The Evaluation of Combustion Mechanisms using Local and Global Sensitivity and Uncertainty Methods.

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Abstract: Complex chemical mechanisms are increasingly used within models describing a range of important chemical processes. Within chemical models, kinetic parameters describing the rates of chemical steps and thermodynamics may be highly uncertain, influencing the uncertainty in final model predictions. Traditionally, local sensitivity analysis is employed within commercial modelling packages but may not be appropriate for highly uncertain data where models are nonlinear. This work compares linear sensitivity methods with global techniques such as Morris and Monte Carlo sampling for a kinetic model describing the influence of fuel sulphur on the oxidation of nitrogen within flames. The kinetics forms an important component of larger models describing pollution formation in combustion devices. The analysis reveals the most important rate and thermo-kinetic parameters contributing to the uncertainty in NO predictions for both rich and lean flames. The level of agreement between local and global techniques is highlighted. The use of reduced model representations using fitting methods is also discussed as a way of improving the efficiency of Monte Carlo based methods. Speed ups of a factor of 15 are seen without significant impact on the predicted mean output and standard deviation. For certain conditions, the mechanism is not capable of observing previous experimental data, highlighting the need for structural developments of the model such as including additional reaction steps for which data is not currently available.

Keywords: kinetic mechanism, sensitivity analysis, reduced model, Monte Carlo, MOAT.

1. INTRODUCTION

The use of computational modelling as a design tool is increasing within engineering applications. One area of importance is that of combustion reactor design. Environmental legislation means that engineers must develop combustion applications with low emissions of pollutants such as nitrogen and sulphur oxides. Understanding the impact of fuel trace elements such as nitrogen and sulphur containing compounds on pollutant emissions is important and requires the description of complex chemical mechanisms within the combustion chamber. In many cases mechanism data, such as rate constants and thermo-chemical parameters, are poorly categorised. If confidence is to be placed in the design process then the uncertainty in output predictions resulting from the use of such complex mechanisms should be investigated. Local/linear sensitivity analysis techniques are commomly used to evaluate such mechanisms. They have been developed in a generic way in the process engineering field using packages such as CHEMKIN [1], which is used for a range of applications including chemical mechanism validation in simplified flow environments such as flow reactors, premixed and diffusion flames. Linear methods are employed because they are computationally efficient, but are problematic where uncertainties in inputs are large and models are highly non-linear. This work therefore describes the development of methods for global uncertainty analysis for application within modelling packages such as CHEMKIN.

The paper will present a comparison of local and global uncertainty methods for a test case describing a 1 dimensional model of the influence of sulphur species on the emission of NO (nitrogen oxide) from methane air flames. Experiments show that the influence of sulphur within the flame can lead to both enhancement and reduction of NO emissions depending on whether the flame is fuel rich or fuel lean [2-4]. The chemical mechanism employed has been developed using evaluated rate data where possible. Predictions from the mechanism are evaluated for experimental data sets for a variety of flames and fuel nitrogen contents. Without sulphur present in the fuel the mechanism has previously been shown to exhibit good agreement with experimental profiles of key species. However, the mechanism does not capture the quantitative influence of sulphur on NOx emissions that is demonstrated in the 1D flame experiments[3]. Local sensitivity analysis has already revealed [3] several important reactions that require improved categorisation of the rate data to lower output variance. The present work presents a comparison between local and nonlocal linear methods, the global screening Morris One at a Time (MOAT) method and random sampling Monte Carlo methods using full and reduced models, coupled with scatter plot and correlation analysis.

