# ANALYSIS OF KINETIC MECHANISM PERFORMANCE IN CONDITIONAL MOMENT CLOSURE MODELLING OF TURBULENT, NON-PREMIXED METHANE FLAMES

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## ABSTRACT

Presented are results obtained from the application of a first-order Conditional Moment Closure (CMC) approach to the modelling of two methane flames of differing geometries. Predictions are based upon a second-moment turbulence and scalar-flux closure, and are supplemented with a wide range of full and reduced chemical kinetic mechanisms, ranging from a simple 12-step to a complex 325-step mechanism. Included with an analysis of the full scheme performances is an appraisal of the behaviour of their derivatives, obtained from mechanism reduction techniques. Despite extensive studies of the predictive ability of such schemes under laminar flame conditions, this is the first time, to the authors' knowledge, that a systematic evaluation has been performed for turbulent reacting flows.

The motivation of these works lies in the authors' current developments of higher-order elliptic CMC methods for application to practical combustion devices. As a precursor, the study presented was undertaken to analyse the practicality of incorporating kinetic models of varying complexity into calculation procedures, and to make comparison of their performance.

The findings of the paper reflect upon the impact that selection of chemical kinetics has upon subsequent calculations, and conclusions include that although the application of reduced schemes is more than adequate to reproduce experimentally derived data, the selection of the parent mechanism is of paramount importance to the prediction of minor species. Although most widely used schemes are very well documented and validated, their performances vary considerably. Thus, careful consideration must be made as to their application and origins during the evaluation of any combustion model.

## INTRODUCTION

The accurate representation of interactions between turbulent flow calculations and complex, finite-rate chemistry is essential for the design and analysis of modern industrial devices; aeronautical combustion chambers, gas turbines, or furnaces being three examples amongst many others. Many of these and other practical devices utilise the liquid injection of fuel, and these reasons provide the motivation for studying the non-premixed flames discussed herein. Demand for universally applicable, computationally efficient, and precise means of prediction is also continually increasing, especially when considering current pollutant emission regulation.

Several methodologies are available for modelling the aforementioned interactions, including the transported probability density function (PDF) approach and the conditional moment closure (CMC) method. The stochastic PDF method (Pope, 1985) provides a rigorous approach to the inclusion of finite-rate chemistry effects, although significant computing resources are required in using this technique. In contrast, the deterministic CMC approach (Klimenko and Bilger, 1999) provides a more economical method that can be readily integrated within computations of complex practical devices.

Although still in a relatively early developmental stage, the CMC method has proven to be a promising technique for predicting a wide range of practical problems. These include premixed and non-premixed combustion; relatively slow chemistry effects, and ignition and extinction phenomena. Parabolic formulations of the CMC equations have been successfully applied to simple diffusion flames of hydrogen (Barlow et al, 1999, Fairweather and Woolley, 2003a), carboxy (Roomina and Bilger, 1999) and hydrocarbon fuels (Roomina and Bilger, 2001). Difficulties in modelling NO in such flames have lead to higher-order studies being carried out on hydrogen flames (Kronenburg et al, 1998, Fairweather and Woolley, 2003b). Other recent work involving higher-order chemistry closure include a method for hydrocarbon fuels applied to piloted methane jet flames described by Kim and Huh (2004).

The results described herein are those obtained from first-order parabolic CMC calculations of $CH_4/H_2/N_2$ (Meier et al, 2000) and $CH_4$/air (Barlow and Frank, 1998) diffusion flames, calculated using three full chemical kinetic schemes alongside seven of their reduced forms. In light of previous works by the authors (Fairweather and Woolley, 2004) involving these flames, all calculations are carried out within a Reynolds stress/scalar flux (RSSF) turbulence-modelling framework.

