mixture at $x/D = 10$ and $x/D = 15$ is compared.

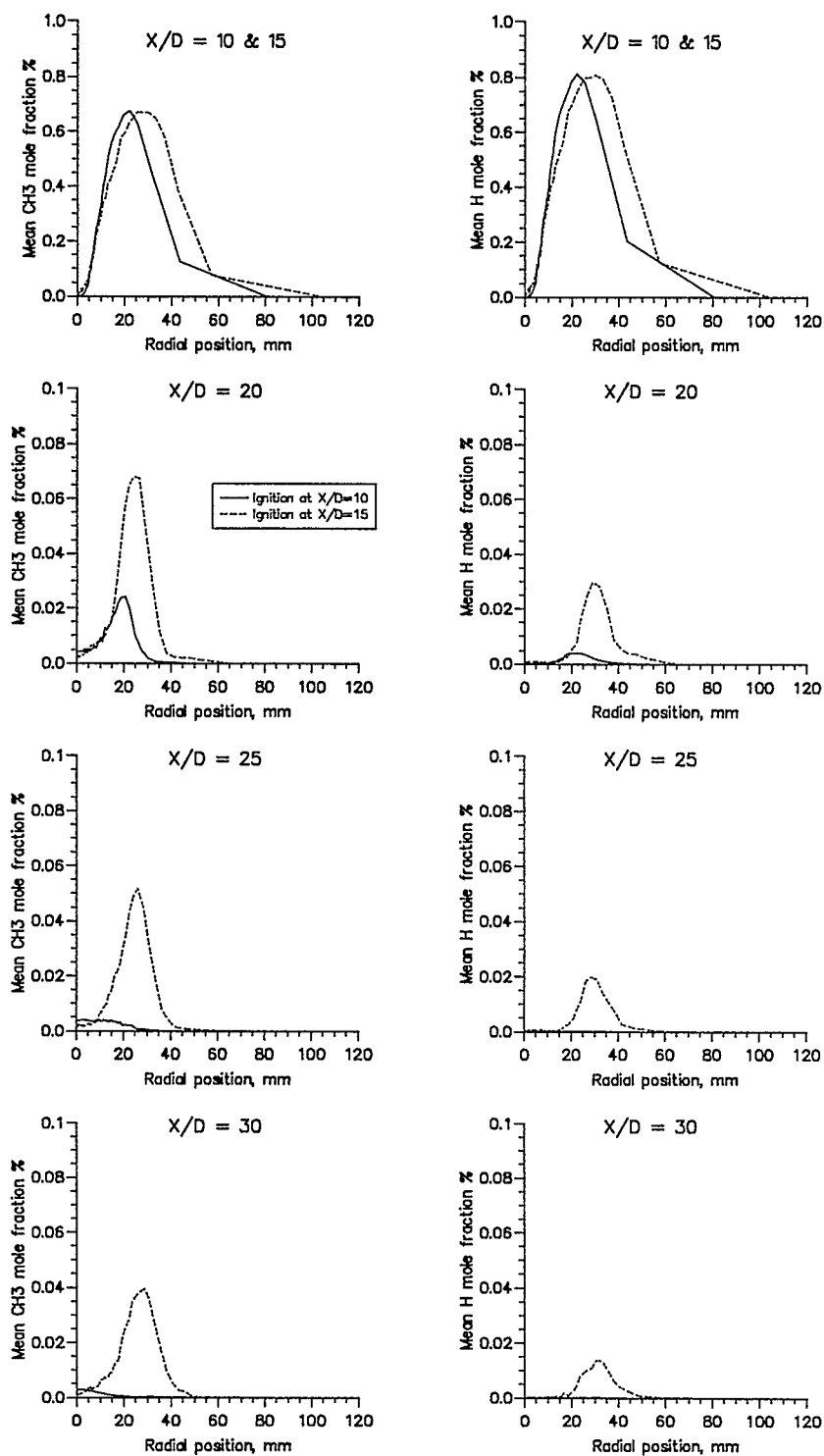


Figure 6.74: Radial profiles of $\tilde{X}_{\text{CH}_3}$ and $\tilde{X}_{\text{H}}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Chelliah et al. [10] is employed, with modified Curl as the mixing model. $k-\epsilon$ is used for the velocity field. In this figure, the effect of igniting the mixture at $x/D = 10$ and $x/D = 15$ is compared.

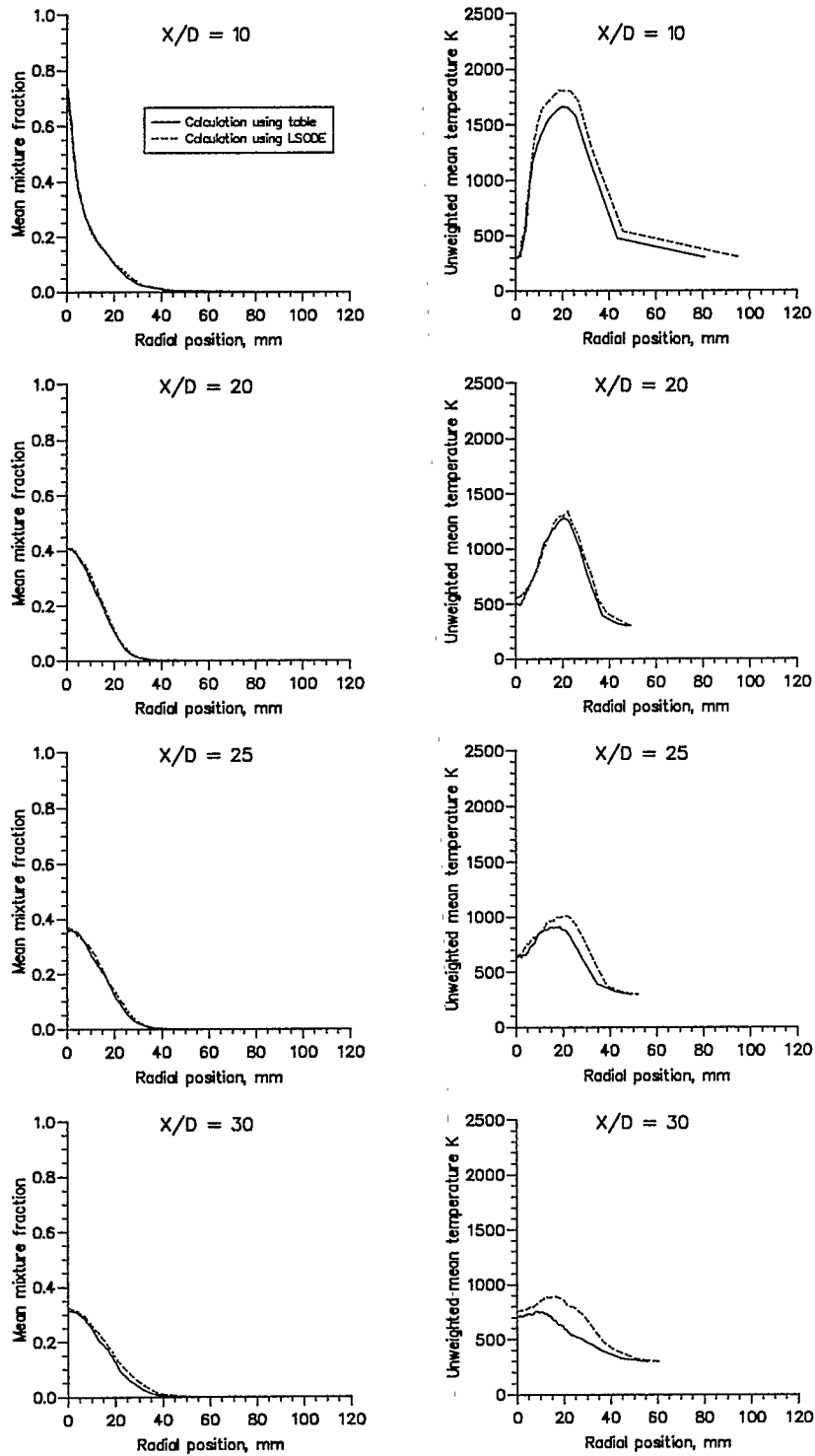


Figure 6.75: Radial profiles of $\tilde{\xi}$ and $\bar{T}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Chelliah et al. [10] is employed, with modified Curl as the mixing model. $k-\varepsilon$ is used for the velocity field. In this figure, the results of the interpolation table are compared with using LSODE for the test case where ignition is performed at $x/D = 10$.

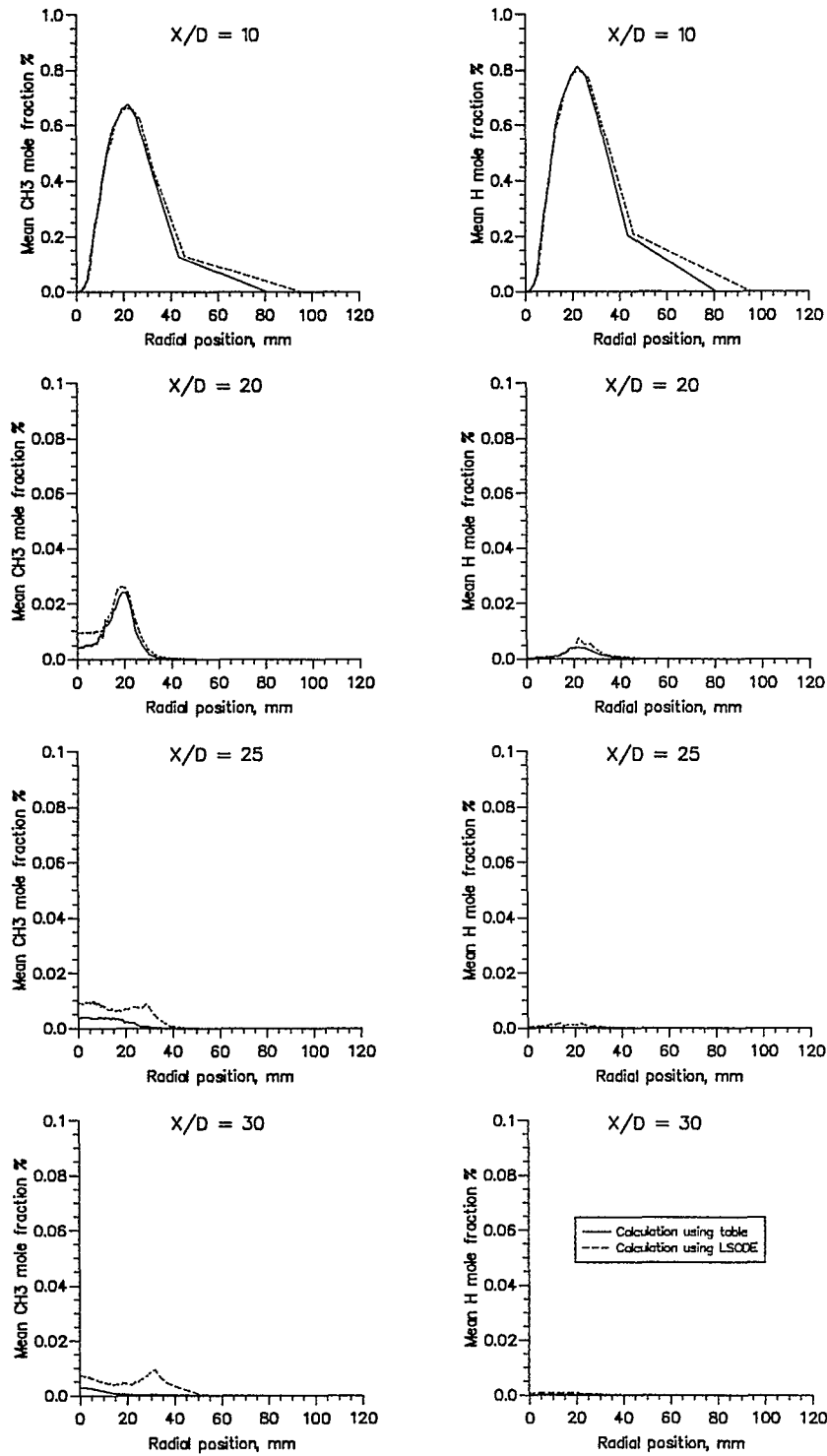


Figure 6.76: Radial profiles of $\bar{X}_{\text{CH}_3}$ and $\bar{X}_{\text{H}}$ for the flame of Hassan et al. [35] (SA2). The reduced scheme of Chelliah et al. [10] is employed, with modified Curl as the mixing model. $k-\epsilon$ is used for the velocity field. In this figure, the results of the interpolation table are compared with using LSODE for the test case where ignition is performed at $x/D = 10$.

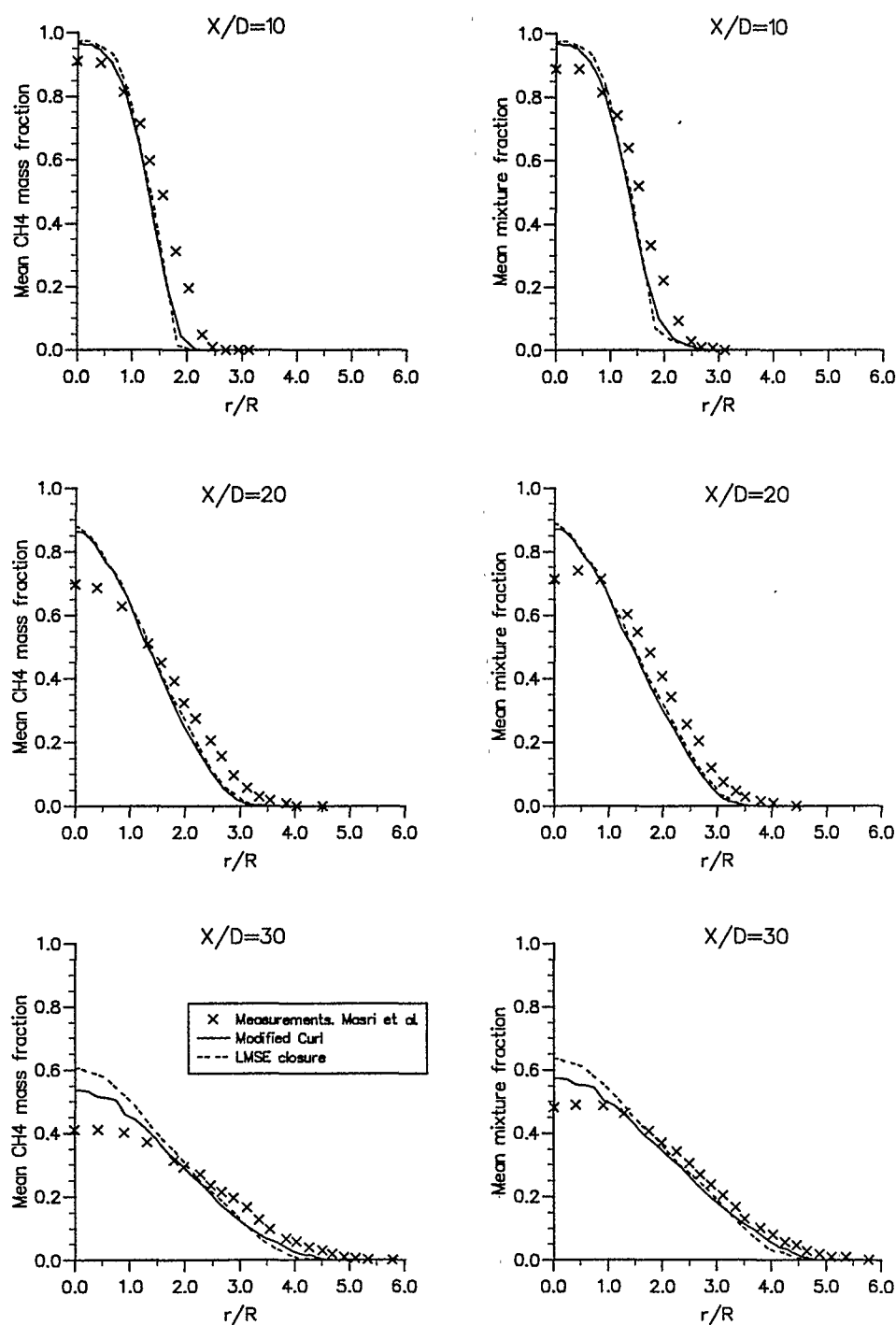


Figure 6.77: Radial profiles of $\bar{Y}_{CH_4}$ and $\bar{\xi}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.