2. MECHANISM STRUCTURE AND CONDITIONS FOR EVALUATION

The mechanism under investigation consists of an updated version of the Leeds methane/NOx mechanism [5-7]. The SOx extension (156 reversible reactions and 24 species) is based on the mechanism of Glarborg et al. [8] and Alzueta et al. [9] and has been augmented by the inclusion of additional reactions of sulphur containing species appropriate for a methane oxidation environment, and reactions describing sulphur-nitrogen interactions that have previously appeared in the literature or widely available databases. The uncertainty study here focuses on reactions of sulphur containing species and heats of formation of sulphur compounds. It is undertaken for selected experimental conditions from [3] where laser induced fluorescence studies were performed for a variety of low pressure methane flames doped with various levels of HCN and SO_2. Comparisons were made of relative NO concentrations for several dopant levels and flame stoichiometries from $\varphi = 0.7$ to $\varphi = 1.6$ (Table 2 of [3]). Whilst showing similar qualitative trends, previous mechanisms have tended to over predict the relative increase in NO on the addition of SO_2 for rich flames when compared to the experiment, and to under predict the reduction in NO for lean flames.

3. SENSITIVITY/UNCERTAINTY METHODS EMPLOYED

3.1 Uncertainties in Input Data

Well categorised kinetic rate parameters k, such as those from evaluations [10], are often quoted with an accuracy expressed as $\Delta \log k$. Here $\Delta \log k = D$ and D is defined by $\log_{10} k = C \pm D$. This is equivalent to the rate parameter k being uncertain by a factor f where $D = \log_{10} f$. For temperature dependant reactions this respresents the uncertainty at 298K ($f(298)$). A temperature dependant form for second order reactions is given by:

$$f(T) = f(298) \exp \left| \frac{\Delta E}{R} \left(\frac{1}{T} - \frac{1}{298} \right) \right|, \qquad (1)$$

where ΔE is the quoted error in the activation energy. The uncertainty of $\log_{10} k$ is usually assumed to be normally symmetric unless the parameter is stated as an upper or lower limit. One can then define a probability density function (pdf) for the rate parameter according to a distribution type. For less well categorised reactions a pdf cannot be determined and a minimum and maximum possible value are chosen, with an equal probability of the value of the

rate constant existing across the range. Local sensitivity coefficients are problematic in these cases since a most likely value cannot be reliably determined. However, if ranges of possible inputs are chosen, a full Monte Carlo analysis allows the user to determine if the model can "observe" target output values by comparison with experiments, allowing the evaluation of possible structural uncertainties in the model such as missing kinetic processes. Where target outputs are observable then the use of correlation factors or global ranking methods, allows the determination of the most important input factors leading to output uncertainties.

3.2 Linear Sensitivities.

In this study non-local linear sensitivities calculated using the *brute force method* are compared against previous local linear sensitivity studies from [3]. The use of the *brute force method* involves performing a base model run with output y_i, using the nominal values of the input parameters, and *n* extra model runs where each uncertain input parameter j is changed by a small factor Δj. The final output (y_i^*) from each run is determined and the sensitivity coefficient given by: $S_{i,j} = \dfrac{y_i - y_i^*}{\Delta j}$. The sensitivity at any temporal or spatial point is determined with a computational effort of order *n* (the number of uncertain inputs).

3.3 The Morris One at a Time (MOAT) Method.

Although they allow the study of non-linear interactions between parameters, global methods can be computationally expensive since thousands of model runs may be required. One example of a potentially more efficient screening method is the One at a Time analysis developed by Morris [11]. The method determines an importance ranking for parameters in terms of their mean effect on output variance as well as determining those parameters with linear additive effects and non-linear interactions [12]. In the MOAT method the inputs x_j are assumed to have values in the set {0, 1/(*p*-1), 2/(*p*-1),…,1} where in practice these values are re-scaled to values from within their uncertainty ranges. A perturbation factor Δ is defined as a multiple of 1/(p-1). A control simulation is then performed based on the random selection of parameters from the set {0,1/(*p*-1),…,1-Δ}. A single parameter is then randomly selected and modified by a factor Δ, and a second simulation performed. This is repeated until each factor has been chosen once, corresponding to *n+1* runs. This procedure is repeated *r* times until stable output statistics are obtained. The average output is computed over *r* runs and the cost of the method scales with *r(n+1)*.