Table 1: Kinetic Schemes Applied in The Study

| Number | Type | Scheme | Species | Steps |
|---|---|---|---|---|
| **1** | **Parent/Full** | **Miller-Bowman** | **46** | **224** |
| 2 | Reduced | Homma&Chen 14 | 18 | 14 |
| 3 | Reduced | Homma&Chen 16 | 20 | 16 |
| **4** | **Parent/Full** | **GRI-Mech2.11** | **49** | **277** |
| 5 | Reduced | Chen 12 | 16 | 12 |
| 6 | Reduced | Sung et al 15 | 19 | 15 |
| **7** | **Parent/Full** | **GRI-Mech3.0** | **53** | **325** |
| 8 | Reduced | Sung et al 13 | 17 | 13 |
| 9 | Reduced | Sung et al 15 | 19 | 15 |
| 10 | Reduced | Sung et al 17 | 21 | 17 |

## MATHEMATICAL MODEL

Solutions of the two-dimensional, axisymmetric forms of the density-weighted flow equations, supplemented with a second-moment (Jones and Musonge, 1988) turbulence closures, were used to obtain flow and mixing field predictions. Closure of the mean density term was achieved using a prescribed β-PDF, with instantaneous values of density derived from adiabatic, equilibrium calculations based on the Miller-Bowman mechanism (Miller and Bowman, 1989). Standard constants were employed in the turbulence models, apart from $C_{\varepsilon 1}$ that was modified between the standard and accepted value of 1.6 to improve spreading rate predictions. Mean and variance equations for mixture fraction were solved, with an improved version (Fairweather et al., 1992) of the original scalar flux model implemented for the second-moment closure employed. Solution of the transport equations was achieved using a modified version of the GENMIX code (Spalding, 1977) that employed expanding finite-difference meshes within a stream function formulation, and in all cases grid-independent solutions were established using resolutions in excess of one million nodes.

A first-order, parabolic CMC model was implemented, based on the set of equations that describe the production and transport of conditionally averaged species mass fractions and enthalpy (Klimenko and Bilger, 1999). For the jet flames modelled, the simplifying assumptions of negligible macro-transport by molecular diffusion and turbulent flux contributions were invoked (Klimenko and Bilger, 1999). Since jet flames display a large degree of radial independence of conditional statistics (Bilger, 1993), radial terms were evaluated using cross-stream averaged velocity and scalar dissipation values, as defined by Klimenko (1990). The conditional axial velocity appearing in the descriptive equations was modelled as a PDF-weighted, cross-stream-averaged value, with the approach of Girimaji (1992) used to represent the conditional scalar dissipation. Non-linear conditional source terms were approximated as for first-order closure, assuming the fluctuations of production rate around the mean to be negligible. Mean values were obtained using the CHEMKIN package (Kee et al., 1996) systematically employing the kinetic mechanisms depicted in Table 1. These include the scheme of Miller and Bowman (1989) and its reduced forms obtained by Homma and Chen (1999); GRI-Mech2.11 (Frenklach et al, 2004) and the derived forms of Chen (1997) and Sung et al (2001); and GRI-Mech3.0 (Smith et al, 2004) and the sub-mechanisms of Sung et al (2001). The conditional enthalpy equation was solved with the source term taken to be the conditional radiation heat loss, modelled using the optically thin assumption as outlined by Marracino and Lentini (1997).

Flow and mixing field information from turbulent flow calculations employing a reacting flow density were passed to the CMC model, where the set of species mass fraction equations plus the enthalpy equation were solved in mixture fraction space. Comparison between densities obtained from the CMC solution and prescribed equilibrium values showed little variation at the majority of locations examined in the flames considered. Coupling of the flow field and CMC calculations was therefore not performed for the calculations reported. Solution of the CMC equations in real space was achieved using a fractional step method, implemented using the stiff ODE solver VODE (Brown et al., 1989) which applies a backward differentiation formula approach to solution of the non-linear equation set. Second-order differential sample-space terms were determined using a central differencing approximation. In all cases, the spatial resolution was in excess of $3x10^3$ nodes.

Meier et al. (2000) considered two jet diffusion flames of $CH_4/H_2/N_2$, the flame under scrutiny here being designated Flame A. Fuel issued from a stainless steel tube with an inner diameter of 8 mm at a velocity of 42.2 m $s^{-1}$, with an air co-flow velocity of 0.3 m $s^{-1}$. Barlow and Frank (1998) considered three piloted turbulent $CH_4$/air diffusion flames, that used in this study being described as Flame D. The burner geometry consisted of an axisymmetric fuel jet, of diameter 7.2 mm, surrounded by a pilot annulus. Fuel issued from the central nozzle at 49.6 m $s^{-1}$, with an ambient air co-flow of 0.9 m $s^{-1}$. Flame A was considered to exhibit little extinction phenomena and some local extinction was noted in Flame D. In modelling these flames, inlet boundary conditions were prescribed from experimental data, and where data profiles were not available, initial conditions were obtained as prescribed by current turbulence theory.