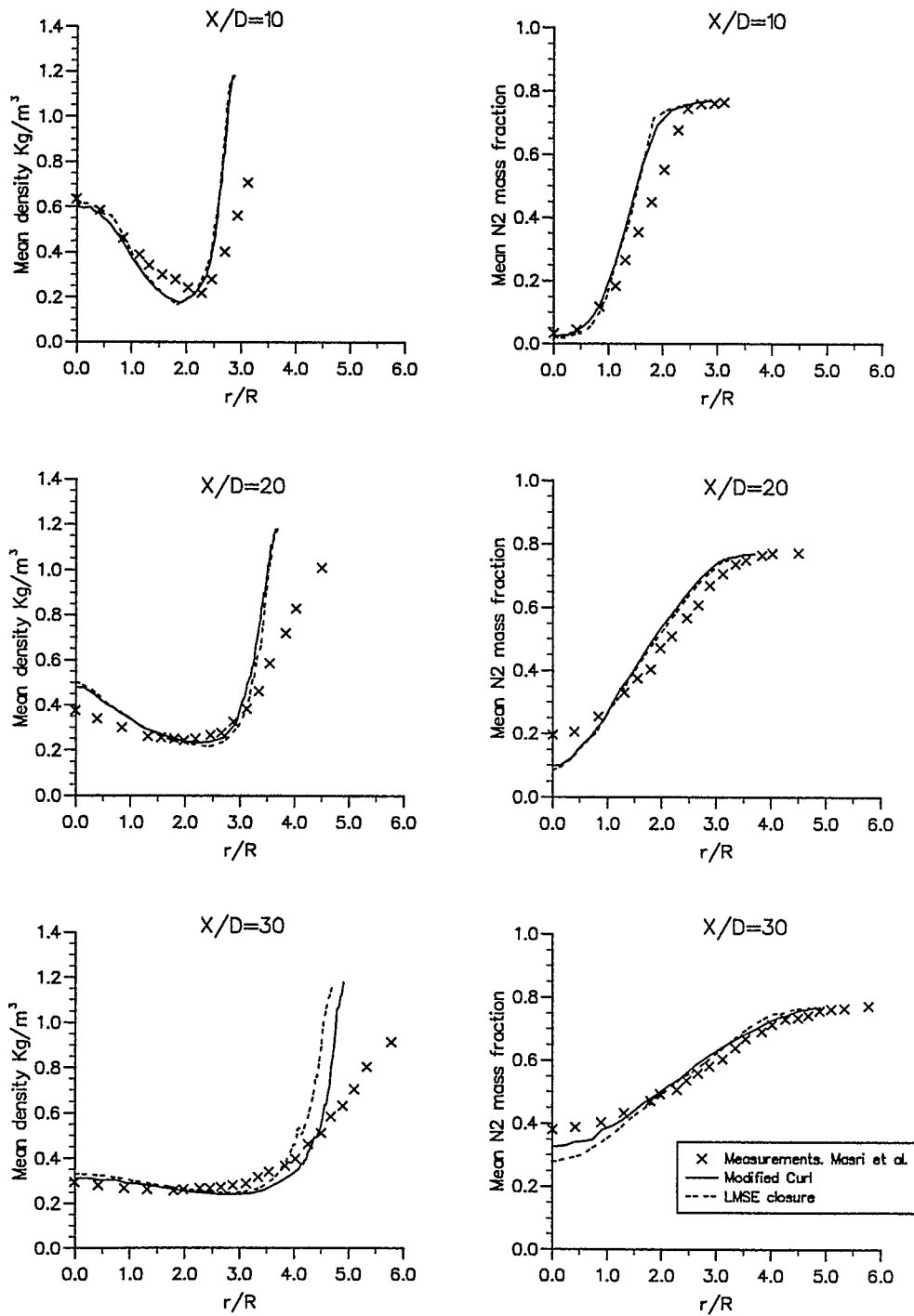


Figure 6.78: Radial profiles of $\bar{\rho}$ and $\bar{Y}_{\text{N}_2}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.

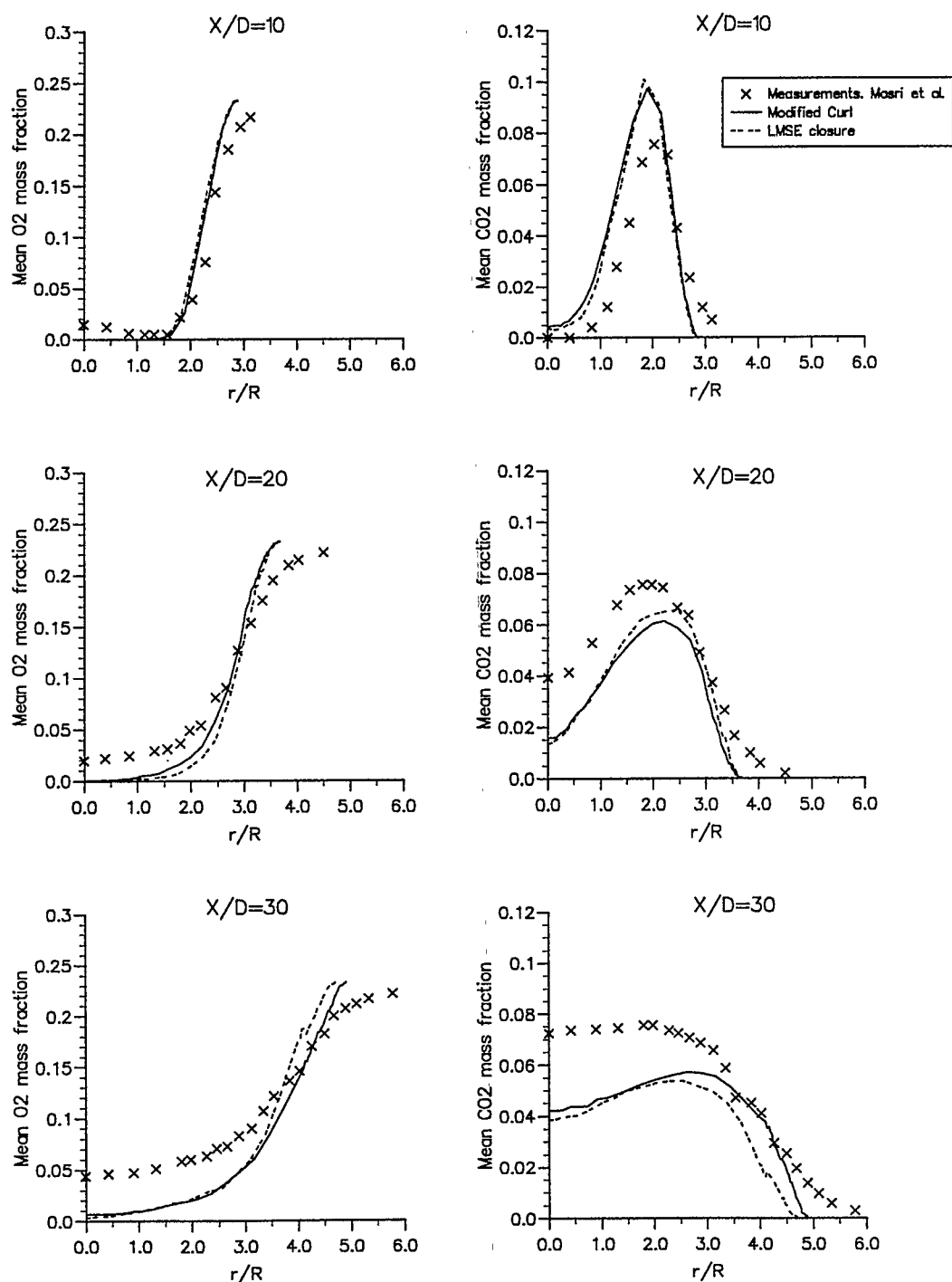


Figure 6.79: Radial profiles of $\bar{Y}_{O_2}$ and $\bar{Y}_{CO_2}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.

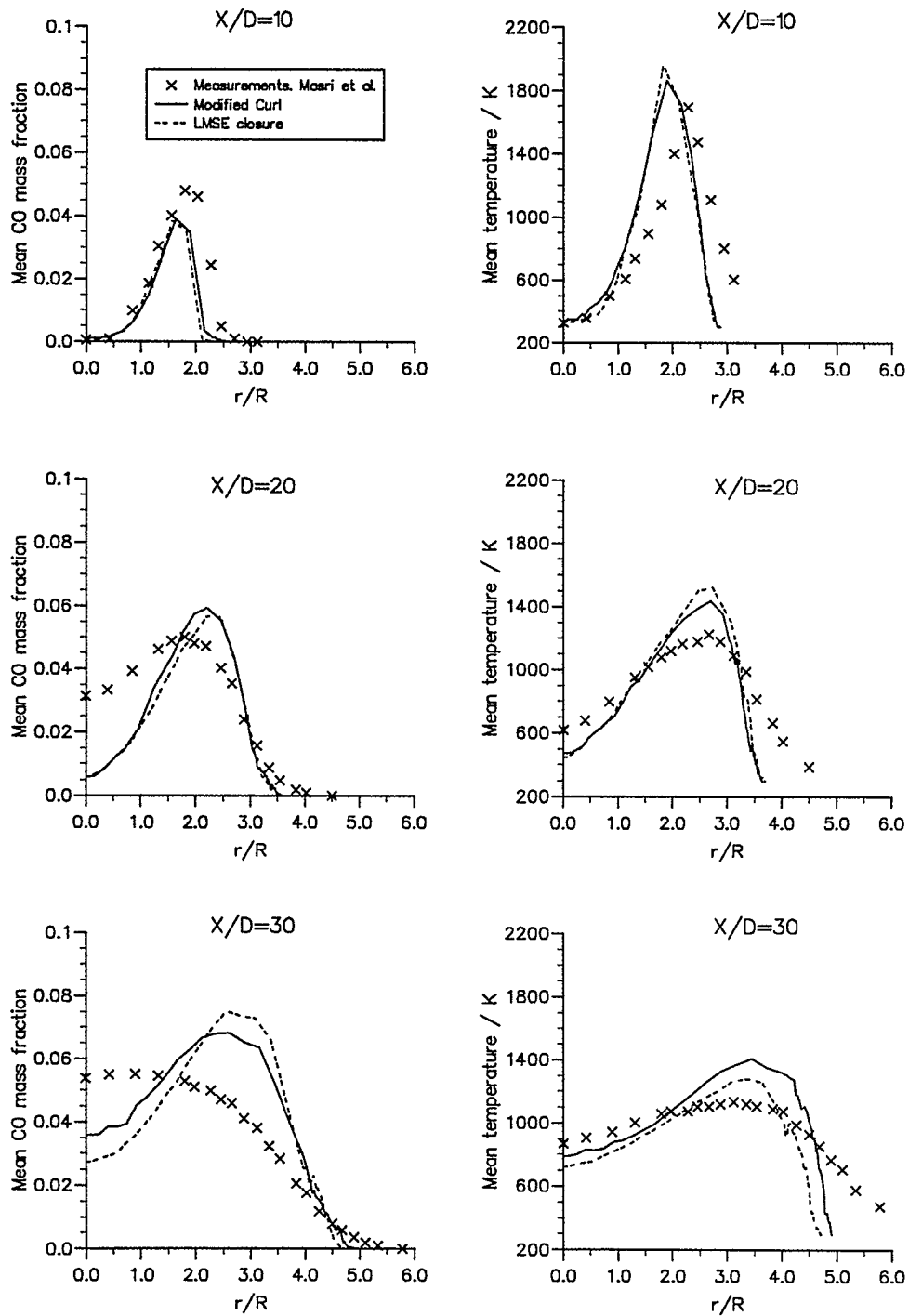


Figure 6.80: Radial profiles of $\bar{Y}_{CO}$ and $\bar{T}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.

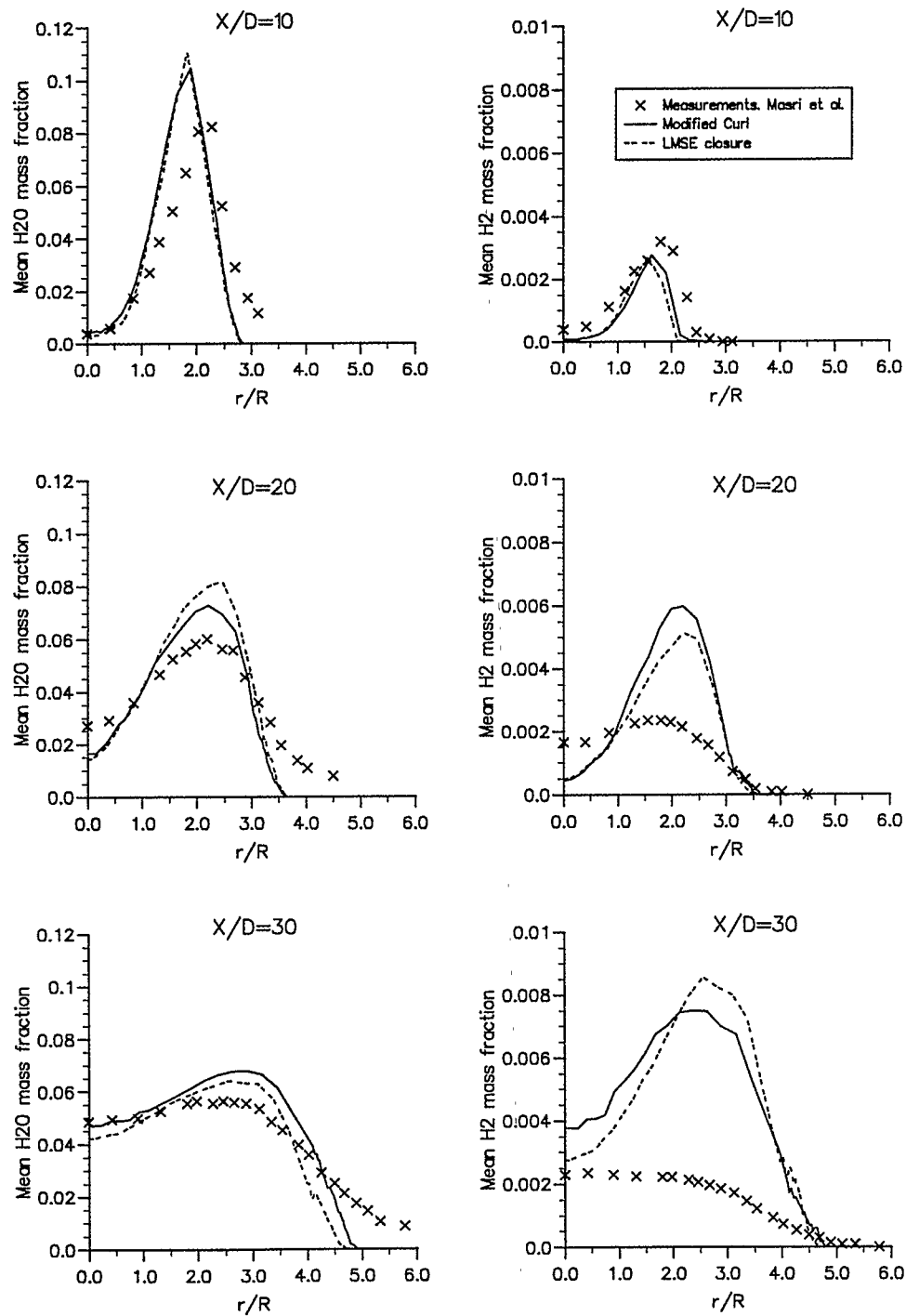


Figure 6.81: Radial profiles of $\bar{Y}_{H_2O}$ and $\bar{Y}_{H_2}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.

