A global sensitivity study of sulphur chemistry in a premixed methane flame model using HDMR

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Outline

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Need for new methods in sensitivity analysis

- Complex chemical mechanisms are increasingly used in models describing a range of important chemical processes (e.g. combustion)
- Models contain a large number of parameters and are often highly non-linear
- Large uncertainty ranges for the parameters
- Models are computationally expensive to run
- Traditional methods for global uncertainty and sensitivity analysis not suitable due to their computational expense and the difficulty in interpreting the results
- Aim: method that can cope with large parameter numbers
- Do we need a screening method, which identifies unimportant parameters beforehand?

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High Dimensional Model Representation (HDMR)

HDMR basics

- Output $f(\mathbf{x})$ of a model can be expressed as finite hierarchical function expansion in terms of the input parameters $\mathbf{x}$

$$f(\mathbf{x}) = f_0 + \sum_{i=1}^{n} f_i(x_i) + \sum_{1 \leq i < j \leq n} f_{ij}(x_i, x_j) + \ldots + f_{12\ldots n}(x_1, x_2, \ldots, x_n)$$

- Usually HDMR expansion to second order provides satisfactory results and a good description of $f(\mathbf{x})$
- Provides detailed input-output mapping suitable to create a model replacement and for global SA
- Several decomposition methods: e.g. cut-HDMR, RS-HDMR

Random Samping (RS)-HDMR

Principles

- Only one set of random samples necessary to estimate all component functions
- Component functions can be approximated by analytical basis functions such as polynomials or splines

$$f_i(x_i) \approx \sum_{r=1}^{k} \alpha_r^i \varphi_r(x_i)$$

$$f_{ij}(x_i, x_j) \approx \sum_{p=1}^{l} \sum_{q=1}^{l'} \beta_{pq}^{ij} \varphi_p(x_i) \varphi_q(x_j)$$

- Here:$\varphi_r(x_i)$, $\varphi_p(x_i)$ and $\varphi_q(x_j)$ orthonormal polynomials

Random Samping (RS)-HDMR extension

Optimisation of the polynomial expansion order (1)

- Usually the same polynomial order is used for all first-order and second-order component functions respectively
- Order of the polynomial approximation should be chosen separately for each component function
- Optimisation algorithm is based on least square method
- Sum of square errors is calculated using the results of the full model runs and the approximation of the component functions by various orders (e.g. 0th to 5th order)
- Smallest sum of square errors indicates the best approximation order for the corresponding component function

Random Samping (RS)-HDMR extension

Optimisation of the polynomial expansion order (2)

- Exclude component functions which do not contribute (identified as 0th order by optimisation)
- Threshold to exclude unimportant component functions (not identified as 0th order, but only very small contribution to overall value)
- **Idea: avoid the need for screening methods**
- Low computational effort to calculate optimal order for all polynomials
- Improvement in the accuracy of the final model replacement

Global sensitivity analysis

Sensitivity Indices

- Sensitivity Indices measure the effect of one or more input parameters on the output
- S_i measures the effect of x_i (fractional contribution)
- S_{ij} measures the interactive effect of x_i and x_j

Computation

- Easily calculated from the HDMR expansion (no additional full model runs required), e.g.

$$D_i \approx \sum_{r=1}^{k_i} (\alpha_r^i)^2 \qquad S_i = \frac{D_i}{D}$$

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Application

Modelling in combustion

- Aim: development of combustion applications with low emissions of pollutants such as nitrogen and sulphur oxides
- Design of low emission technologies depends on accurate computational models describing combustion processes
- Trace amounts of sulphur in fuel can have an impact on the extent of nitrogen oxide emissions
- Models required which describe the interaction of sulphur containing compounds with other species within flames

Application

Methane flame model

- Premixed methane flame model describing the influence of sulphur containing compounds on the formation of nitrogen oxides
- Assessment of the resulting uncertainty in predictions of nitrogen oxide emissions is important to improve the confidence in the design process
- Modelling using CHEMKIN and simulation using PREMIX
- Model contains large number of parameters (with large uncertainty ranges)
- 177 uncertain parameters: 153 reactions rates and 24 heats of formation (calculated by NASA polynomials)

Methane flame model

Investigated scenarios

- Output of interest: NO concentration
- Uncertainty ranges for all parameters according to min and max value with equal probability
- Ranges for the 153 reaction rates according to Tomlin (2006)
- 3 different sets of ranges for the 24 heats of formation:

1. **ΔHf = $\pm$10KJ for all** 24 parameters (assume equal uncertainties)
2. Ranges according to Burcat table if available (**ΔHf SN = $\pm$105KJ**) (`http://technicon.ac.il/~aer0201`)
3. Ranges according to Burcat table, but **ΔHf SN = +13KJ** (based on updated value for heats of formations for SN according to Peebles and Marshal (2002) with smaller uncertainty range)

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Methane flame model

HDMR set up

- Second order RS-HDMR expansion
- Approximation of the component function by orthonormal polynomials
- Quasi-random sampling (N=1024)
- Optimisation of the polynomial order:
  - Maximum order for approximation of first-order component functions: 10
  - Maximum order for approximation of second-order component functions: 3
- Excluding component functions via threshold
- Correlation method for variance reduction (Li et. al 2002)
- HDMR method including all 177 parameters

Results - Scenario 3 (Burcat table, ΔHf SN = +13KJ)

- Sample size: N=1024
- Threshold to exclude component functions: 1 %

Optimal polynomial order

- **5** of 177 first-order component functions are non-zero
  - **2** have been approximated by **1st-order** polynomials
  - **2** have been approximated by **2nd-order** polynomials
  - **1** has been approximated by **8th-order** polynomial
- **0** of 15576 second-order component functions are non-zero

Results - Scenario 3 (Burcat table, ΔHf SN = +13KJ)

- Sample size: N=1024
- Threshold to exclude component functions: 0.001 %

Optimal polynomial order

- **51** of 177 first-order component functions are non-zero
  - **25** have been approximated by **1st-order** polynomials
  - **17** have been approximated by **2nd-order** polynomials
  - **6** have been approximated by **3rd-order** polynomials
  - **1** has been approximated by **4th-order** polynomial
  - **1** has been approximated by **5th-order** polynomial
  - **1** has been approximated by **9th-order** polynomial
- **4** of 15576 second-order component functions are non-zero
  - **4** have been approximated by **1st-order** polynomials

Results - Scenario 3 (Burcat table, ΔHf SN = +13KJ)

Accuracy - variance

- No additional full model runs required
- Variance full model runs (N=1024): $D = 1.7367 \cdot 10^{-8}$
- Variance 1st-order model replacement:
  $\hat{D}_{1st} = 1.7002 \cdot 10^{-8} \rightarrow 97.89\,\%$
- Variance 2nd-order model replacement:
  $\hat{D}_{2nd} = 1.7043 \cdot 10^{-8} \rightarrow 98.13\,\%$

Accuracy - Relative Error (RE)

- Additional set of full model runs required (N=2000)
- 1st-order model replacement 5 % RE: 99.50 %
- 2nd-order model replacement 5 % RE: 99.65 %

Results - Scenario 3 (Burcat table, ΔHf SN = +13KJ)

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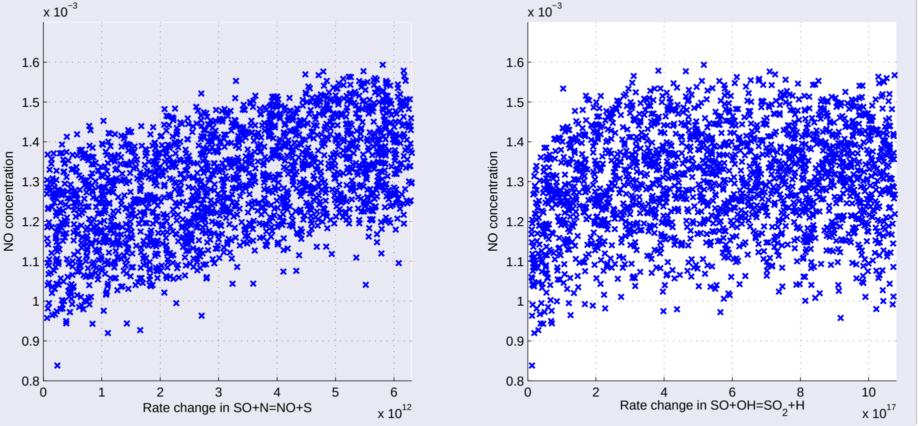
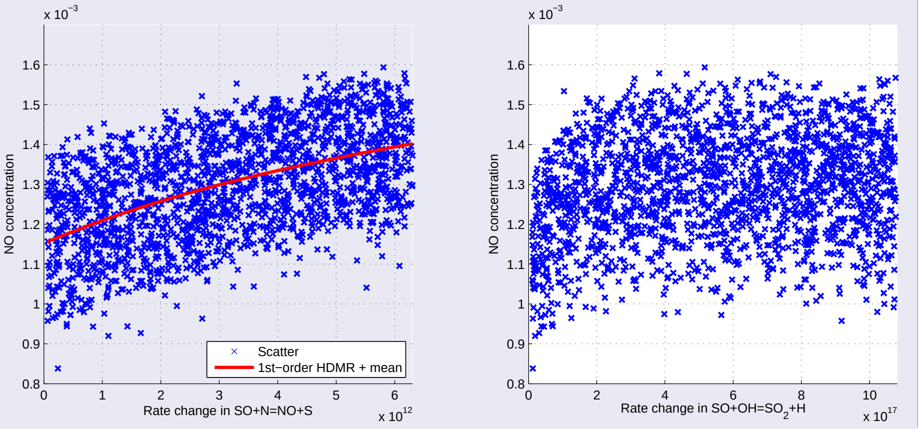
Scatter plots