The elementary effect of the j'th component of **x** on the output y_i where x_j has been changed by a factor Δ is given by:

$$d_{ij}(\mathbf{x}) = \frac{y_i(x_1,...,x_{j-1}, x_j \pm \Delta, x_{j+1,...,}x_m) - y_i(\mathbf{x})}{\Delta}. \tag{2}$$

The mean effect across *r* runs is given by: $\overline{d}_{ij} = \dfrac{\sum\limits_{l=1}^{r}\left|d_{ij}^l\right|}{r}$, and the variance:

$$\sigma^2(d_{ij}) = \frac{r\sum\limits_{l=1}^{r}(d_{ij}^l)^2 - \left(\sum\limits_{l=1}^{r}d_{ij}^l\right)^2}{r(r-1)}. \tag{3}$$

In this study, ten runs were sufficient to produce stable outputs and a value of *p*=4 was chosen. Parameters with the highest mean effect have a significant impact on overall output variance and require improved categorisation to lower output uncertainty. If the variance between runs is low then the effect is said to be linear or additive. Parameters with a high variance exhibit non-linear or interactive effects, which is important since it indicates parameters with sensitivities that may change if other parameter ranges are changed through improved categorisation.

3.4 Sampling Based Methods and Monte Carlo Analysis.

The standard method of Monte Carlo analysis is to perform a large number of model runs where in each run a sample of input parameters: $\mathbf{x_k} = [x_{k1}, x_{k2}, \ldots, x_{knx}]$, $k = 1, 2, \ldots, n_s$,

of size n_s is randomly selected from the possible values for $\mathbf{x}$ from the chosen distributions [12]. For a uniform distribution each sample region is assigned equal probability and therefore the sample points are chosen randomly from any region. The corresponding outputs $\mathbf{y}$ for each sample are determined by rerunning the model n_s times. Examination of the mapping from inputs to outputs can then be performed via a variety of methods including scatter plots, correlation analysis, regression analysis etc. The computational cost is of order n_s where n_s must be large enough for the mean output and the output variance to converge.

4. OVERALL METHODOLOGY

Simulation of the low pressure flames described in [3] has been performed using PREMIX [1] at a pressure of 40 Torr for fuel to air ratios Φ=1.6 and Φ=0.7. 0.3% and 0.5% of SO_2 has been added to the lean and rich flames respectively in line with the experiments [3]. The output y($\mathbf{x}$) is the NO mole fraction in the burnt gas region. Reactions have been treated as reversible with reverse rates calculated from the appropriate equilibrium constants. The sensitivity to the heats of formation therefore forms an important part of the study. Each PREMIX run is computationally fairly expensive since a larger number of coupled non-linear equations must be solved in order to determine the concentration profiles of over 75 species in the flame. The use of a fitted model within the Monte Carlo analysis will therefore also be discussed as a method of reducing the computational expense resulting from large numbers of PREMIX simulations.

The following methods will be presented for comparison:

For uncertainties in rate parameters of sulphur containing reactions:
1. linear sensitivities using the brute force method and a relative change of 10%,
2. MOAT analysis,
3. Monte Carlo analysis using up to 2000 model runs based on full and fitted models.

For uncertainties in heats of formation for the sulphur containing compounds:
4. Monte Carlo analysis using up to 2000 model runs.

Uncertainty ranges for kinetic rate parameters were assigned using *f* factors with 95% confidence limits where data evaluations existed. For parameters derived from a single experimental or modelling study a factor of 2 was used. Where data derived from a single RRKM calculation, or was estimated, a factor of 10 was assumed. Where the temperature dependence was estimated, an uncertainty factor in ΔE of 2 was assumed. Of the 155 parameters 18% derived from evaluated rate data, 18% from a low number of measurements, 7% from measurements with no evaluation, 8% from a single RRKM study and 49% were estimated. For this reason, only input ranges were determined and not pdfs. The analysis will not therefore allow pdfs of the outputs to be determined, but rather allows the evaluation of the

current mechanism in order to inform its future development. Thermodynamic data and their uncertainties were obtained from the databases of Burcat [13] and NIST [14]. Coefficients for a large number of the sulphur compounds originated from modelling techniques [15]. Where a single model value was used an uncertainty of ± 100 (kJmol^{-1}) was assumed. Because almost half the thermodynamic data was of this type uniform distributions were used.