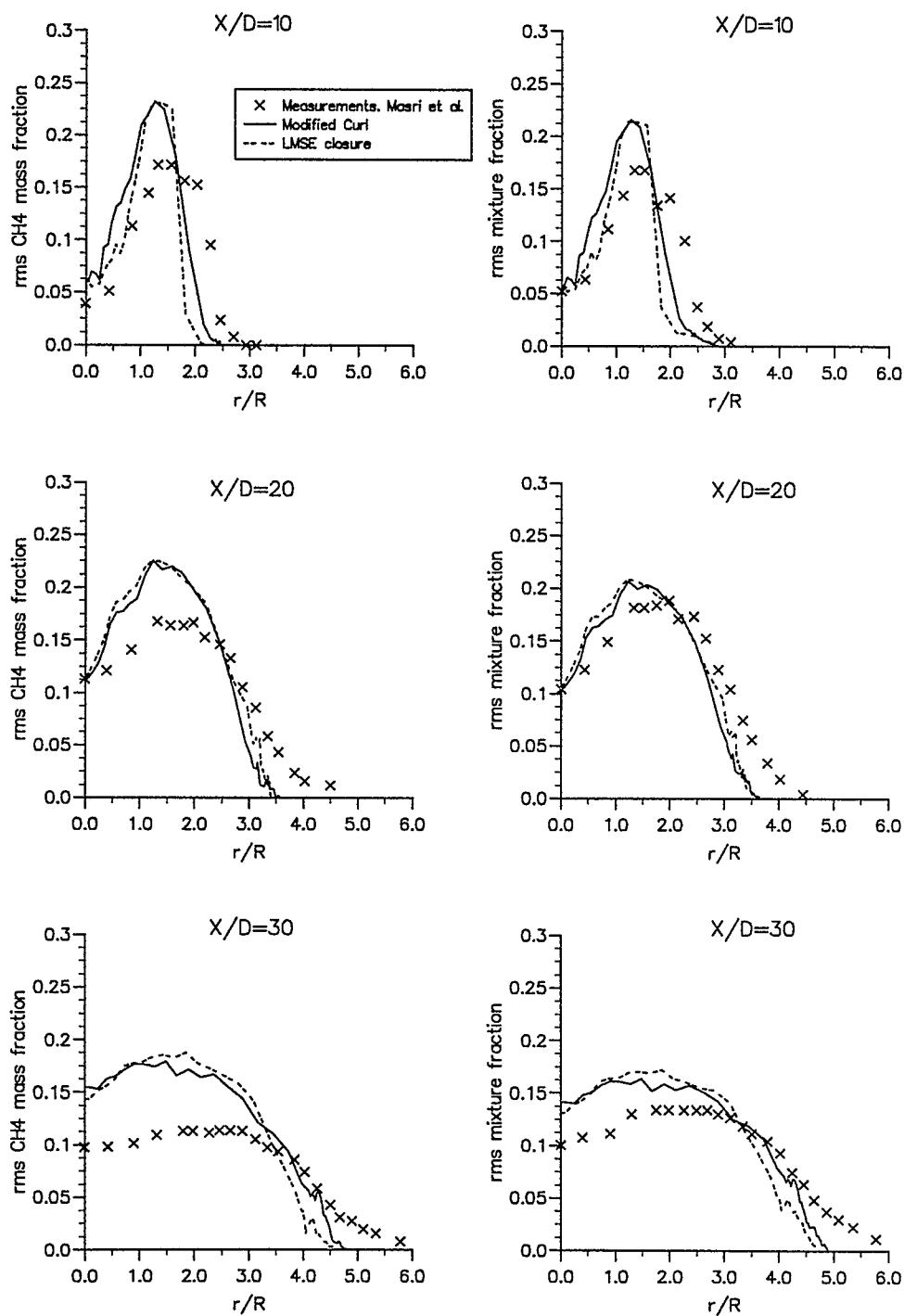


Figure 6.82: Radial profiles of $(\widetilde{Y_{CH_4}''^2})^{1/2}$ and $(\widetilde{\xi''^2})^{1/2}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.

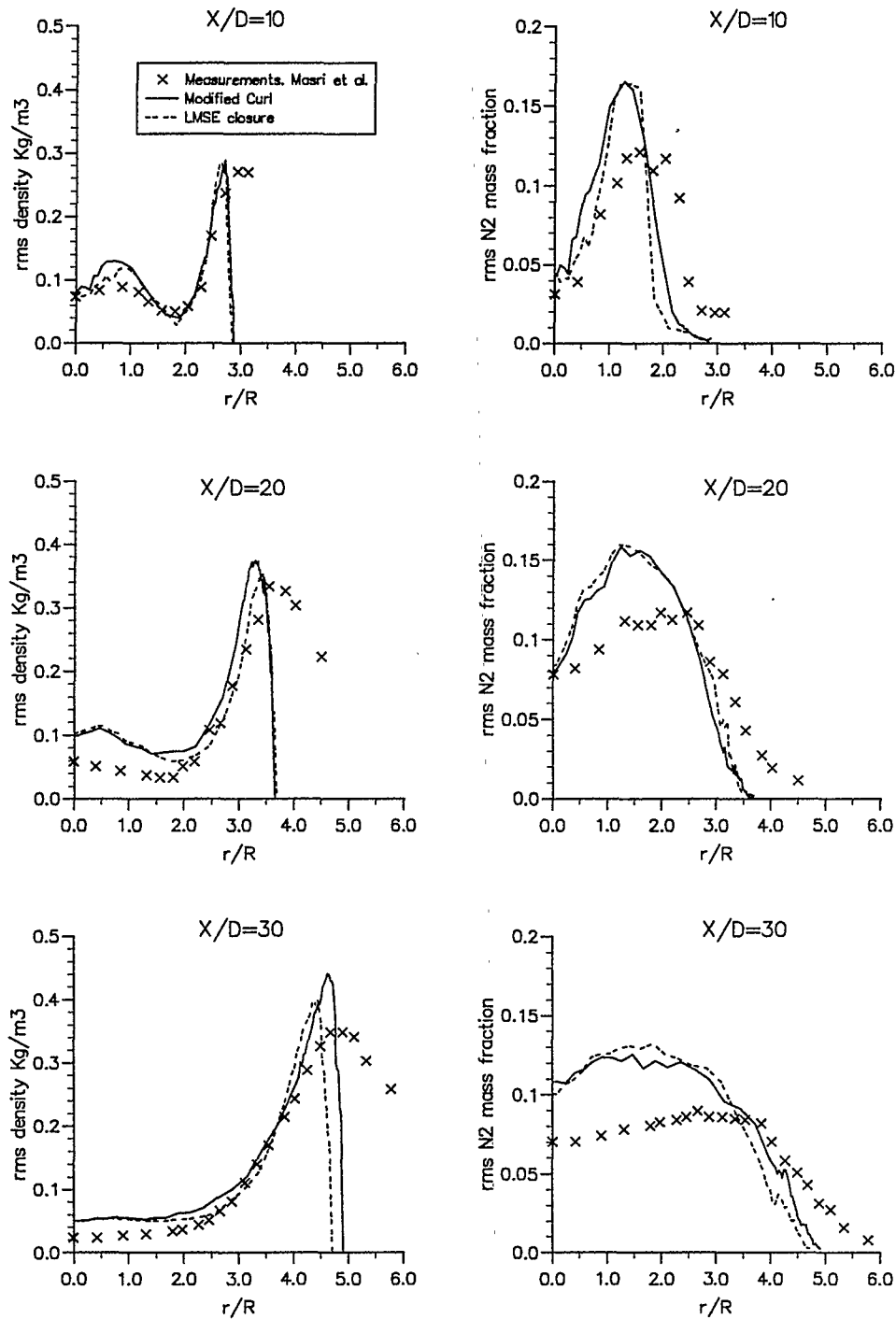


Figure 6.83: Radial profiles of $(\overline{\rho'^2})^{1/2}$ and $(\overline{Y_{N_2}'^2})^{1/2}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.

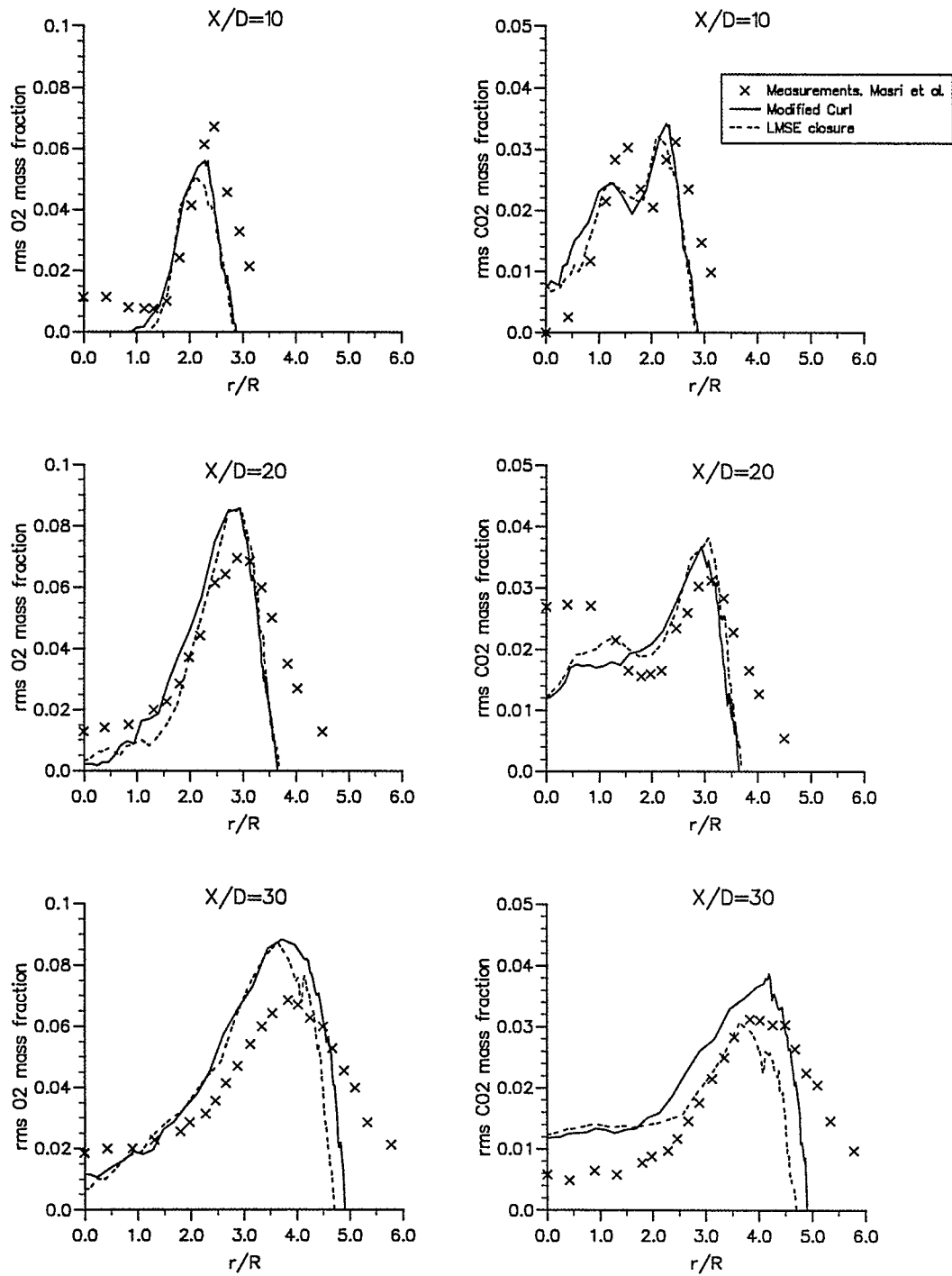


Figure 6.84: Radial profiles of $(\widetilde{Y_{O_2}''})^{\frac{1}{2}}$ and $(\widetilde{Y_{CO_2}''})^{\frac{1}{2}}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.

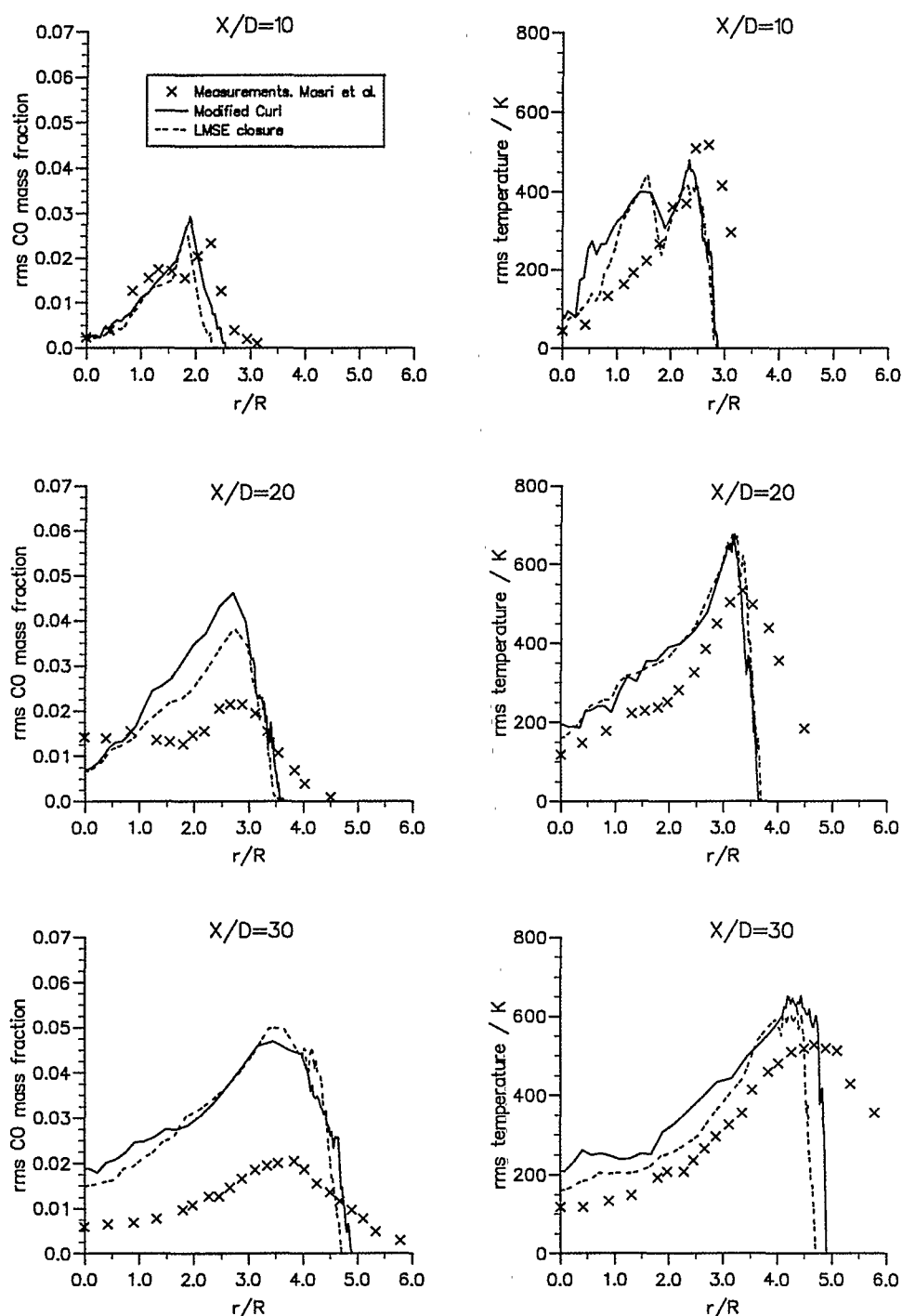


Figure 6.85: Radial profiles of $(\widetilde{Y_{CO}''^2})^{1/2}$ and $(\widetilde{T'^2})^{1/2}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.

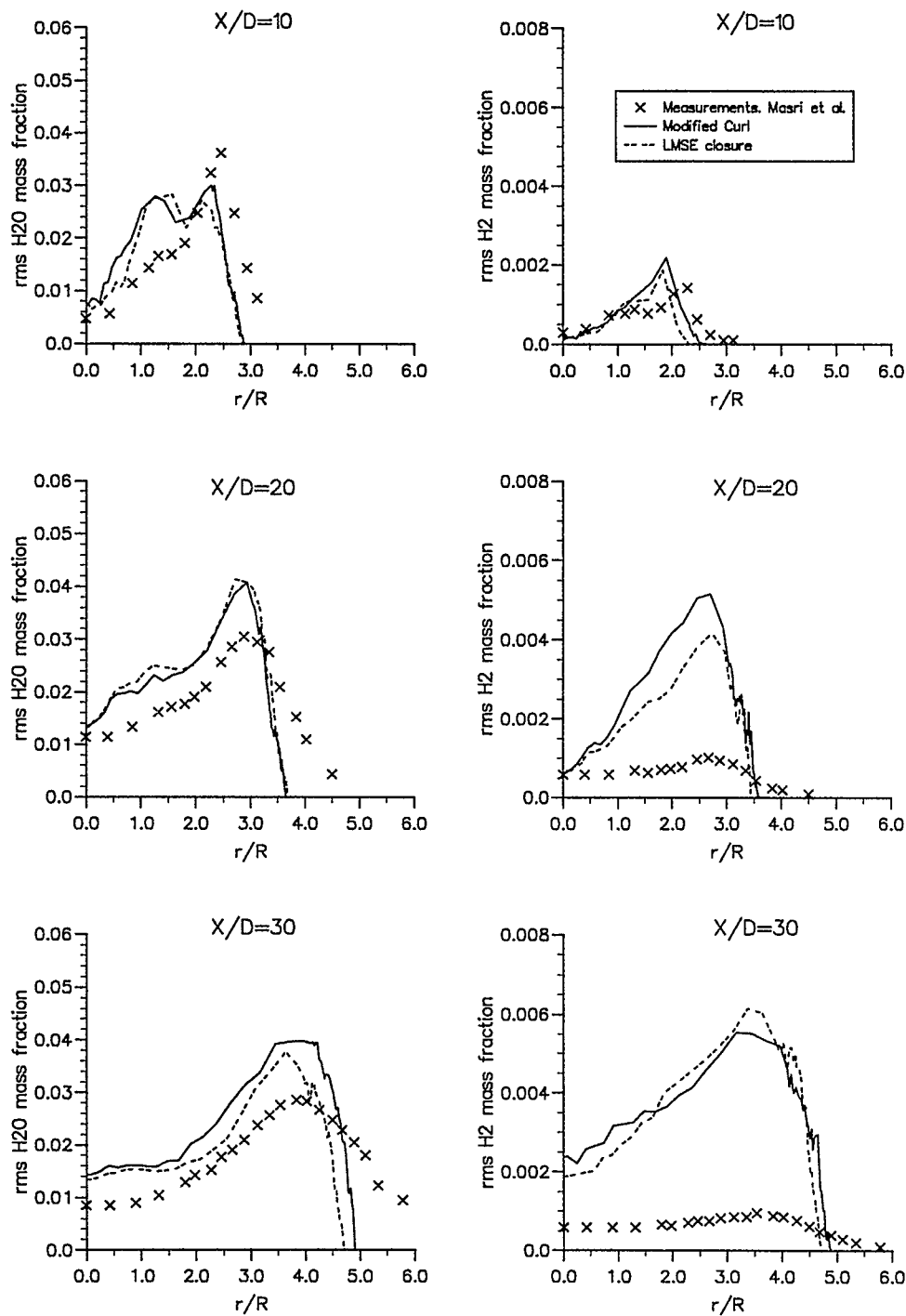


Figure 6.86: Radial profiles of $(\widetilde{Y_{H_2O}^{1/2}})^{1/2}$ and $(\widetilde{Y_{H_2}^{1/2}})^{1/2}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.