Results - Scenario 3 (Burcat table, ΔHf SN = +13KJ)

Scatter plots + first-order RS-HDMR component function



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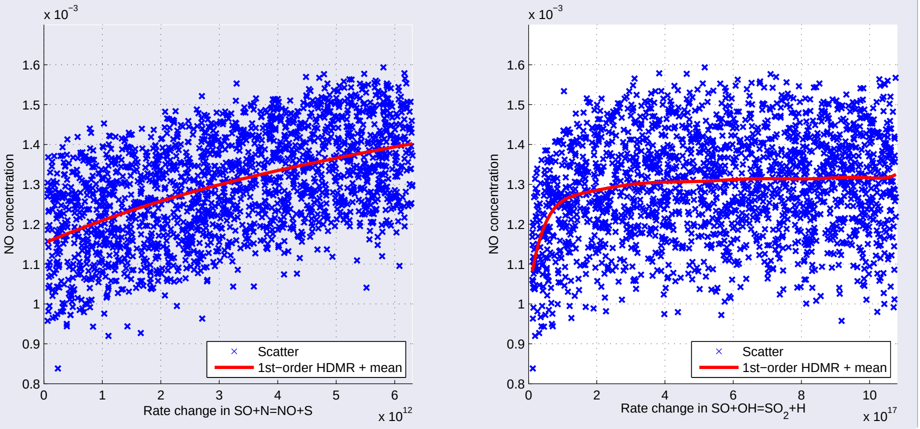

Results - Scenario 3 (Burcat table, ΔHf SN = +13KJ)

First and second-order sensitivity indices (N=1024)

Parameter	Rank	S_i
$SO + NH = NO + SH$	1	0.5956
$SO + N = NO + S$	2	0.2758
$SO + OH = SO_2 + H$	3	0.0735
ΔHf SO	4	0.0140
$SH + NH = NS + H_2$	5	0.0111
$\sum S_i$		0.9784
Parameter 1	**Parameter 2**	S_{ij}
$SO + OH = SO_2 + H$	$SO + NH = NO + SH$	0.0018
$SH + H = H2 + S$	$SH + NH = NS + H_2$	0.0015
$S + CS_2 = CS + S_2$	$HS_2 + H + M = H_2S_2 + M$	0.0005
$H_2S + M = H_2 + S + M$	$S + NO_2 = NO + SO$	0.0004
$\sum S_{ij}$		0.0042
S		0.9826

Results - comparison of all scenarios

First-order sensitivity indices (N=1024)

Parameter	Scenario 1 (ΔHf = $\pm$10KJ) S_i (Rank)		Scenario 2 (ΔHf SN = $\pm$105KJ) S_i (Rank)		Scenario 3 (ΔHf SN = +13KJ) S_i (Rank)	
$SO + NH = NO + SH$	0.2297	(3)	0.3631	(1)	0.5956	(1)
$SO + N = NO + S$	0.1007	(4)	0.1219	(3)	0.2758	(2)
$SO + OH = SO_2 + H$	0.0255	(5)	0.0316	(4)	0.0735	(3)
$\Delta Hf\ SO$	0.3082	(1)	0.0101	(5)	0.0140	(4)
$SH + NH = NS + H_2$	0.0035		0.0019		0.0111	(5)
$\Delta Hf\ SO_2$	0.2874	(2)	0		0	
$\Delta Hf\ SN$	0.0001		0.3479	(2)	0.0001	
$\sum S_i$	0.9771		0.8904		0.9784	
S	0.9842		0.9055		0.9826	