5. RESULTS AND DISCUSSION

5.1 Linear Sensitivity Coefficients.

$\Phi=1.6$: The importance ranking from the non-local linear sensitivity study for the rich flame is presented in Table 1. The highest ranked reaction is SO+NH=NO+SH in agreement with the local sensitivity study in [3]. The second highest ranked reaction from the local study was SO_2+H=SO+OH, which is represented here by its reverse rate [16] and ranked 5th. The second highest ranked reaction from this study is SH+NH=NS+H_2, which was also highlighted by the local sensitivity study and has an estimated rate giving scope for improvement. The reaction H_2S+M=H+SH+M, ranked third here, was not identified using local sensitivity coefficients. SO+O_2=SO_2+O was highlighted by the previous study and again has a negative sensitivity when using non-local methods. There is therefore some broad agreement between the local and non-local linear sensitivity methods as well as notable differences in ranking.

Table 1 – Comparison of importance ranking of sulphur containing reactions in the rich flame (Φ = 1.6) from the linear brute force (lbf) and MOAT analyses.

React. No.	Reaction	lbf Rank	MOAT Rank	Pearson Coeff.	Source of Data
1	SO+NH=NO+SH	1	1	0.79	Single meas.
2	SH+NH =NS+H_2	2	5	0.12	Estimated
3	H_2S+M=H+SH+M	3	7	-0.08	Unevaluated measurements.
4	SO+O_2=SO_2+O	4	14	-0.03	Evaluated
5	SO+OH=SO_2+H	5	4	0.2	Single meas.
6	S+OH=SH+O	6	15	-0.07	Estimated
7	HSO+H=SH+OH	7	33	-0.03	Estimated
8	S+H_2=H+SH	8	10	-0.06	Unevaluated measurements.
9	SO+N=NO+S	9	2	0.56	Estimated
10	H_2S+M=H_2+S+M	10	55	0.01	Unevaluated measurements.
11	HSOH=SH+OH	69	3	0.04	Estimated
12	SH+H=H2+S	11	6	0.06	Unevaluated measurements.
13	SH+NO=SN+OH	13	8	0.01	Estimated
14	SN+O=SO+N	39	9	0.05	Estimated

$\Phi=0.7$: In the lean case a 10% increase in the selected rate parameters was not sufficient to cause any detectable change in the NO mole fraction. An factor of 10 increase was required to produce a detectable difference, making it impossible to determine an importance ranking using a linear method. The analysis indicates that for lean conditions, the NO concentration is highly insensitive to the forward rate parameters of the sulphur reactions around their nominal values.

5.2 MOAT Analysis

$\Phi=1.6$: Figure 1a shows the variance of the factor effects plotted against the mean effects from the MOAT analysis for the rich flame. Parameters with a low significance in terms of output

variance appear at the bottom left of the plot. Those in the bottom right segment have a high linear effect on model output and those in the upper portion show strong non-linear or interactive effects. The reaction SO+NH=NO+SH appears at the bottom right of the figure showing a strong linear effect on the output in agreement with the linear methods. SO+N=NO+S also shows a strong linear effect and ranks second in the MOAT analysis as shown in Table 1, although lower in the linear analyses. HSOH = OH+SH shows a strong mean effect and a high variance, indicating that the sensitivity to this reaction strongly depends on the values of the other parameters. Not surprisingly this reaction was not identified as important by the linear methods. Its high ranking by the MOAT analysis is strong evidence of the importance of using global uncertainty techniques.
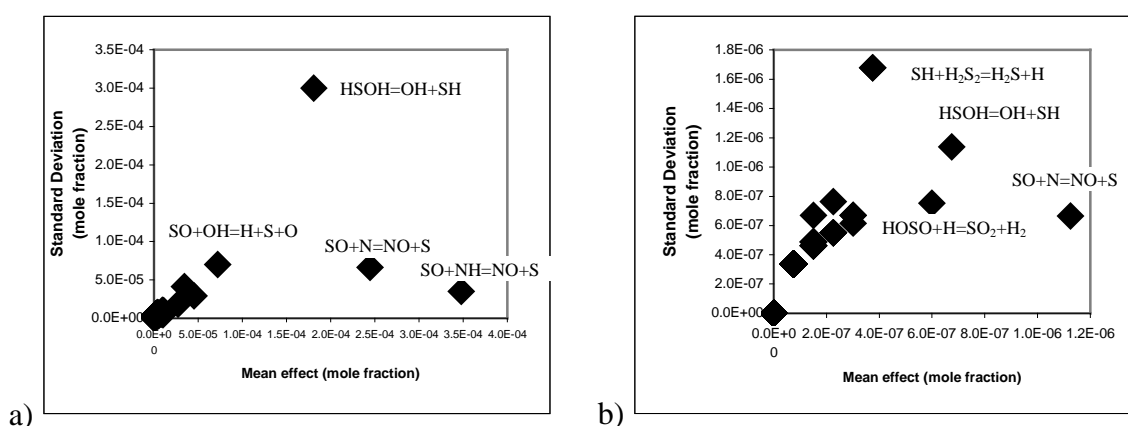