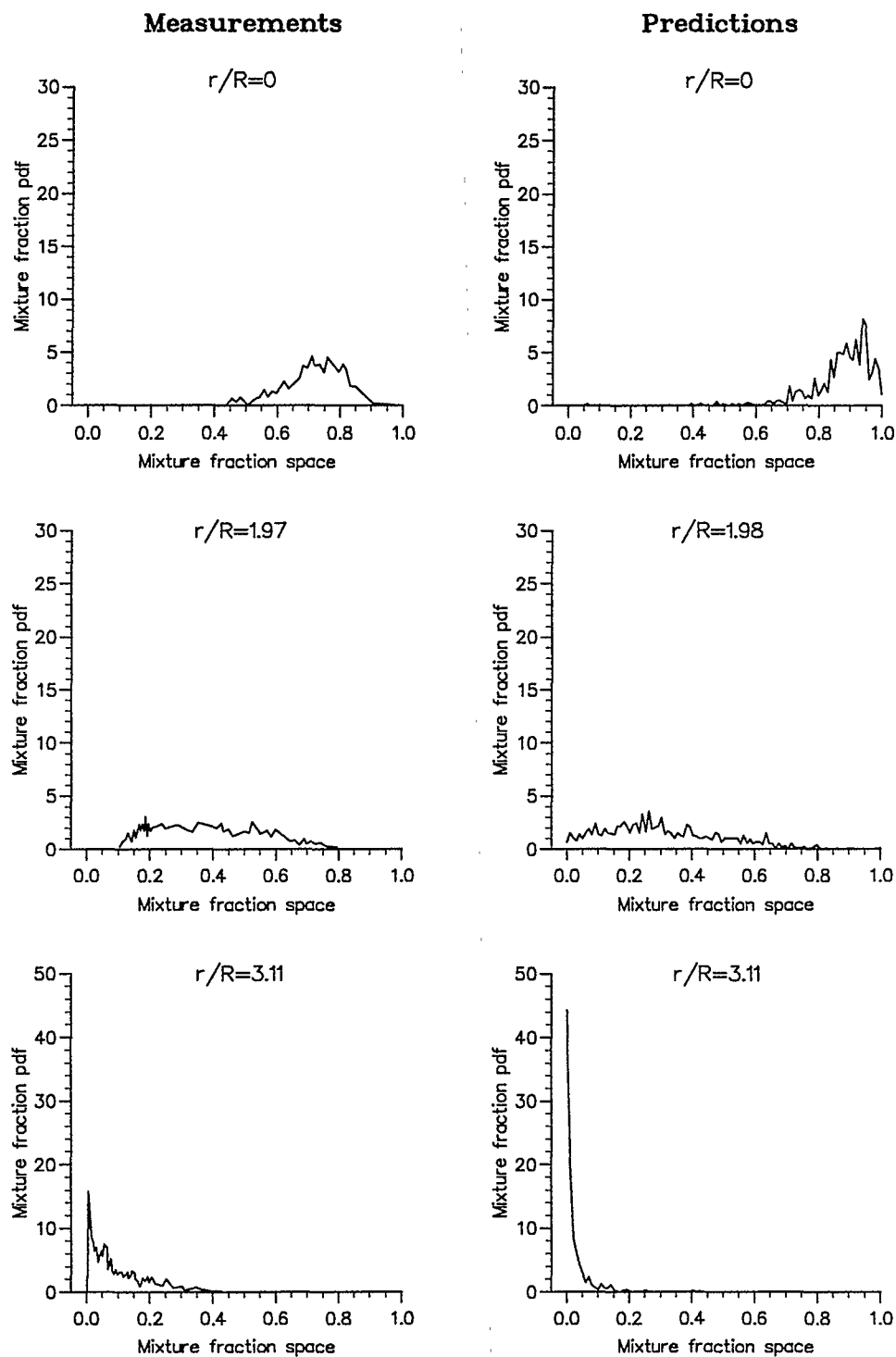


Figure 6.87: Pdf profiles at $x/D = 20$ of $\tilde{P}(\hat{\xi}; x, \omega)$ for the (L) flame of Masri et al. [67]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. $k-\varepsilon$ is used for the velocity field.

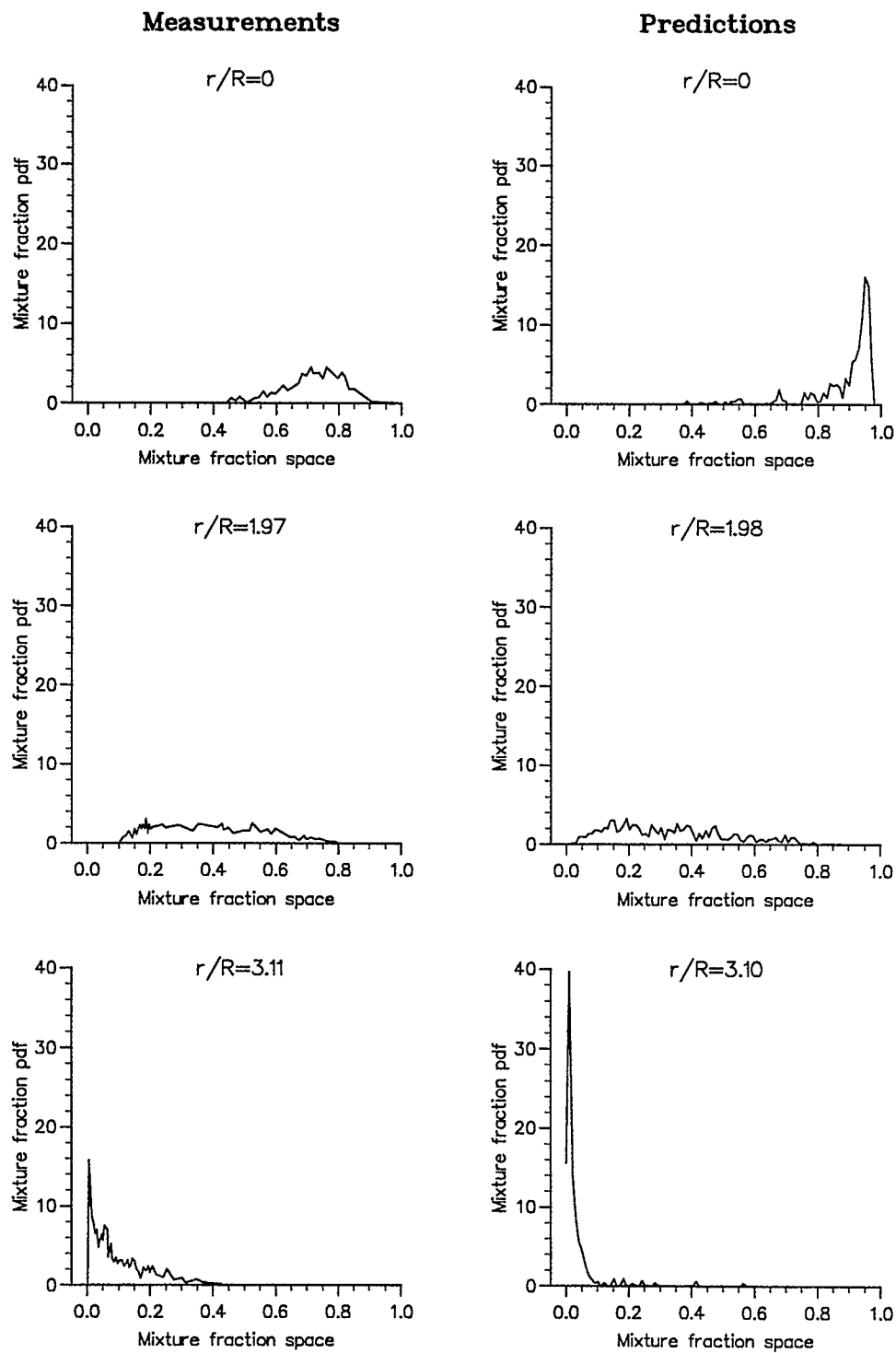


Figure 6.88: Pdf profiles at $x/D = 20$ of $\tilde{P}(\xi; x, \omega)$ for the (L) flame of Masri et al. [67]. The reduced scheme of Jones and Lindstedt [46] is employed, with LMSE as the mixing model. $k-\varepsilon$ is used for the velocity field.

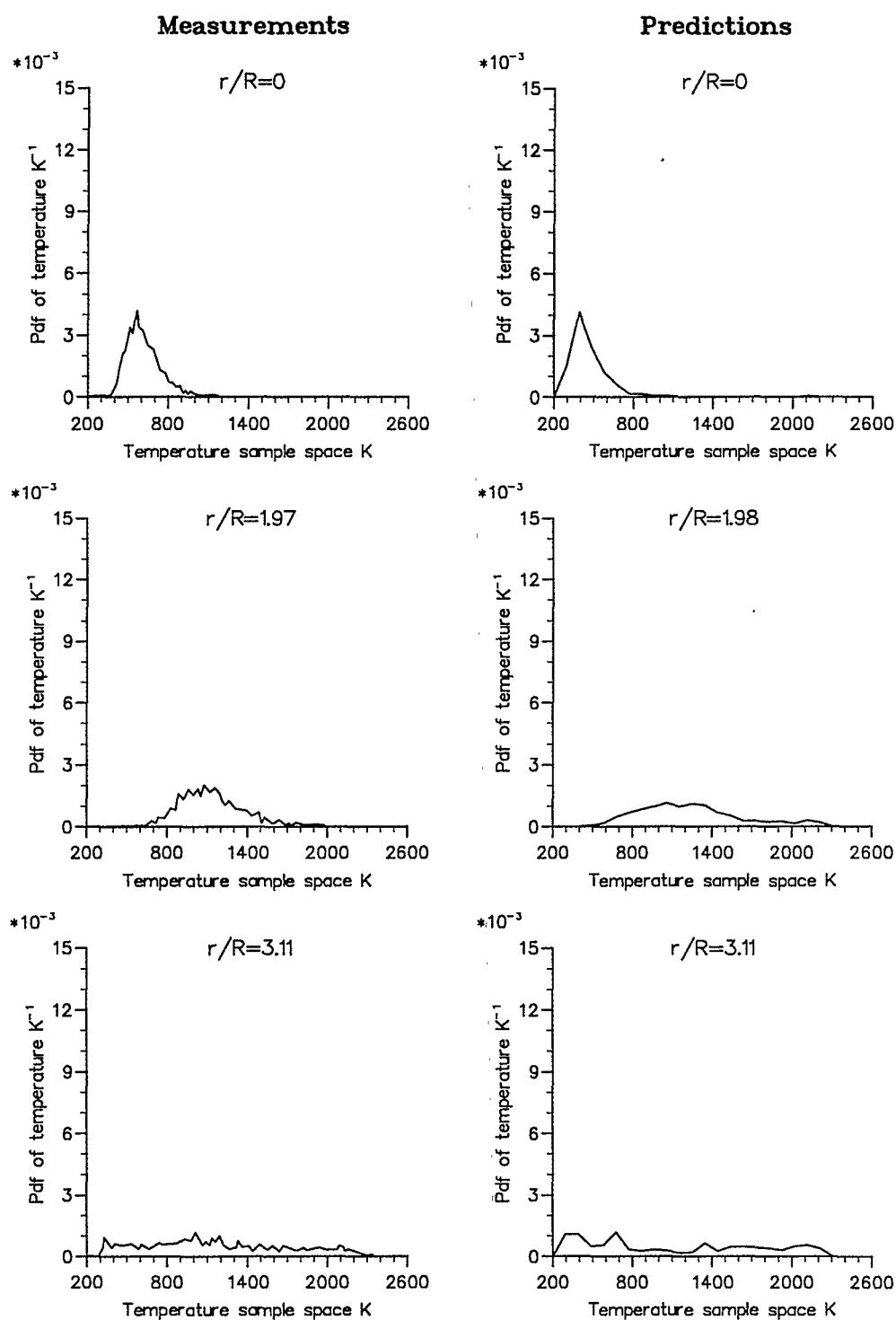


Figure 6.89: Pdf profiles at $x/D = 20$ of $\tilde{P}(\hat{T}; x, \omega)$ for the (L) flame of Masri et al. [67]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. $k-\varepsilon$ is used for the velocity field.

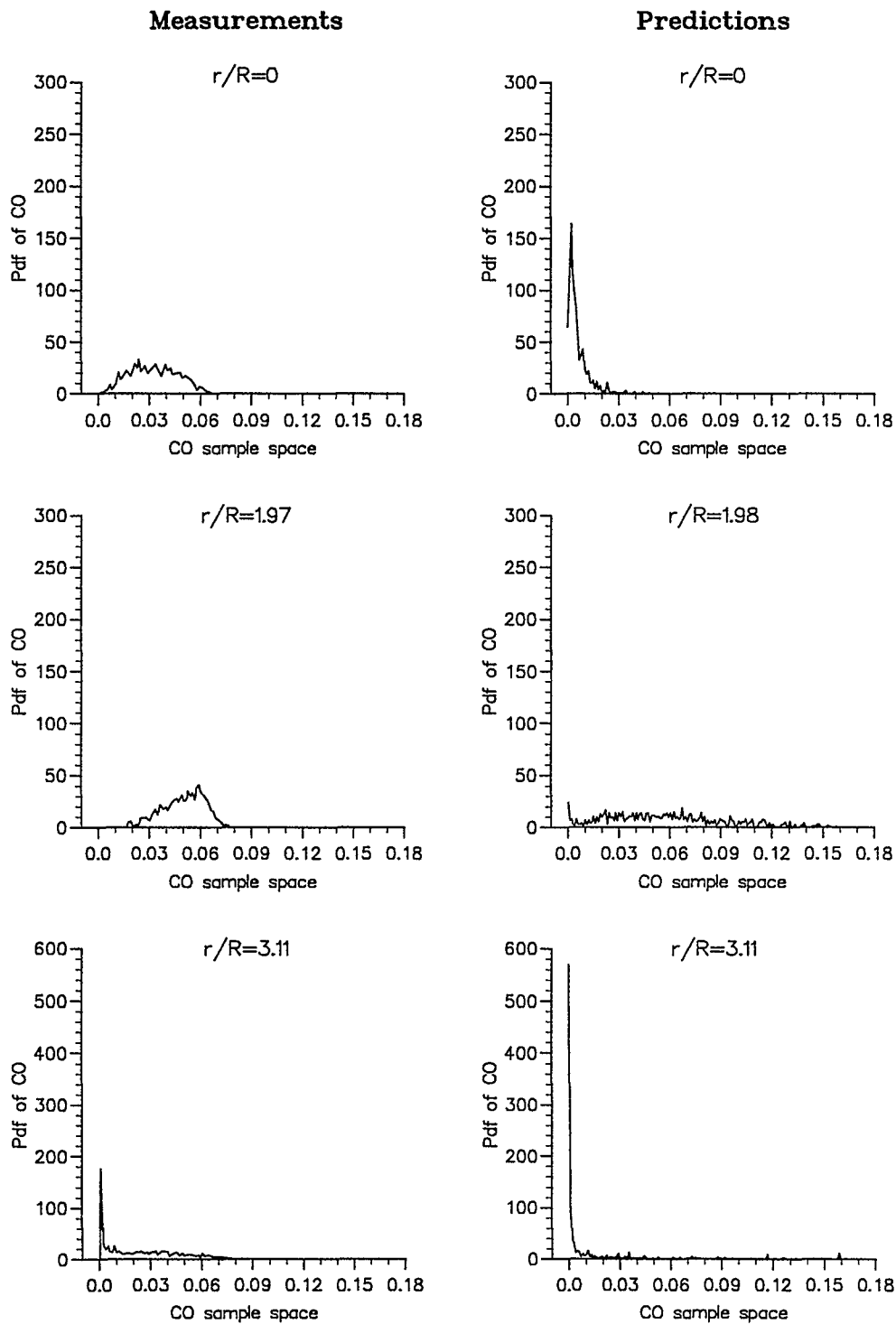


Figure 6.90: Pdf profiles at $x/D = 20$ of $\tilde{P}(\hat{Y}_{CO}; x, \omega)$ for the (L) flame of Masri et al. [67]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model. $k-\epsilon$ is used for the velocity field.