## RESULTS AND DISCUSSION

Space restrictions preclude a detailed consideration of the accuracy of the velocity and mixing field predictions. However, in the case of both flames A and D, predictions of radial profiles of mean mixture fraction and its fluctuations, mean axial velocity and its fluctuations, and Reynolds stresses were in good agreement with data. A detailed appraisal of the methods used and results obtained can be found in Fairweather and Woolley (2004)

Figure 1 and Figure 4 portray measured and predicted major and minor conditional species mass fractions at one axial location in Flame A and Flame D respectively. Predicted data for both examples have been obtained using the Miller-Bowman mechanism (Miller and Bowman, 1989) and its two derivations (Homma and Chen, 1999). With respect to major species of flame A, all three models perform similarly in all instances excepting the temperature profile furthest downstream (not shown) which shows the reduced

mechanisms indicating a marginally lower conditional temperature. Results are generally very good, although slight under-predictions of $H_2O$ and over-predictions of $CO_2$ are evident over rich stoichiometries at near-nozzle locations (not shown), which become less prominent with increasing axial distance due to the reduced incidence of encountering fuel rich regions within the flame. In comparison, results for Flame D are less satisfactory and show a notable over-prediction of temperature at all fuel-rich mixture fractions in the near-field of the flame at x/d = 15 (not shown) and x/d = 30 (Figure 4). At the same locations, and in contrast to results for Flame A, $H_2O$ is now over-predicted, and $CO_2$ under-predicted, in fuel-rich regions, with $CH_4$ and $O_2$ also significantly under-predicted in these regions. The over prediction of temperatures, and under prediction of $CH_4$ and $O_2$, under fuel-rich conditions should lead to an over prediction of $CO_2$ and $H_2O$. This does occur for the latter species, but $CO_2$ appears to be very much in line with experimental data due to the overestimation of CO levels.

Turning to an analysis of the minor species predictions, all three schemes show very similar results regarding CO in both flames. A slight over-prediction is observed by all models in fuel-rich regions at x/d = 10 in Flame A (not shown), which leads to a slight under-prediction of peak values further down the flames length. The two reduced schemes digress from the path of their parent slightly at x/d = 40 (not shown), predicting a slightly lower peak value. Conversely, data obtained for Flame D see an over-prediction made by all models at peak and rich stoichiometries, the reduced schemes showing slightly higher values than their parent in the rich regions. Regarding the nitrogenous species, all three kinetics schemes perform similarly in their predictions of NO for the burner-stable flame, although where the 16-step mechanism follows the behaviour of the parent, the 14-step predicts slightly higher NO in the fuel-rich regions whilst predicting slightly lower NO in the fuel-lean. By x/d = 40 (not shown), a much lower prediction is observable across most of mixture fraction space, and the peak is seen to move slightly further towards the richer stoichiometries. Similar observations can be made of results obtained for Flame D, although effects appear greatly accentuated. A general over-prediction of NO is now seen from the Miller-Bowman and 16-step reduced scheme, and more notable deviations can be seen in the 14-step data. This under-prediction by the 14-step mechanism conforms with the findings of the authors' investigations (Homma and Chen, 1999) using a $CH_4$-air opposed diffusion flame experiment, although the slight shift of the peak concentration in mixture fraction space is an anomaly of the current study. As observed by Homma and Chen (1999), and verified in these works, the 14-step mechanism is unable to predict prompt NO formation accurately due to the assumption of quasi-steady-state of the HCN molecule. With respect to $NO_2$ predictions (not shown), and in line with the previous work (Homma and Chen, 1999), the 14-step mechanism was seen to under-predict levels in comparison to its counterpart reduced scheme and the parent, which perform very similarly (not shown). In addition to this, the 14-step scheme notably over-predicts $NO_2$ in comparison in the rich-region peak of mixture fraction space of Flame D. Again, this behaviour in $NO_2$ predictions can be attributed to the assumptions made regarding HCN during the model construction. With mention of OH radical predictions, all three models perform in a similar manner throughout the lengths of the two flames, displaying a substantial over-prediction. The smaller of the reduced schemes appears to deviate from the others' results marginally with downstream progression.