Figure 1. Morris One at a Time Analysis for (a) ($\Phi = 1.6$) and (b) ($\Phi = 0.7$)

Table 2 – Importance ranking of sulphur reactions in lean flame ($\Phi = 0.7$) from MOAT analysis.

React. No.	Reaction	MOAT Rank	Source of Data
9	SO+N=NO+S	1	Estimated
11	HSOH=SH+OH	2	Estimated
15	HOSO+H=SO$_2$+H$_2$	3	Estimated
16	SH+H$_2$S$_2$=H$_2$S+HS$_2$	4	Single low temp. meas.
17	S$_2$+H+M=HS$_2$+M	5	Estimated
1	SO+NH=NO+SH	6	Single meas.
18	SO2+OH=SO3+H	7	Estimated
19	SO+M=S+O+M	8	Unevaluated measurements.

$\Phi=0.7$: Figure 1b and Table 2 represent the output from the MOAT analysis for the lean flame. The overall mean effect of the rate parameters on the NO mole fraction in the burnt gas region is much lower than for the rich flame. SO+N=NO+S shows the highest overall mean effect and since this has an estimated rate there is some scope for improvement in predictions by its better categorisation. There are some differences between the reactions ranked highly by the MOAT analysis and by the linear studies in this and previous work [3]. For example reactions 11, 16, and 17 had no impact in the linear study despite being modified by up to a factor of 10. This indicates interaction effects between rate parameters in the scheme. The dominant uncertainty in the conversion from SO$_2$ to SO$_3$ identified by the MOAT analysis is via OH rather than the pressure dependant reaction in the linear study. The low mean effect of all reactions for this flame again highlights the low sensitivity of NO to the sulphur chemistry for lean conditions.

5.3 Monte Carlo analysis.

The output values resulting from the randomly sampled input distributions for the rich flame are presented in Fig. 2 as scatter plots for high ranking reactions from the MOAT analysis. Pearson correlation functions [12] can also be used to determine the strength of the linear response of the output to the input parameters (see Table 1). A high correlation implies a strong linear response of NO concentrations to an increase in the rate parameter. These correlation coefficients do not take into account interactive effects.
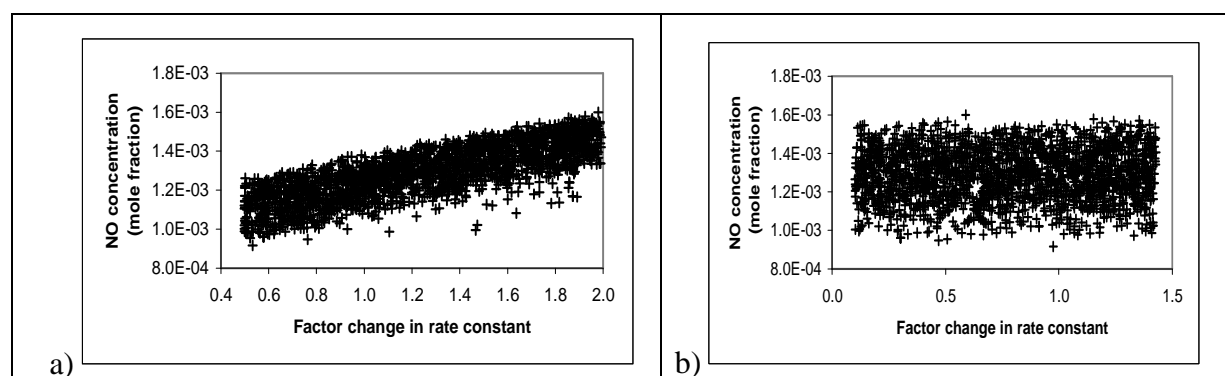