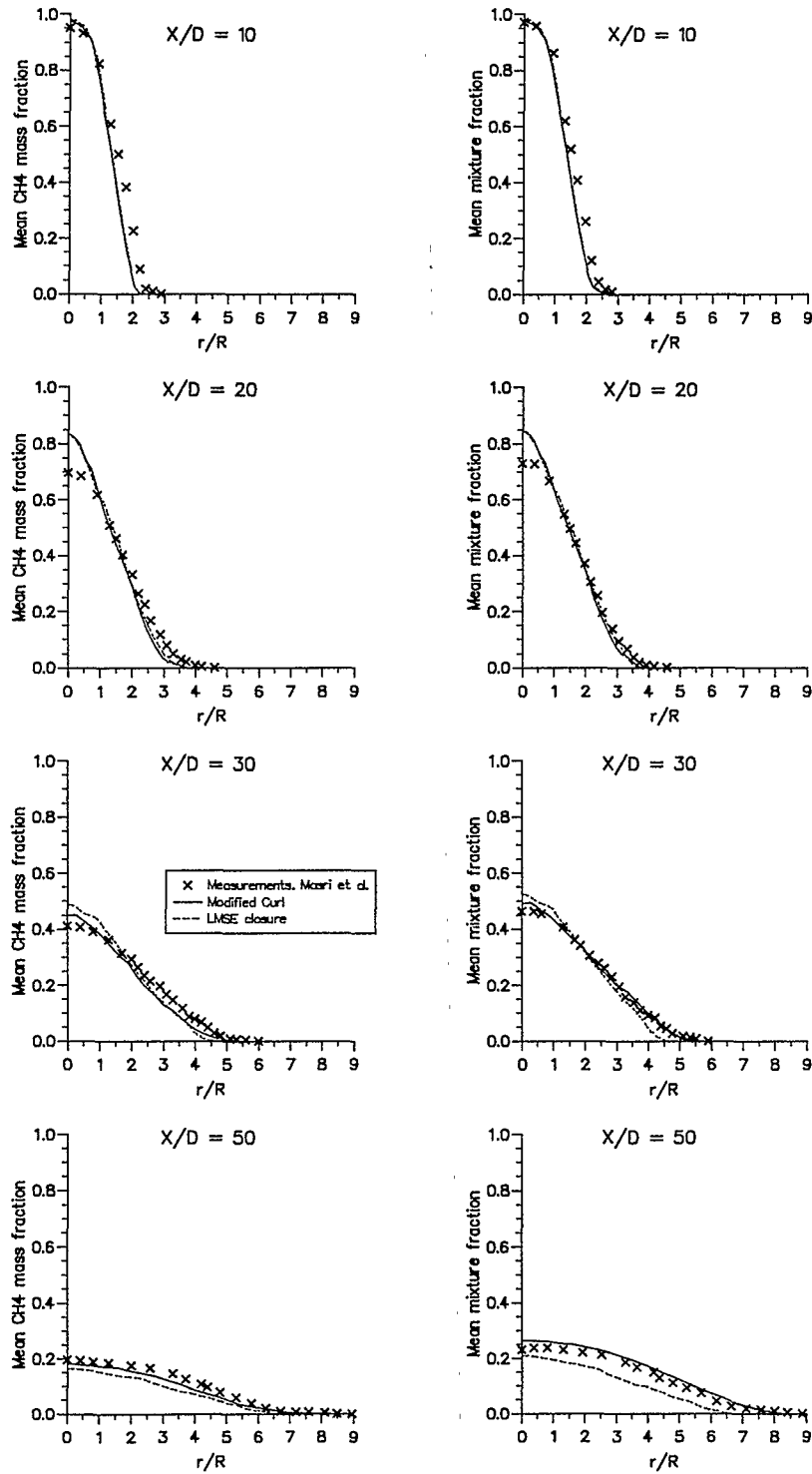


Figure 6.91: Radial profiles of $\bar{Y}_{\text{CH}_4}$ and $\bar{\xi}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.

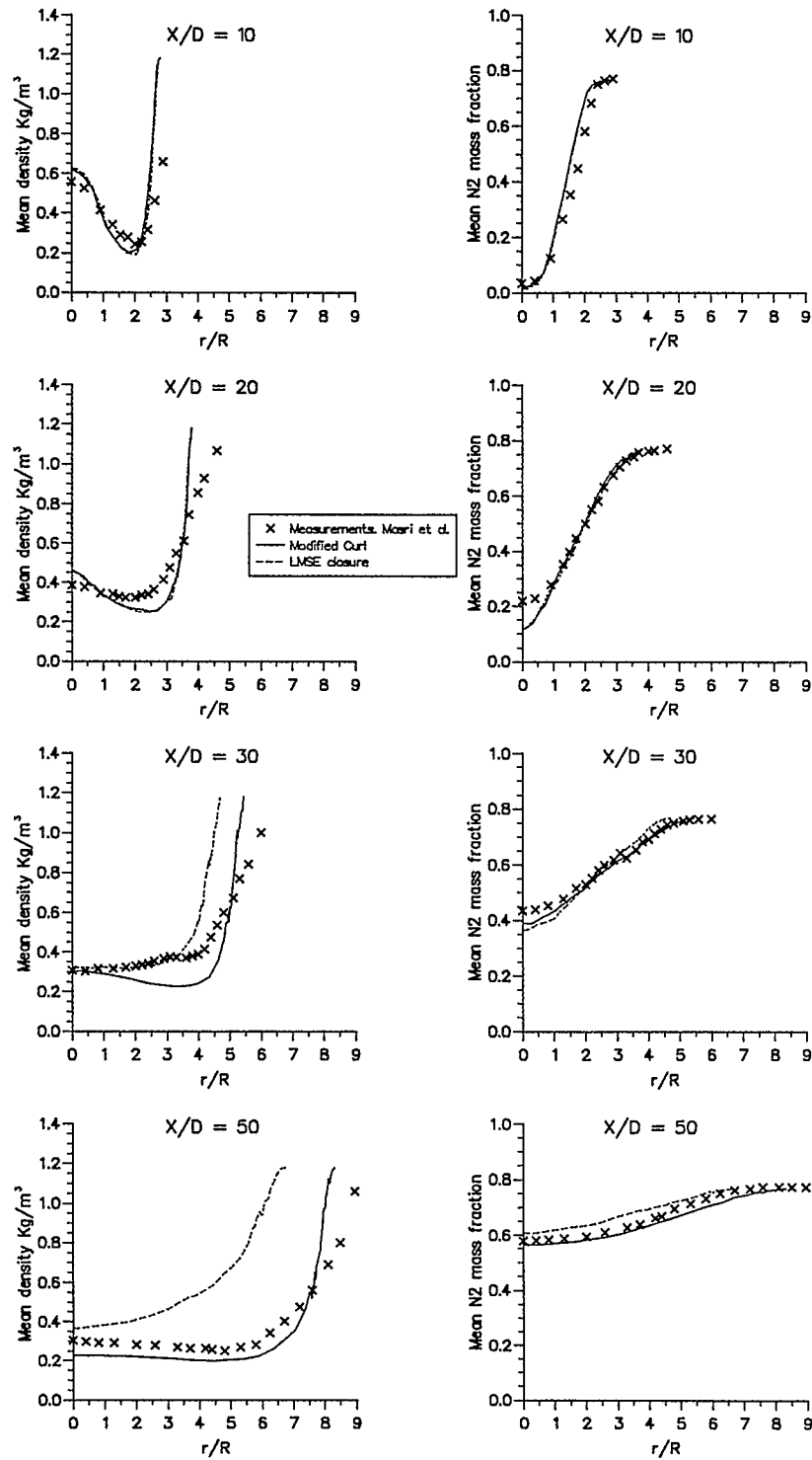


Figure 6.92: Radial profiles of $\bar{\rho}$ and $\bar{Y}_{N_2}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.

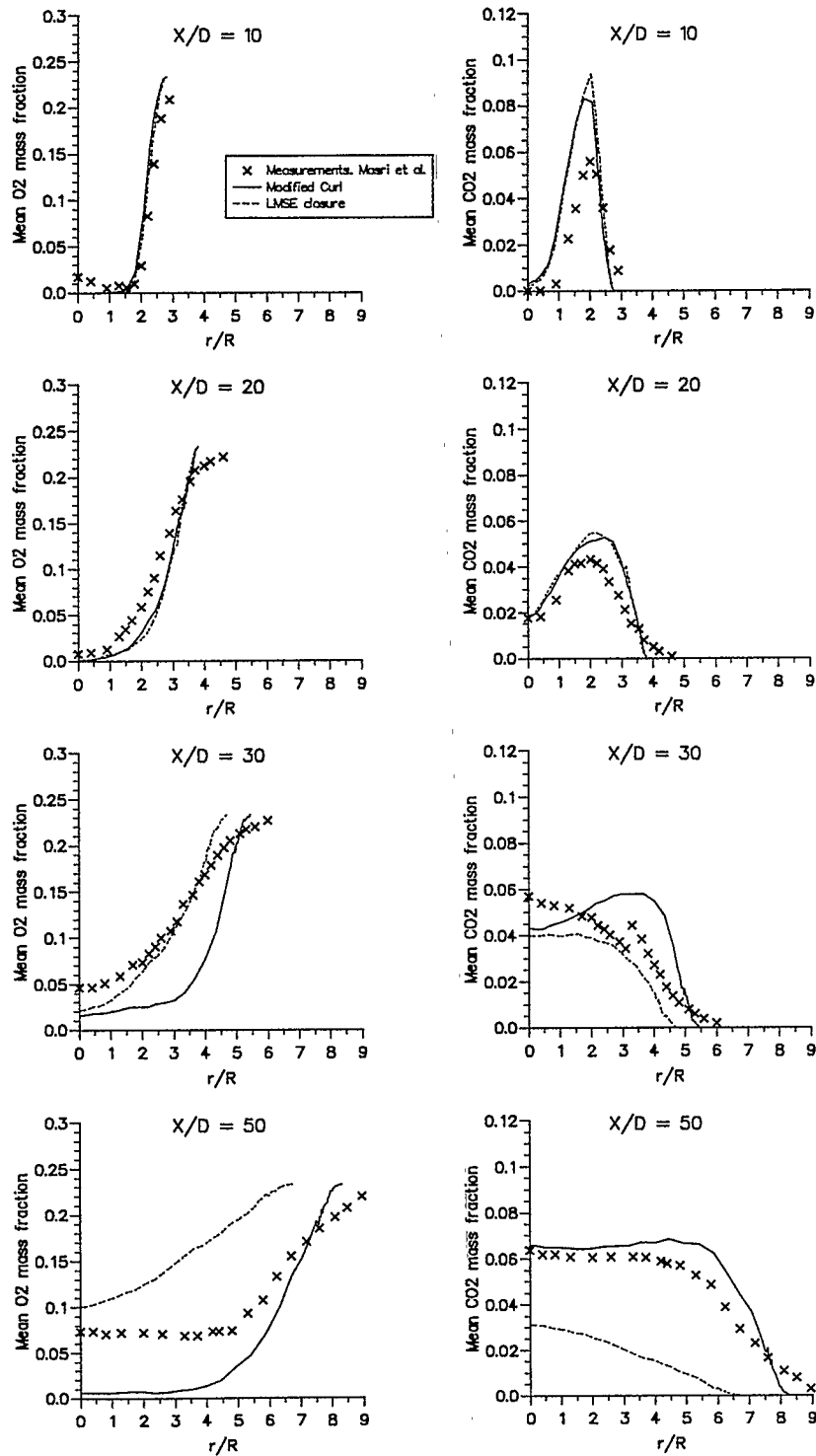


Figure 6.93: Radial profiles of $\bar{Y}_{O_2}$ and $\bar{Y}_{CO_2}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.

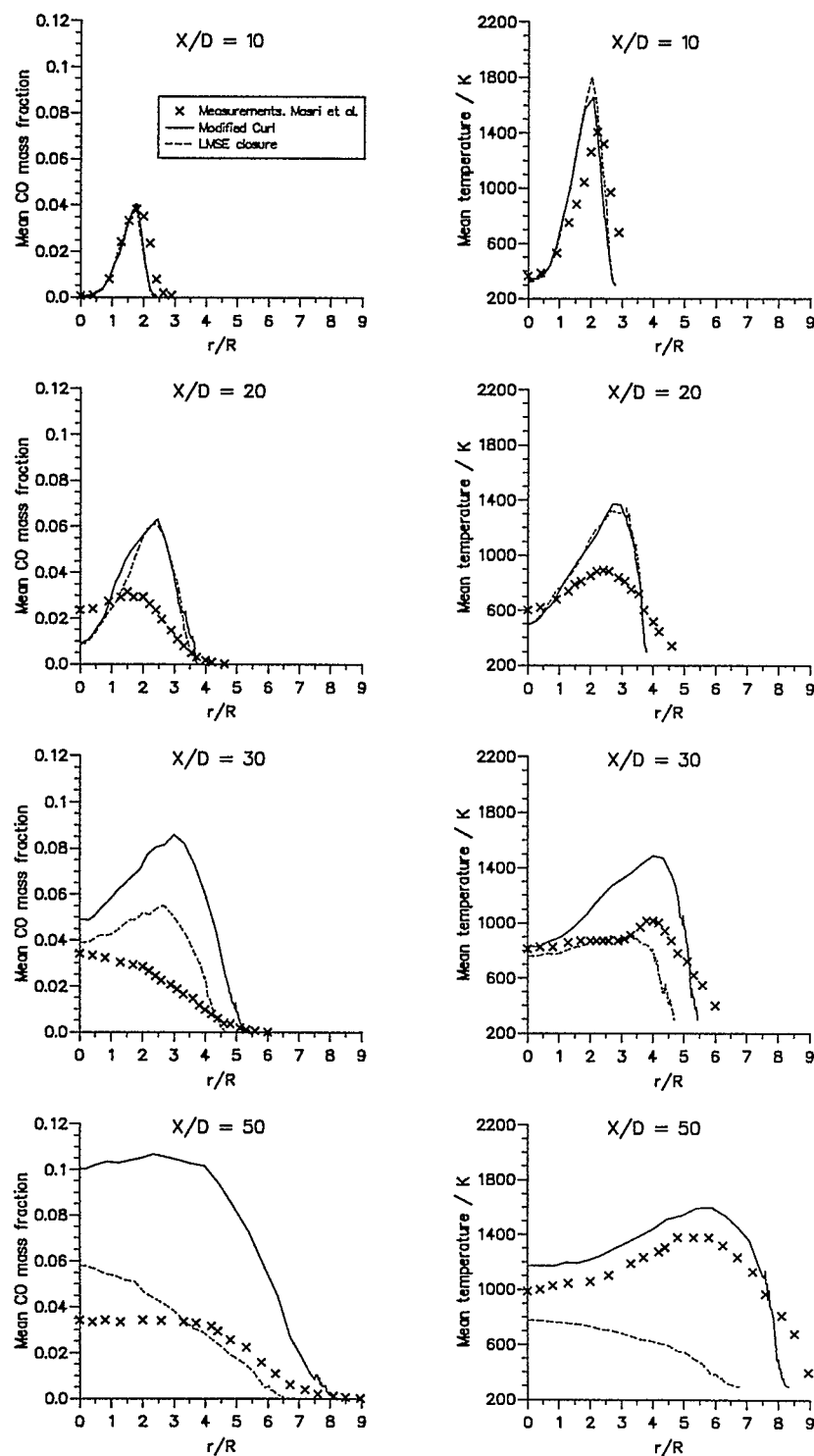


Figure 6.94: Radial profiles of $\bar{Y}_{CO}$ and $\bar{T}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.