Turning to an analysis of GRI-Mech2.11 and its 12- and 15-step derivatives, Figures 2 and 5 depict the major and minor species predictions plotted against experimental data at x/d = 20 and x/d = 30 for Flames A and D respectively. The behaviour of the parent scheme is very similar to that of the Miller-Bowman mechanism with similar trends observed in both flames. Little discerns between the two models in the prediction of major species, but Miller-Bowman displays a slight improvement in the prediction of NO at fuel-lean and peak mixture fractions. This manifests as a lowering of predicted levels, and a similar effect is observable in $NO_2$ results (not shown). However, GRI-Mech2.11 reports a notably smaller CO peak and subsequently a reduced fuel-rich region prediction. This aids to improve conformity with experimental data in calculations of Flame D, but has the converse effect in Flame A. As with the major species, OH radical levels show little difference in the magnitude of their predictions. Considering the performance of the reduced mechanisms of GRI-Mech2.11, both the 12- and the 15-step schemes provide almost identical results for all species and temperature in both flames. These profiles follow those of the parent mechanism closely, although they do display a slightly lower prediction of CO levels at fuel-rich stoichiometries in Flames A and D. They also show a marginally lower fuel-lean and peak prediction in Flame D, and marginally higher fuel-rich prediction in Flame A, with respect to NO.

Figures 3 and 6 show the results of calculations carried out using GRI-Mech3.0 and its three reduced forms comprising 13-, 15-, and 17-step schemes of Flames A and D respectively. An initial inspection of major species and temperature indicates no observable difference between the two GRI-Mech issues. Inspection of the minor species does however reveal some differences in behaviour. Peak and fuel-rich regions of CO predictions by GRI-Mech3.0 prove to be lesser in magnitude than their counterpart, which leads to an improvement in predictions in Flame D, but a slight deterioration in results for Flame A. The most striking observable differences are in the NO and $NO_2$ (not shown) plots which show predicted levels by GRI-Mech3.0 to be between two and three times greater than those of the earlier issue. The reasons for this behaviour are unclear at present, although observations do indicate a number of possible sources of error in the preparation of both schemes in a mechanistic and a validatory sense. A number of recorded applications of the schemes exist in the literature, which report their relative performances based upon a number of differing criteria, and all indicate a superior performance of GRI-Mech3.0 over its predecessor. Although the optimisation procedure involved in GRI-Mech3.0 appears to produce superior results in a number of analyses, likely being due to new data being included in the targets, the more recent mechanism does not accurately describe NO concentrations in the atmospheric diffusion flames considered in this work. Comment is made of this behaviour in the optimisation notes of the authors (Smith et al, 2004),

indicating that the mechanism over-predicts NO re-burn in reactor experiments but under-predicts re-burn in low pressure flames. It is suggested therein, that either experimental discrepancies exist or some mechanistic problem remains. Observed in (Sung et al, 2001), GRI-Mech3.0 does not contain reactions of $NH_2$ + NO which lead to the thermal de-$NO_x$ processes. This may be some indication as to the differing performances of the models, although additional studies by this manuscript's authors of other full mechanisms attributed to Konnov (2004) and Warnatz et al (2001), which both contain routes to such processes, also display a similar over-prediction of NO concentrations as GRI-Mech3.0.

## CONCLUSIONS

A comprehensive study of the interaction of a number of commonly used kinetic mechanisms with the parabolic CMC model applied to two methane flames of differing geometries has been presented.

Of the selected schemes applied, those of Miller-Bowman and its derivatives perform in the most conforming manner to the experimental data. Although the kinetic models describe major species and temperature to a similar degree of accuracy, the aforementioned provide superior predictions of minors such as CO and NO also.

The reduced schemes derived from the larger parent schemes reproduce the predictions made by the latter to a very high degree of accuracy. The findings of this work therefore include that reduced schemes of well validated larger mechanisms are suitable for inclusion in further studies which may require more computationally expensive procedures. This being the case with the elliptic and higher-order hydrocarbon combustion models recently developed by the authors, using the 16-step reduced scheme of Homma and Chen (1999), the results currently being prepared for publication.