Figure 2. Scatter plot showing Monte Carlo simulation of changes in final NO concentration resulting from changes in reaction rate constant (a) SO+NH=NO+SH, (b) HSOH = OH+SH.

The Pearson coefficient of SO+NH=NO+SH is $r = 0.79$ (Fig. 2a) showing a strong linear response in line with both the linear sensitivity and MOAT methods. The scatter about the mean effect in Fig. 2a is due to the influence of other parameters on the output. This reaction is ranked highest by all the methods of analysis and is clearly important for the rich flame. SO+N=NO+S also shows strong linear behaviour as demonstrated by $r = 0.56$, in agreement with the MOAT analysis. As expected, the Monte Carlo analysis coupled with linear regression techniques agrees well with the MOAT analysis for reactions with strong linear effects. Interestingly this reaction is fairly low down the importance ranking using both linear methods. Because it is estimated however, this parameter has a highly uncertain input range and therefore its overall contribution to the output uncertainty is high, even though its sensitivity may not be. HSOH=SH+OH is ranked third by the MOAT analysis. It has an extremely low ranking using the linear method and as Fig. 2b shows there is a large amount of scatter in the Monte Carlo results. Its correlation coefficient is very low ($r = 0.04$). This stems from the fact that the sensitivity of NO mole fraction to this reaction rate parameter changes sign in different regions of the input parameter space. In order to highlight the importance of such non-linear responses either the MOAT method or higher order correlation techniques combined with Monte Carlo based methods must be used. The ranking of reactions SO+OH=SO$_2$+H and SH+NH =NS+H$_2$ as derived from the correlation coefficients and the MOAT analysis are similar due to their high mean effects on the output but low variances. Scatter plots and Pearson coefficients are not presented for the lean flame since insufficient changes in NO mole fraction occur. Each method therefore highlights the lack of sensitivity to the sulphur chemistry in the lean flame leaving limited scope for model improvement via better characterisation of the current rates.

5.4 Agreement with experiment.

The experimental study in [3] showed a 16% increase in the NO mole fraction in the burnt gas region of the rich flame on the addition of 0.5% SO$_2$, with an experimental uncertainty of

±12%. In order to overlap with the experimental predictions the NO mole fraction in the burnt gas region must be < 9.545 x 10^{-4}. The scatter plots show that in order to achieve this, several of the parameters must be chosen from the extremes of their input ranges. A view could be taken that the likelihood of these parameters being at their extreme values is low. This leads to the possibility of structural problems within the model, such as missing reaction sets or rate parameters that are significantly more uncertain than has been assumed. For the lean flame, the model does not observe the concentration determined in the experiments with any combination of the forward rate parameters for the sulphur reactions used here.