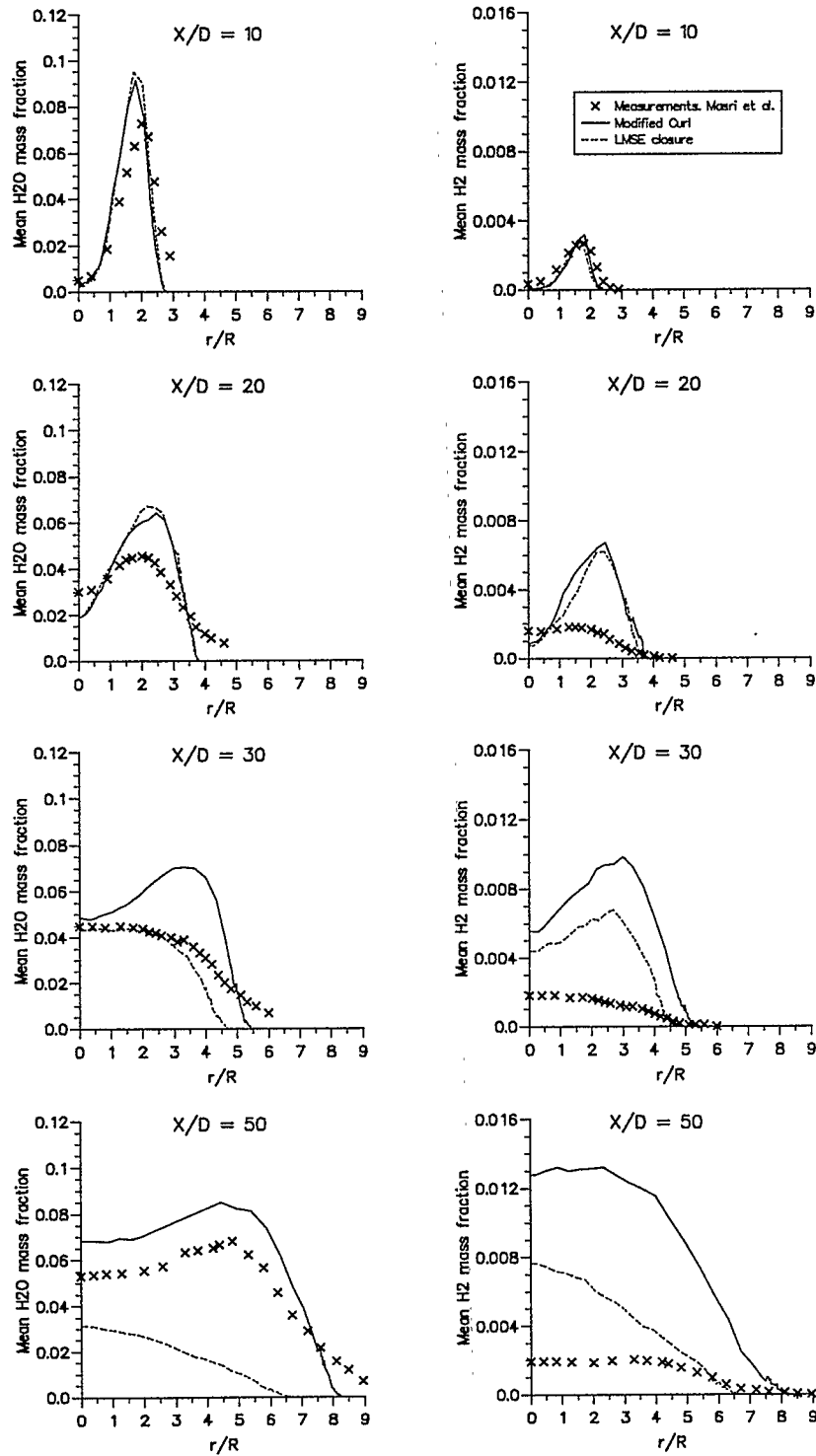


Figure 6.95: Radial profiles of $\tilde{Y}_{\text{H}_2\text{O}}$ and $\tilde{Y}_{\text{H}_2}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. $\tau = \frac{D}{2} = 3.6\text{mm}$.

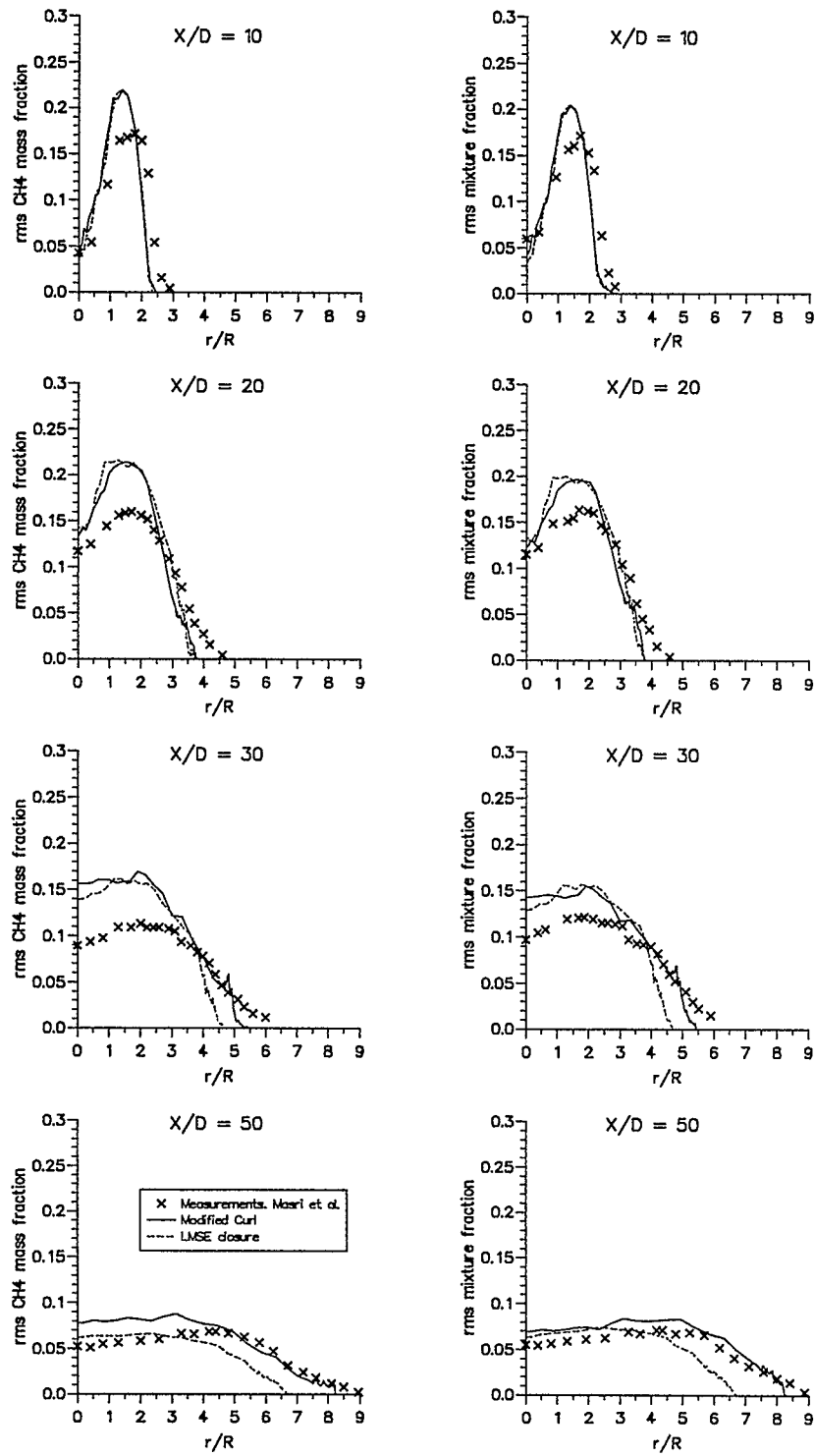


Figure 6.96: Radial profiles of $(\widetilde{Y_{CH_4}''^2})^{1/2}$ and $(\widetilde{\xi''^2})^{1/2}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.

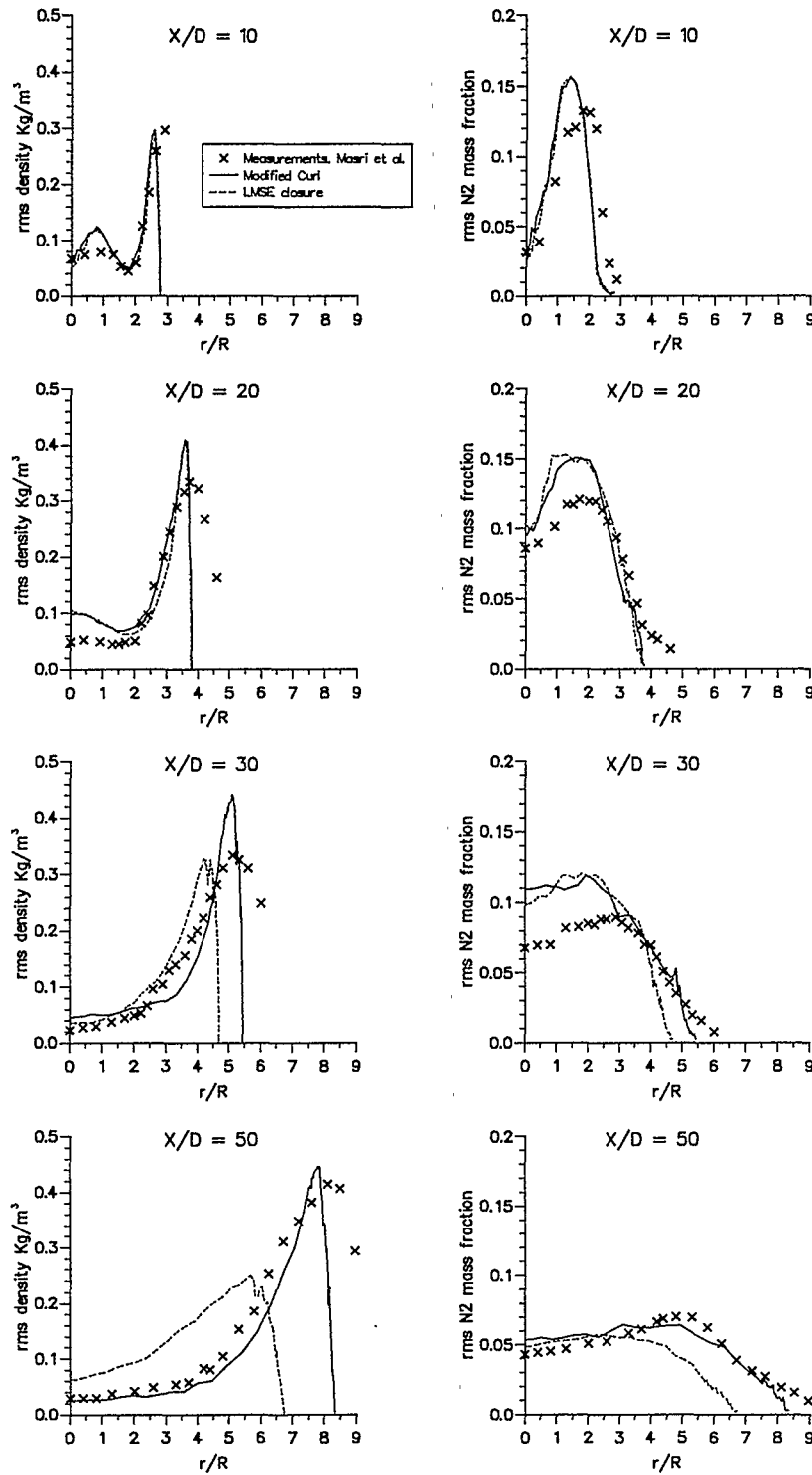


Figure 6.97: Radial profiles of $(\overline{\rho'^2})^{1/2}$ and $(\overline{Y_{N_2}'^2})^{1/2}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. $k-\epsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.

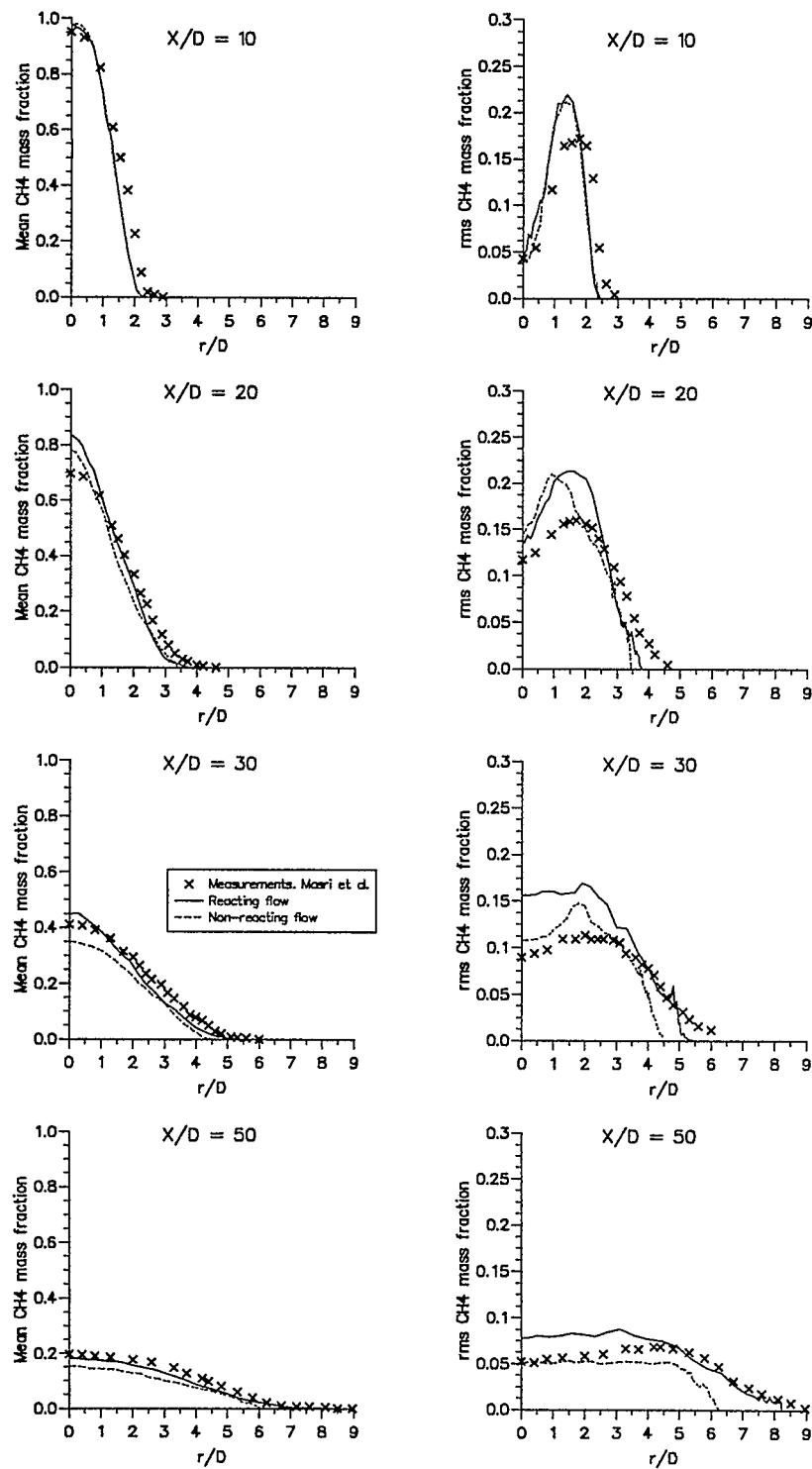


Figure 6.98: Radial profiles of $\tilde{Y}_{CH_4}$ and $(\tilde{Y}_{CH_4}''^2)^{1/2}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model (solid line) and this is compared with a test case where chemical reaction is not performed (dashed line). $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.

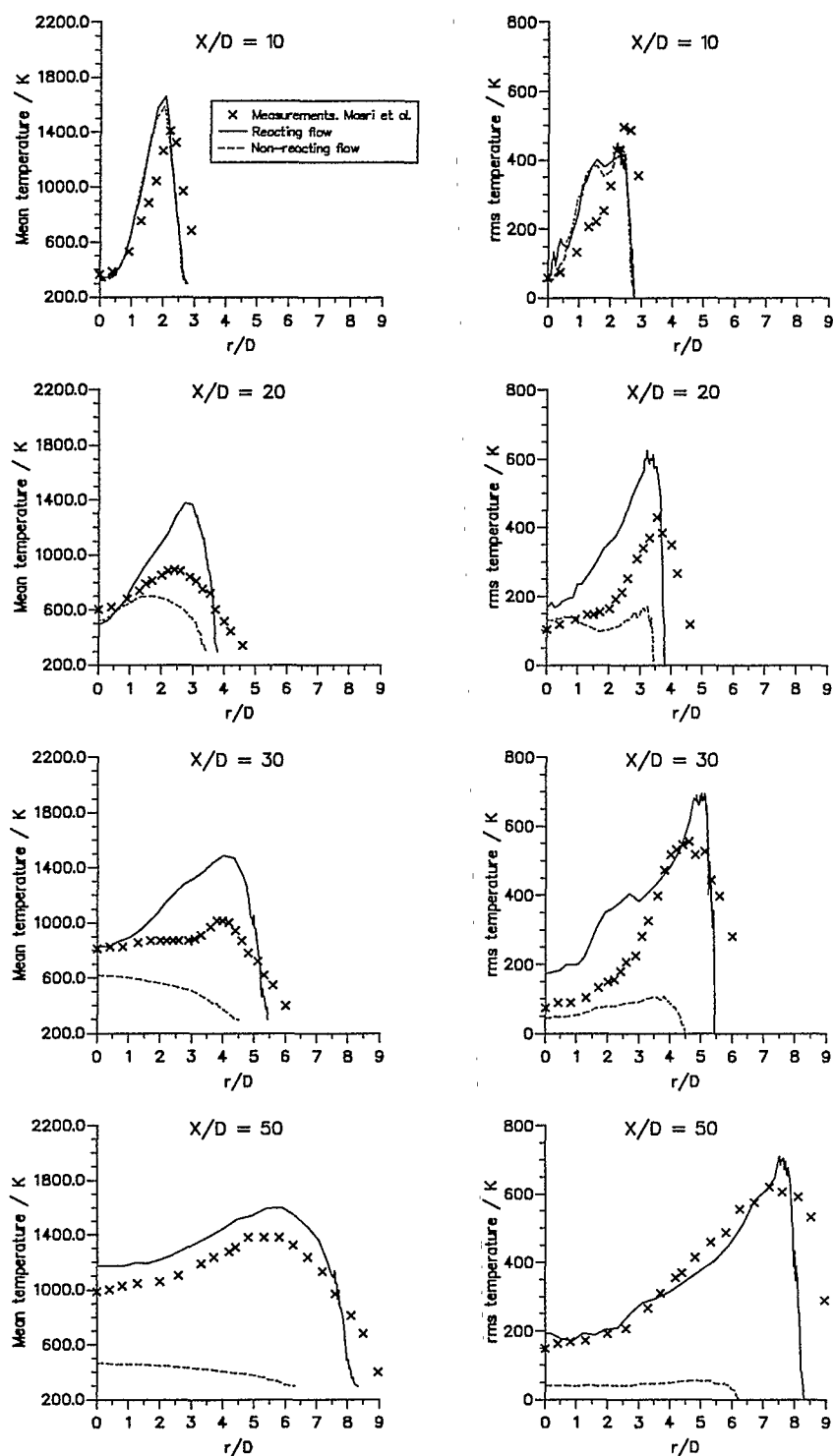


Figure 6.99: Radial profiles of $\bar{T}$ and $(\overline{T'^2})^{1/2}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model (solid line) and this is compared with a test case where chemical reaction is not performed (dashed line). $k-\varepsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.