It is apparent that the complexity of a kinetic model is not indicative of its ability to reproduce experimental data. Of the full schemes investigated, the smallest of the three proffers the most accurate results, as indeed do its derivate schemes. What does appear to have a great bearing upon their performance is the data used in the calibration and validation of the schemes. This is perhaps highlighted in the comparative behaviours of the two GRI-Mech models.

Although sound validation of the individual schemes studied here exist, the variation of their constituents such as rate parameters and elemental reactions is sufficient to give results of minor species such as NO which diverge from experimental data and each other by as much as a factor of four. Hence, the analysis of a combustion model performance can be subject to the effects of the kinetic scheme applied. The present study demonstrates that the CMC model has difficulty reproducing experimentally derived NO data using GRI-Mech3.0. However, use of GRI-Mech2.11, Miller-Bowman and the schemes' derivatives indicate that the CMC model can predict these minor species very accurately. The Miller-Bowman mechanism succeeds in a relatively good representation of CO profiles also.

It is concluded that results obtained from the CMC model, and indeed other combustion models, are heavily reliant upon the kinetic scheme used in its application. Simple analysis of the magnitude of the terms which construct the CMC equation demonstrates the dominance of the chemical source term in such calculations. Consequentially, great care needs to be taken during the analysis of overall combustion model performance to the consideration of the kinetics being applied.

Finally, first-order CMC is capable of yielding reliable predictions for methane flames displaying little or some local extinction. Contrary to previous experience and observations (Fairweather and Woolley, 2004) which limit the model's reliability to flows displaying little or no extinction effects, it is evident that careful consideration of the kinetics applied to the calculations considerably improves the range of flows over which the first-order model is valid.

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## FIGURES

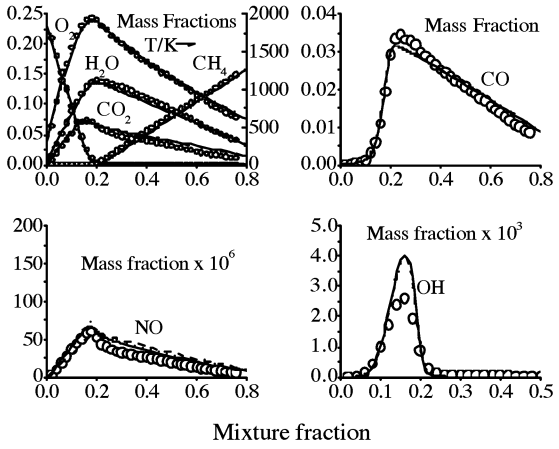


Figure 1: Measured and predicted conditional species mass fractions and temperature at x/d = 20 in flame A (o measured, — predicted Miller-Bowman, --- predicted 14-step Homma and Chen, ··· predicted 16-step Homma and Chen).

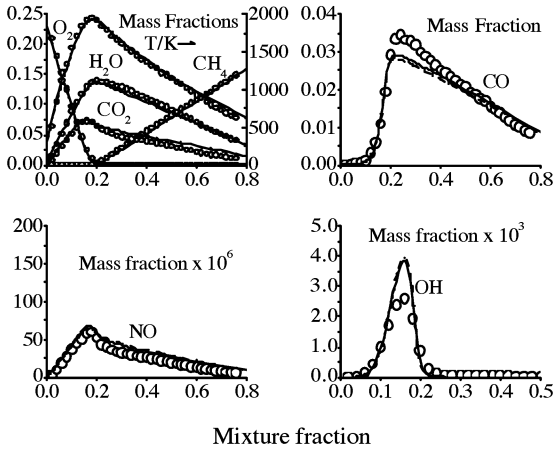


Figure 2: Measured and predicted conditional species mass fractions and temperature at x/d = 20 in flame A (o measured, — predicted GRI-Mech2.11, --- predicted 12-step Chen, ··· predicted 15-step Sung et al).