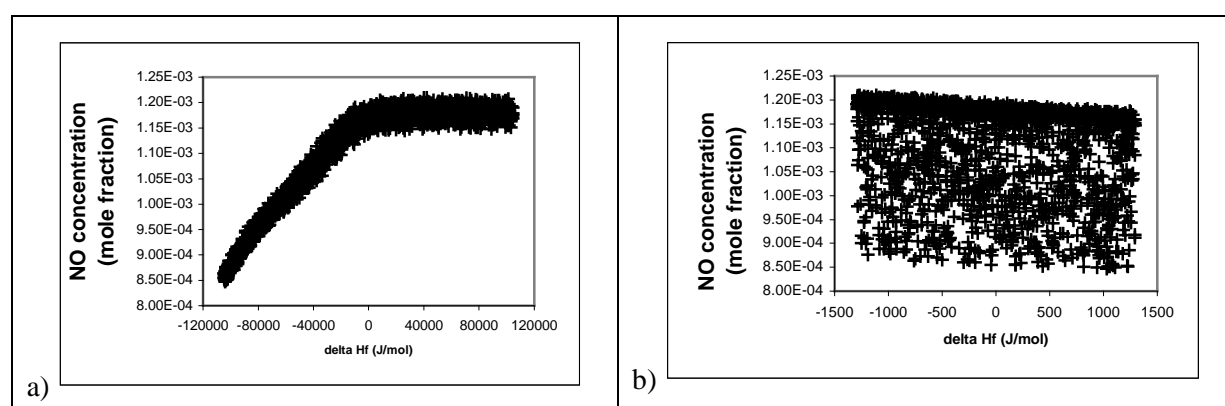## 5.5 Influence of Heats of Formation.

Figure 3. Scatter plot showing Monte Carlo simulation of changes in final NO concentration resulting from changes to heats of formation of a) NS, b) SO.

Uncertainties in the heats of formation of sulphur containing species may affect the reaction kinetics as the rates of the reverse reactions are determined through the equilibrium constant. Figure 3 shows scatter plots from a Monte Carlo analysis of uncertainties in heats of formation for the two most significant parameters for the rich flame. The influence of the heat of formation for NS ($\Delta_fH_{298}(NS)$) is extremely non-linear with a strong response at values lower than the quoted value, which flattens off at higher values. The output NO mole fraction will therefore be dominated by ($\Delta_fH_{298}(NS)$) only if the quoted value is too high. At higher values other compounds start to have an effect as shown by the scatter plots for SO. The large amount of scatter stems from the dominant influence of NS in its lower range. However, as the effect of NS saturates, a negative linear response to Δ_fH_{298} for SO can be seen. The influence of ($\Delta_fH_{298}(SO)$) is therefore highly dependant on the value chosen for ($\Delta_fH_{298}(NS)$).

5.6 Computational Requirements and Stabilisation of Output Statistics.

Theoretical estimations of the number of Monte Carlo runs required for the analysis of 155 uncertain parameters would be extremely high. In reality, 2000 runs are sufficient for output statistics to settle. Because of the large number of uncertain input parameters attempts could be made to reduce the computational costs by focussing the Monte Carlo analysis on a smaller number of parameters, for example those highest ranked from the MOAT analysis. For comparison purposes therefore, a second Monte Carlo run has been performed for the rich flame, where only the top 15 reactions from the MOAT analysis vary randomly within their input uncertainty ranges. All other parameters are fixed at their nominal values. The final means for the full and 15 parameter runs compare well at 1.290x10^{-3} and 1.283x10^{-3} respectively, as do the final standard deviations of 1.321x10^{-4} and 1.320x10^{-4}. This shows that the top 15

parameters identified by the MOAT analysis account for almost all the variance in the final output. However, the 15 parameter run does not settle statistically any faster than the full run, meaning that large computational savings have not been made, since a similar order of full model simulations have been required. This indicates that the number of samples used in the Monte Carlo analysis depends not on the total number of uncertain parameters, but on the number of important parameters that significantly affect the output statistics. In many cases, the number of samples required may not rise dramatically with the number of uncertain input parameters, since only a few parameters may dominate the output uncertainty.
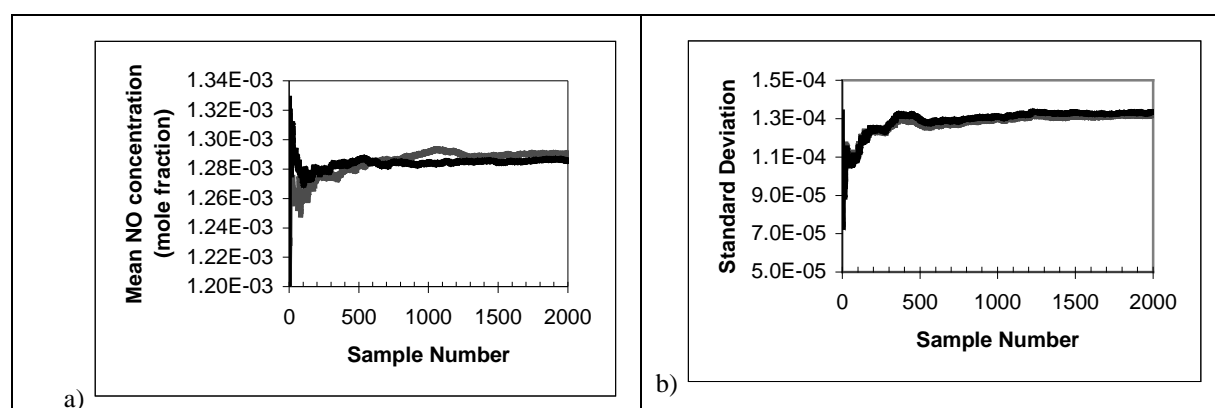