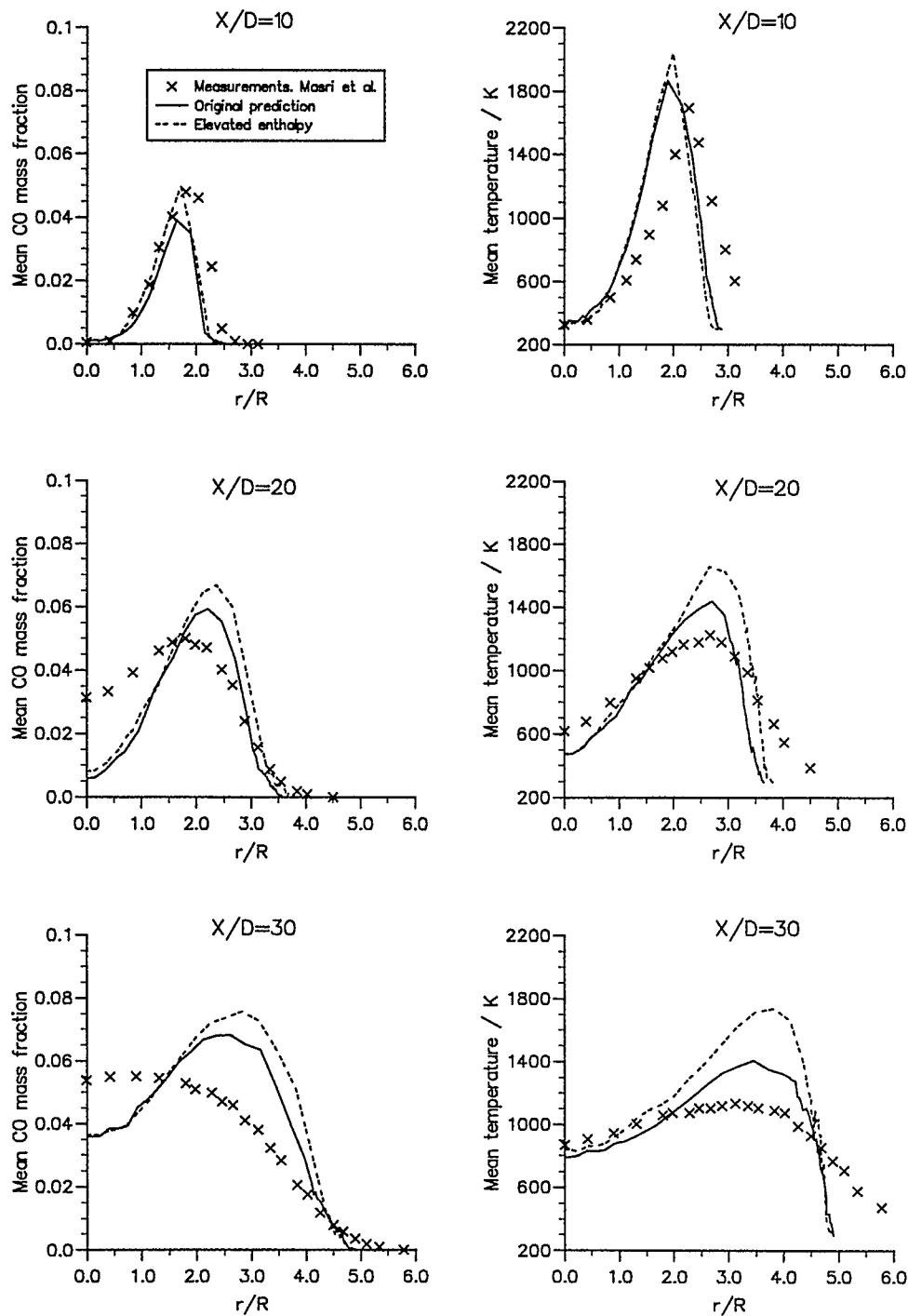


Figure 6.100: Radial profiles of $\bar{Y}_{CO}$ and $\bar{T}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl as the mixing model; the dashed line incorporates enthalpy as an extra scalar, and this is compared with the solution (corresponding to the solid line) presented in figure 6.80.

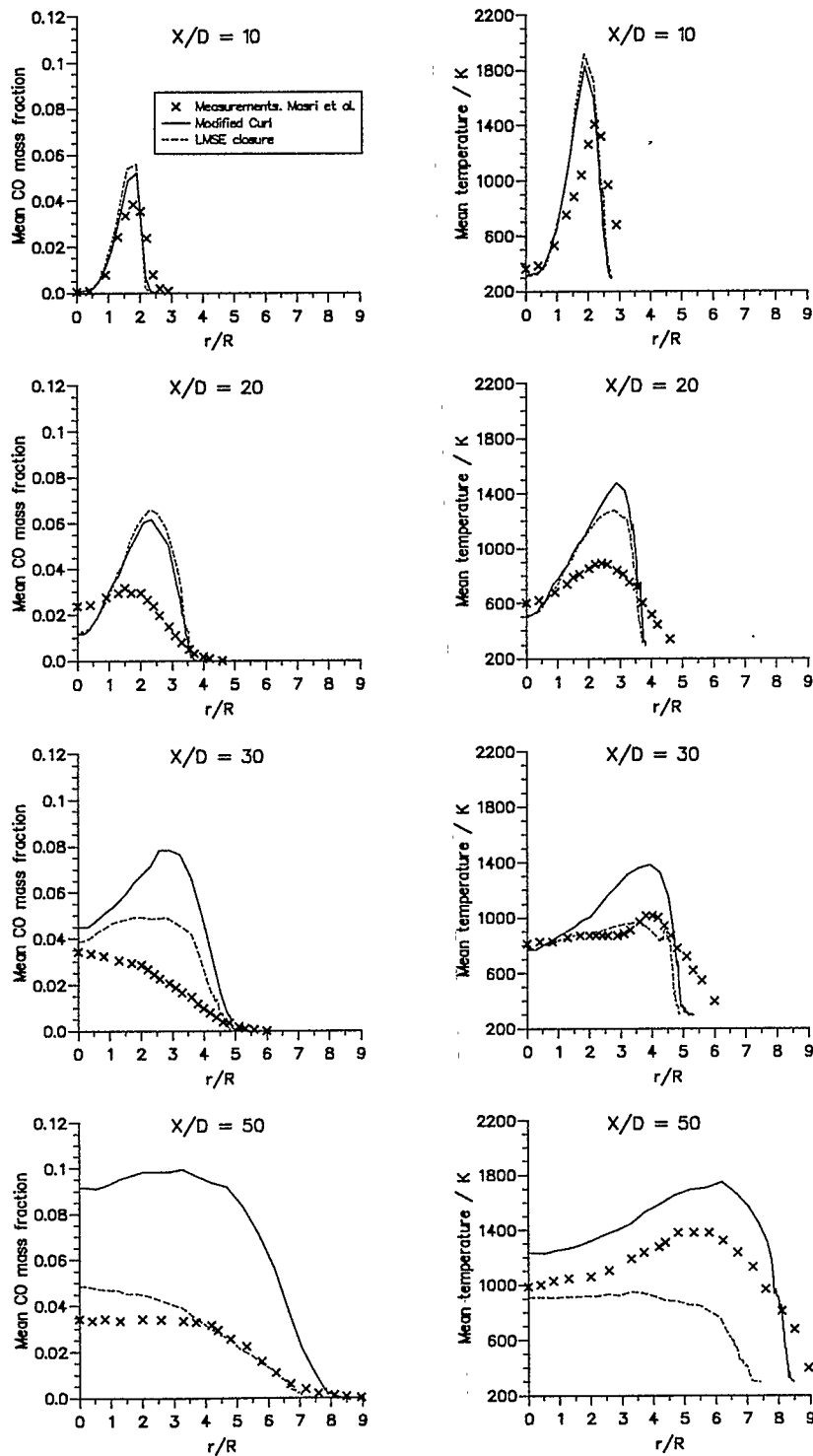


Figure 6.101: Radial profiles of $\bar{Y}_{CO}$ and $\bar{T}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. In these calculations, enthalpy is included as an extra scalar, and the result should be compared with figure 6.94.

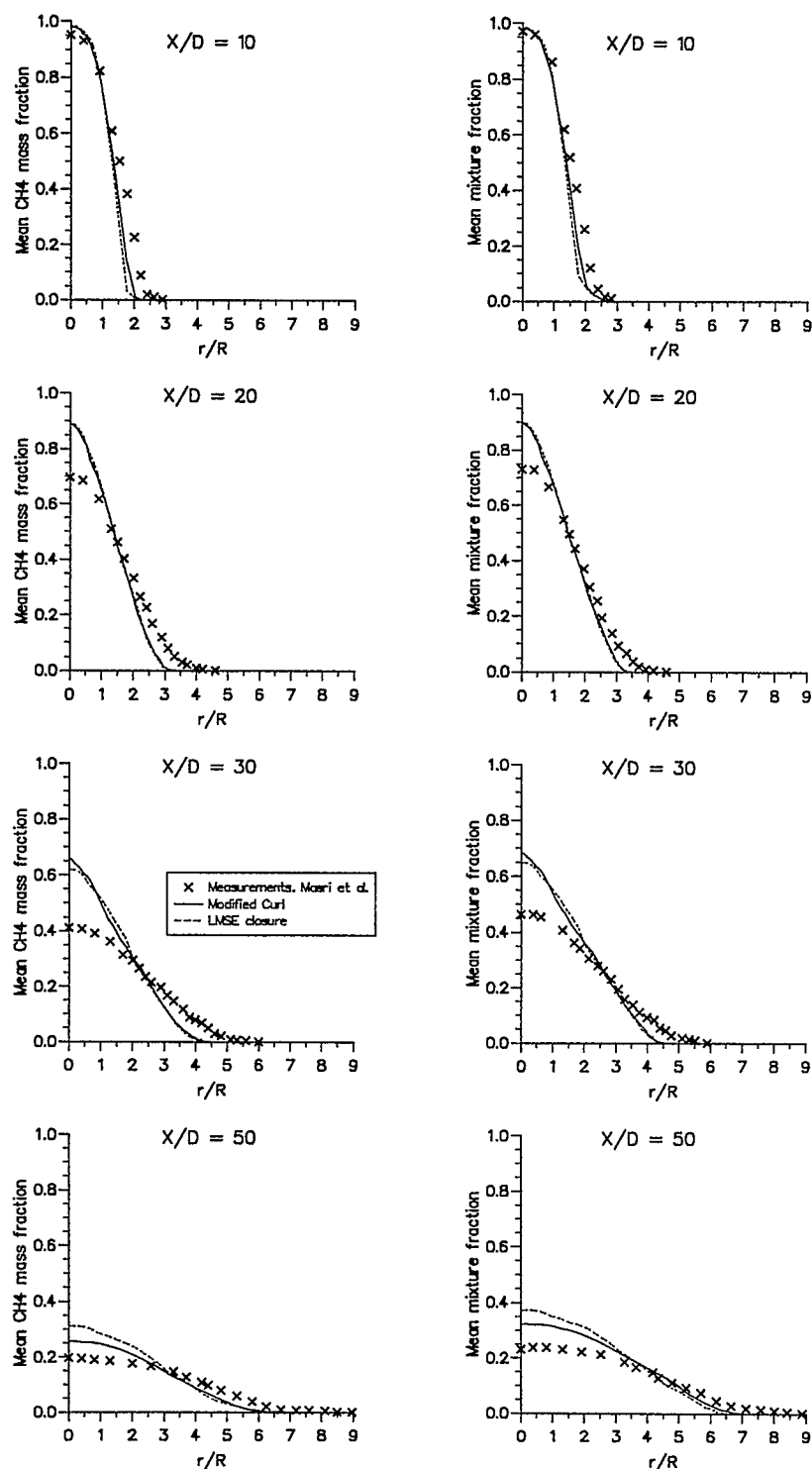


Figure 6.102: Radial profiles of $\bar{Y}_{CH_4}$ and $\bar{\xi}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. A Reynolds stress model is used for the velocity field, with $C_{\epsilon_2} = 1.8$. $r = \frac{D}{2} = 3.6\text{mm}$.

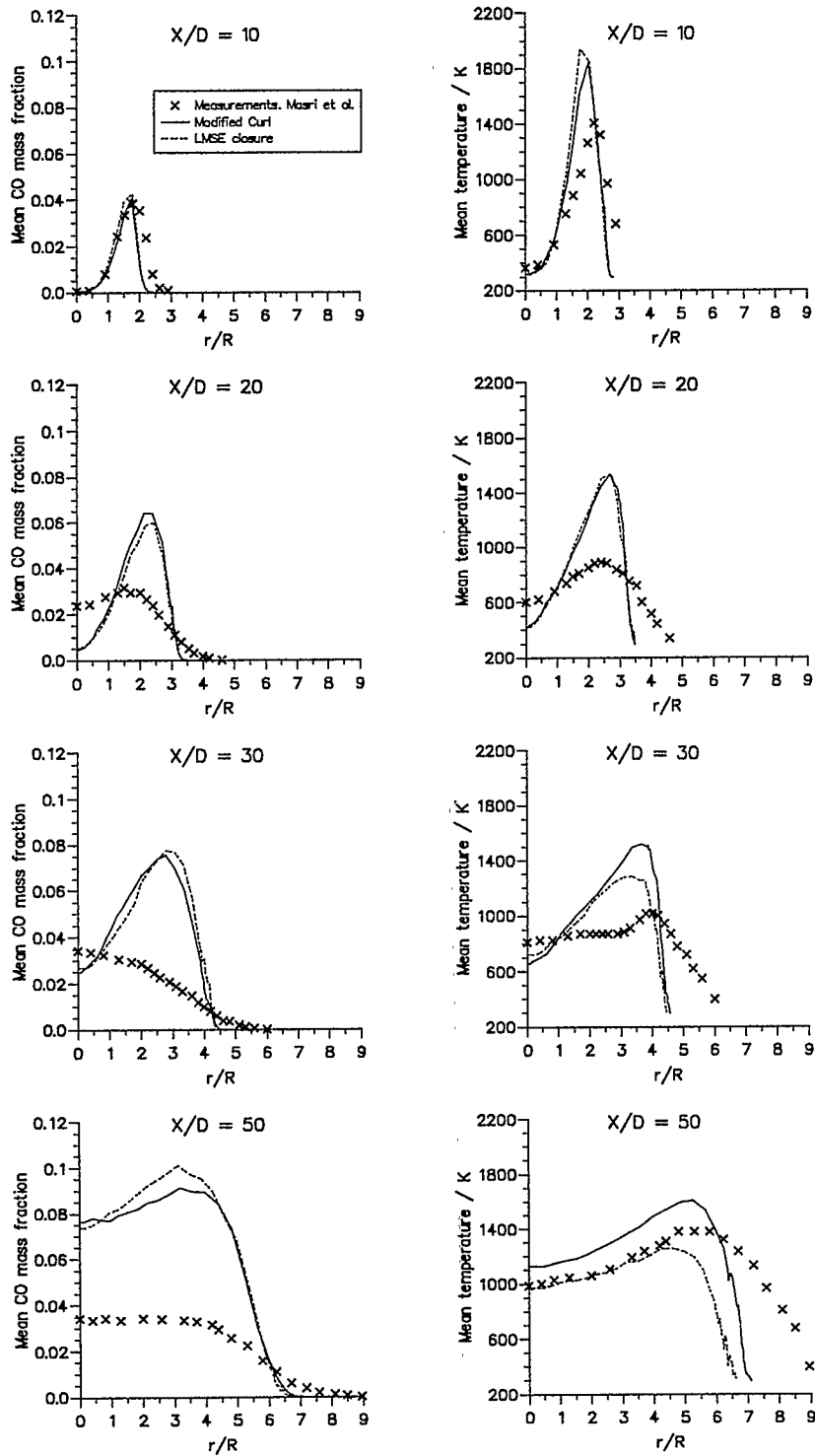


Figure 6.103: Radial profiles of $\bar{Y}_{CO}$ and $\bar{T}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with modified Curl and LMSE as the mixing models. A Reynolds stress model is used for the velocity field, with $C_{\epsilon_2} = 1.8$. $r = \frac{D}{2} = 3.6\text{mm}$.