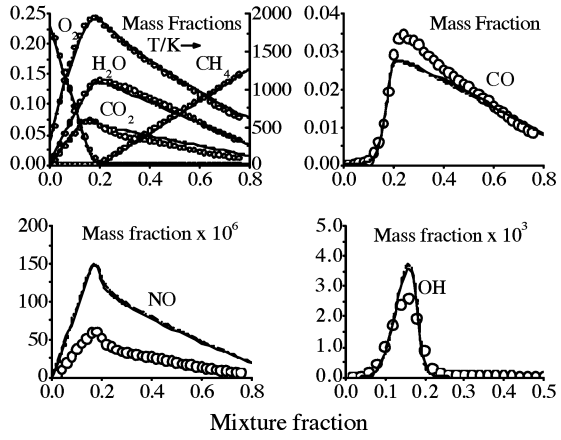


Figure 3: Measured and predicted conditional species mass fractions and temperature at x/d = 20 in flame A (o measured, — predicted GRI-Mech3.0, -·- predicted 13-step Sung et al, --- predicted 15-step Sung et al, ··· predicted 17-step Sung et al).

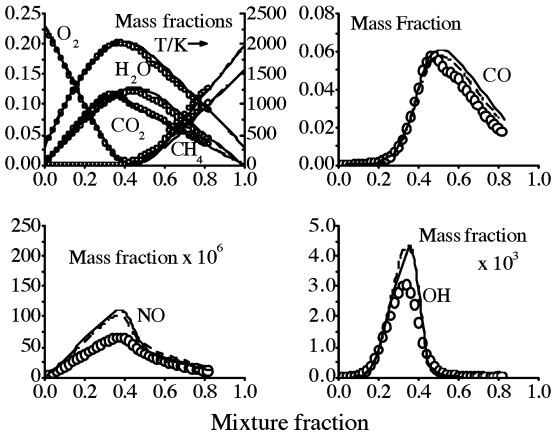


Figure 5: Measured and predicted conditional species mass fractions and temperature at x/d = 30 in flame D (o measured, — predicted GRI-Mech2.11, --- predicted 12-step Chen, ··· predicted 15-step Sung et al).

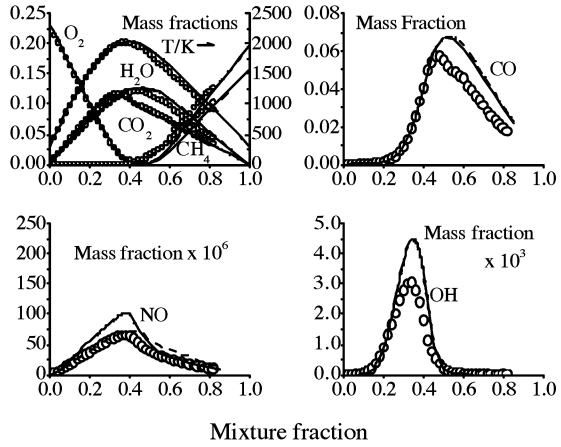


Figure 4: Measured and predicted conditional species mass fractions and temperature at x/d = 30 in flame D (o measured, — predicted Miller Bowman, --- predicted 14-step Homma and Chen, ··· predicted 16-step Homma and Chen).

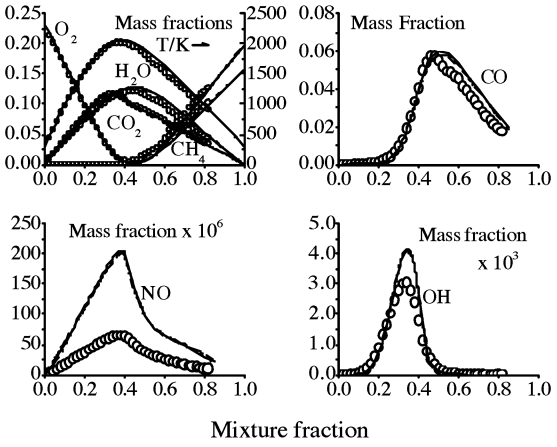


Figure 6: Measured and predicted conditional species mass fractions and temperature at x/d = 30 in flame D (o measured, — predicted GRI-Mech3.0, -·- predicted 13-step Sung et al, --- predicted 15-step Sung et al, ··· predicted 17-step Sung et al).