Figure 4 Comparison of output from Monte Carlo simulations using full and reduced models. Grey line - full PREMIX runs, black line – runs using fitted polynomial.

Further attempts may be made in order to reduce the computational cost of the Monte Carlo runs. In this work a second order polynomial equation has been fitted to represent the input output relationships from a small number of randomly sampled PREMIX runs using a Gram-Schmidt orthonormalisation procedure [17]. Again 15 input parameters are utilised as determined by the MOAT analysis and 120 samples are used for the fit. The polynomial coefficients are determined by minimizing the root mean square error (rms) of the fitted function with respect to the target output data derived from the full model. Terms not reducing the rms error are discarded. The final polynomial is factorised using Horner equations to minimize the number of arithmetic expressions required in the final simulation. Figure 4 shows that both the mean output and the output variance behave in a very similar way with increasing sample size for the full PREMIX simulations and those using the polynomial model. After 2000 simulations both give extremely similar results, despite the reduced model being formulated using only 120 full simulations. The final output means from the full and reduced models are 1.283×10^{-3} and 1.286×10^{-3} respectively, and the final standard deviations 1.320×10^{-4} and 1.337×10^{-4}, showing that the polynomial model gives similar results for >15 times lower computational costs. In contrast, if only 120 full model runs had been used then the final mean and standard deviations would not have settled down and would have been 1.272×10^{-3} and 1.217×10^{-4} and therefore do not represent the final values as well as using 2000 polynomial model runs. A further point is that the polynomial model directly reveals second order interactions between parameters.

6. FINAL DISCUSSION AND CONCLUSIONS

The analysis shows that useful information can be obtained from linear sensitivities, although both the linear sensitivities and the regression analysis fail to identify important reactions with strong non-linear interactions. Using combined global methodologies however, highlights a

range of reaction rates and thermo-kinetic parameters leading to output uncertainties in NO predictions providing useful information for further kinetic studies. Each method agrees that the sensitivity of NO mole fraction to sulphur containing reactions is much higher in the rich flame than the lean flame, which is extremely insensitive to the parameters tested here. Possible mechanistic problems relevant to the lean flame must therefore be identified. Since the mechanism utilised in the study contained all reactions present in the literature with measured, modelled or estimated rates, significant further improvements may involve new elementary reactions being postulated with a further requirement for the categorisation of their rate data.

The MOAT analysis performed in this study successfully identified those reactions making the major contribution to the overall output uncertainty as determined by Monte Carlo techniques. Once this group of 15 parameters had been identified it was demonstrated that the compuional expense of using Monte Carlo analysis could be significantly reduced by fitting a polynomial model describing the relationship between the 15 important parameters and the chosen model output. 120 random sampling runs proved sufficient for the fit, that was then capable of predicting the mean output and standard deviation across many runs with a high degree of accuracy when compared to analysis using full model runs. Since the computation of polynomials is so fast, the use of the reduced model gave speed ups of greater than a factor of 15. This approach of combining a global screening method with random sampling analysis using a fitted model could therefore have potential benefits for the future application of global uncertainty methods where individual model runs are computationally time consuming.

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