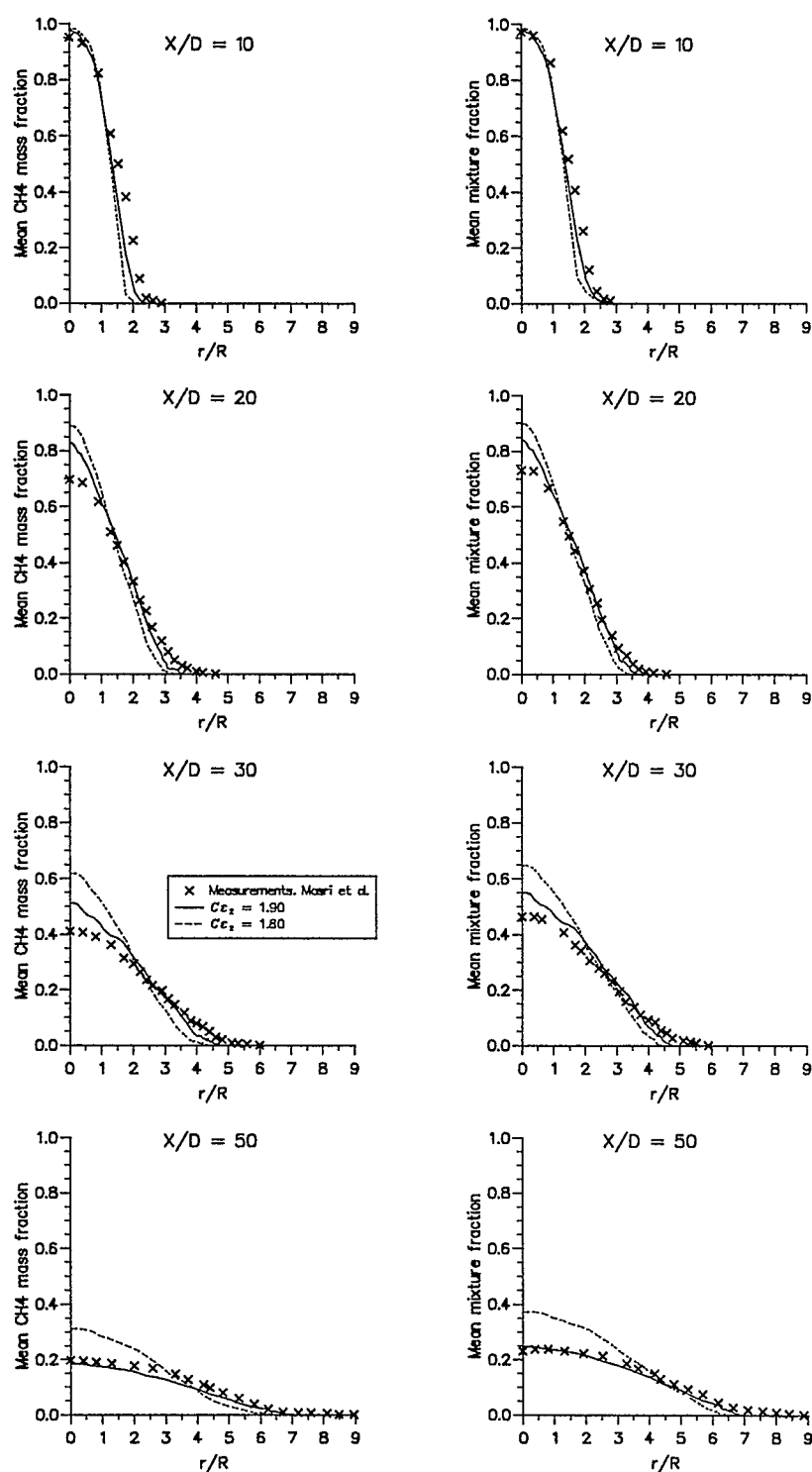


Figure 6.104: Radial profiles of $\bar{Y}_{CH_4}$ and $\bar{\xi}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with LMSE as the mixing model. A Reynolds stress model is used for the velocity field, with the solutions for $C_{\varepsilon_2} = 1.8$ (also shown in figure 6.102) and $C_{\varepsilon_2} = 1.9$ being compared. $r = \frac{D}{2} = 3.6\text{mm}$.

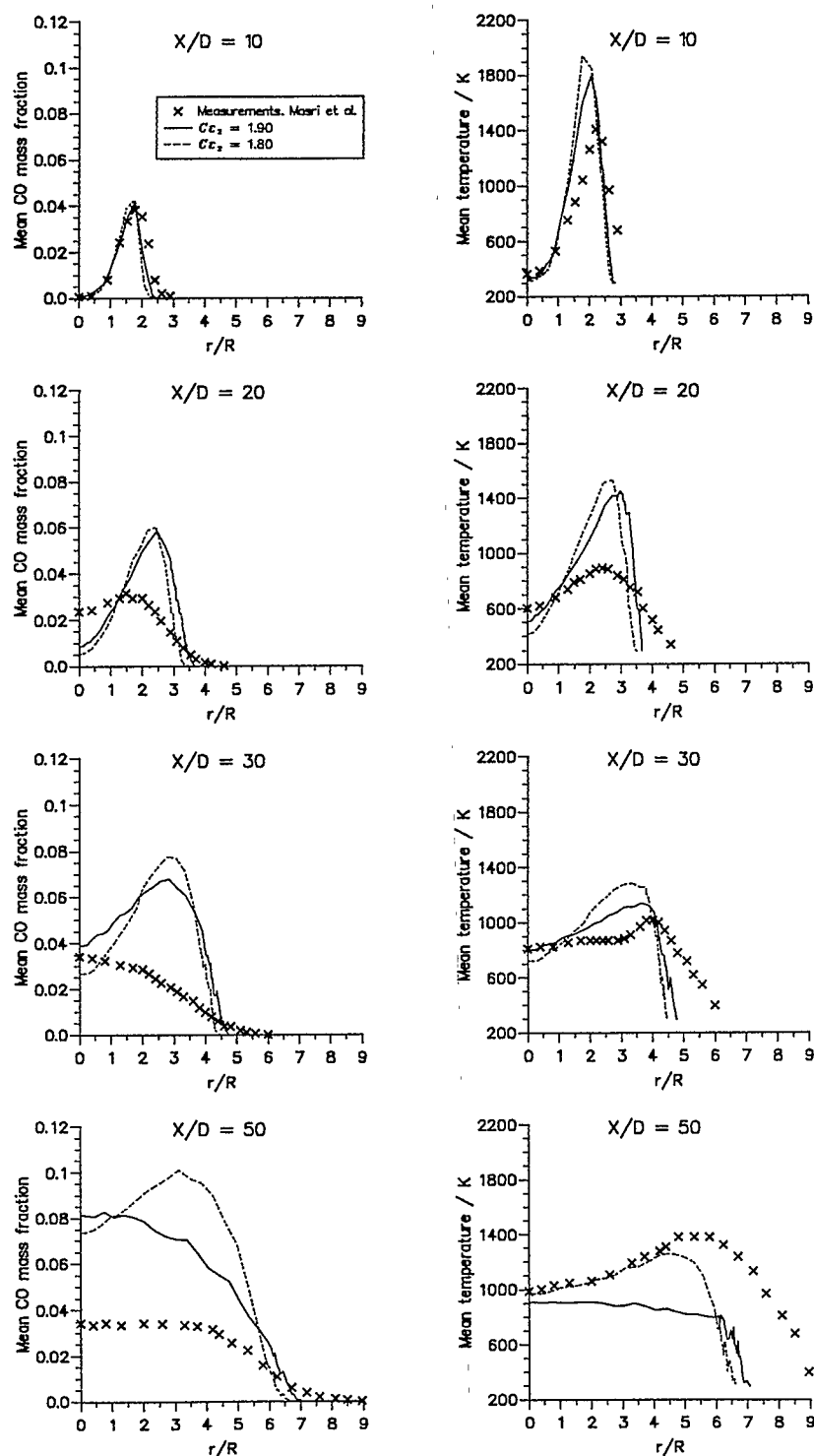


Figure 6.105: Radial profiles of $\bar{Y}_{CO}$ and $\bar{T}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with LMSE as the mixing model. A Reynolds stress model is used for the velocity field, with the solutions for $C_{\epsilon_2} = 1.8$ (also shown in figure 6.102) and $C_{\epsilon_2} = 1.9$ being compared. $r = \frac{D}{2} = 3.6\text{mm}$.

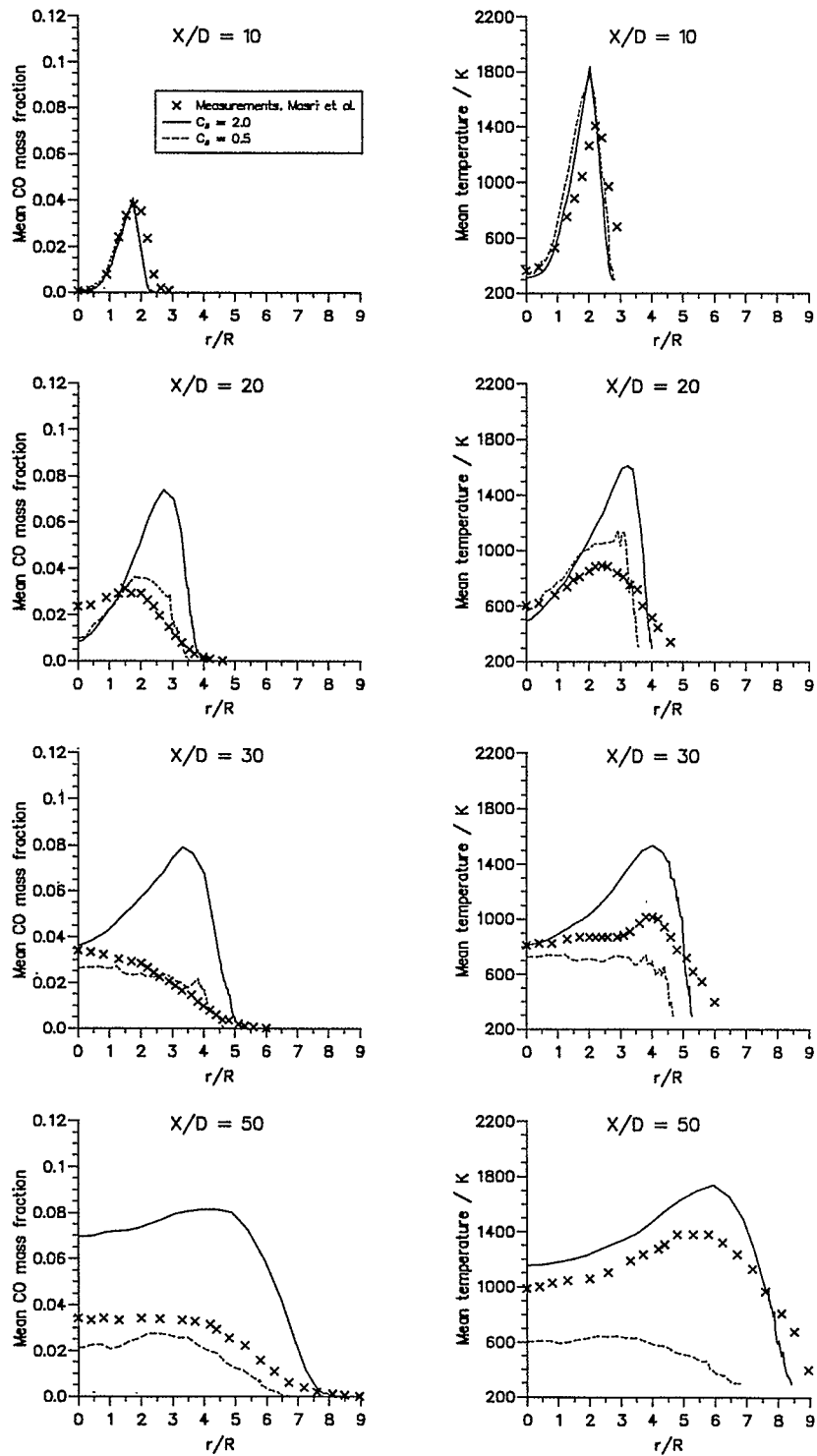


Figure 6.106: Radial profiles of $\bar{Y}_{CO}$ and $\bar{T}$ for the (B) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with LMSE as the mixing model. The effect of C_D is investigated. $k-\epsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.

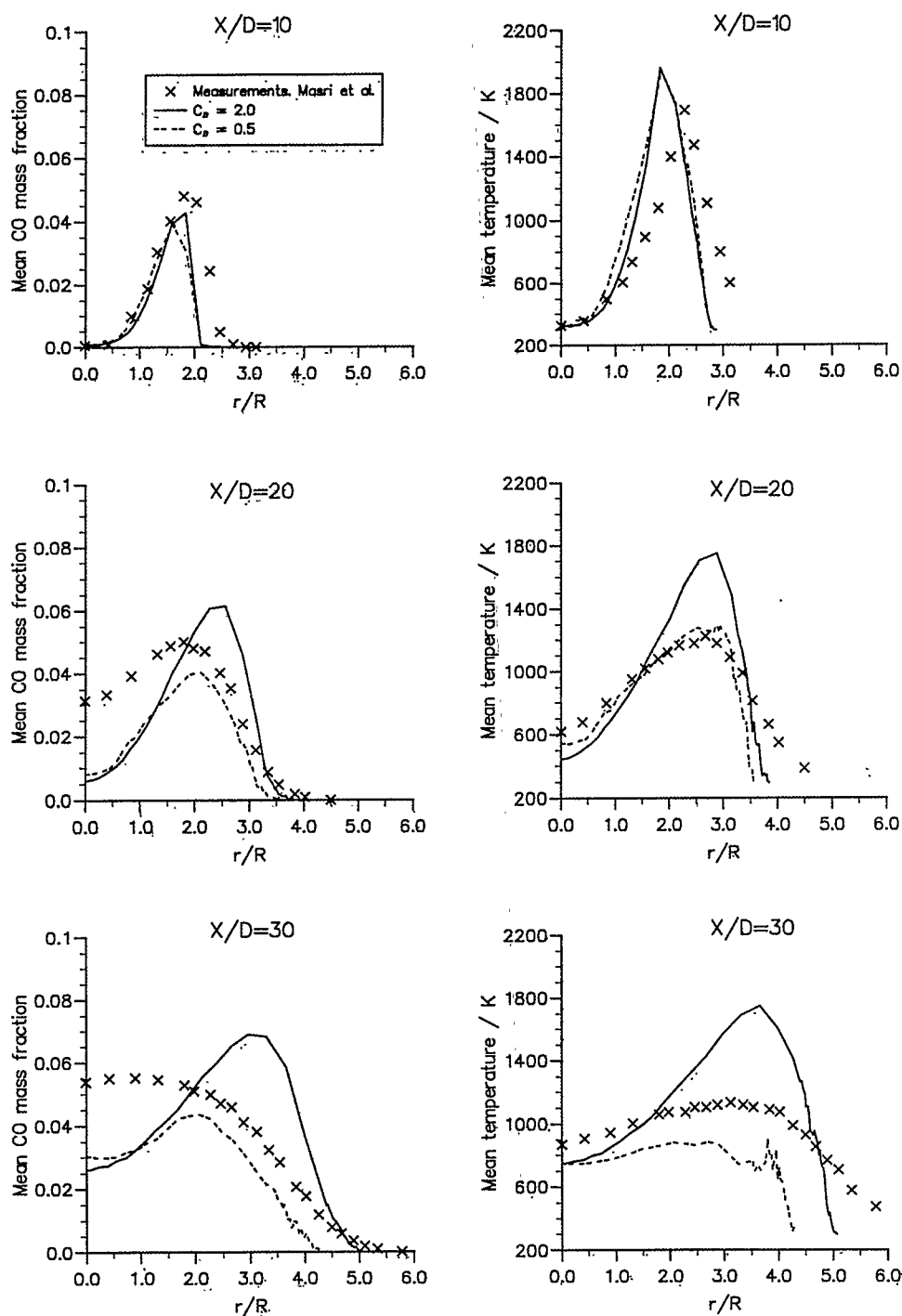


Figure 6.107: Radial profiles of $\bar{Y}_{CO}$ and $\bar{T}$ for the (L) flame of Masri et al. [66]. The reduced scheme of Jones and Lindstedt [46] is employed, with LMSE as the mixing model. The effect of C_D is investigated. $k-\epsilon$ is used for the velocity field. $r = \frac{D}{2} = 3.6\text{mm}$.