Results - Scenario 2 (Burcat table, ΔHf SN = $\pm$105KJ)

Improvement of the accuracy

1. RS-HDMR method using 177 inputs (N=4096)
2. Morris Method to identify unimportant parameters first (N=1780) + RS-HDMR method using 47 inputs (N=1024)

	RS-HDMR (N=1024)	**RS-HDMR** (N=4096)	**Screening + RS-HDMR** (N=1780+1024)
1st-order 5 % RE	85.85 %	**90.40 %**	**89.90 %**
2nd-order 5 % RE	86.25 %	**94.80 %**	**95.30 %**

Results - Scenario 2 (Burcat table, ΔHf SN = ±105KJ)

First-order sensitivity indices

1. RS-HDMR method using 177 inputs (N=4096)
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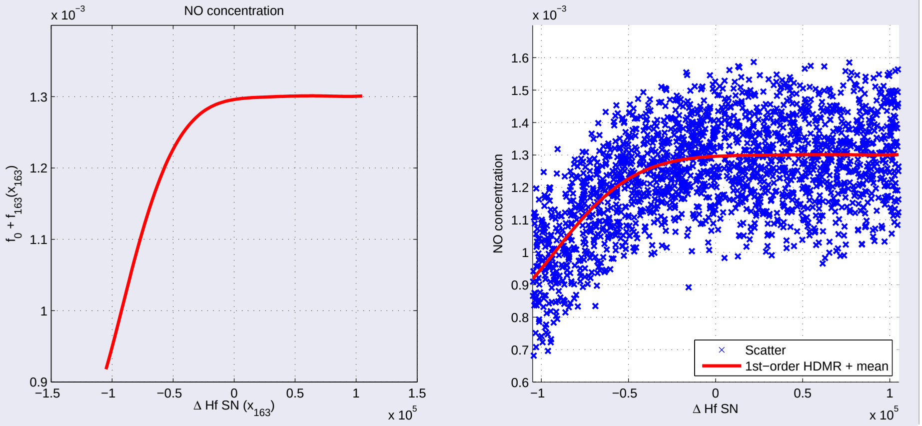
Parameter	RS-HDMR S_i	RS-HDMR (Rank)	Screening + RS-HDMR S_i	Screening + RS-HDMR (Rank)
ΔHf SN	0.3815	(1)	0.3855	(1)
$SO + NH = NO + SH$	0.3700	(2)	0.3719	(2)
$SO + N = NO + S$	0.1344	(3)	0.1365	(3)
$SO + OH = SO_2 + H$	0.0410	(4)	0.0386	(4)
ΔHf SO	0.0086	(5)	0.0087	(5)
$\sum S_i$	0.9524		0.9590	
S	0.9695		0.9755	

Results - Scenario 2 (Burcat table, ΔHf SN = ±105KJ)

First-order RS-HDMR component functions and scatter plot



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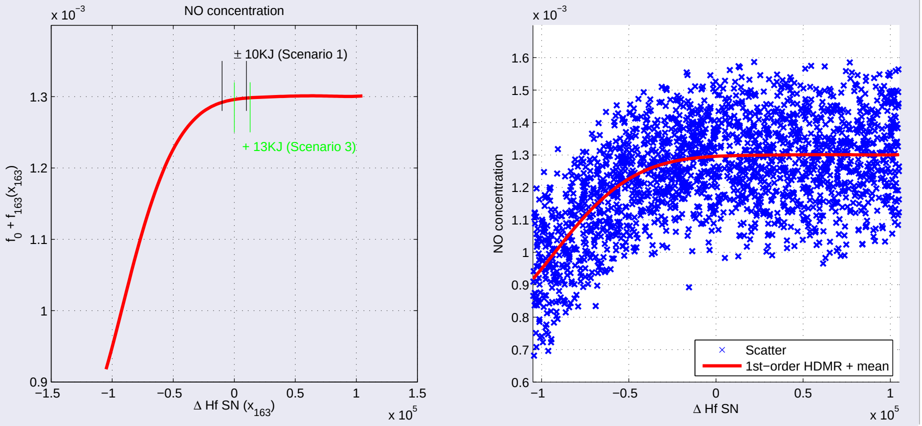
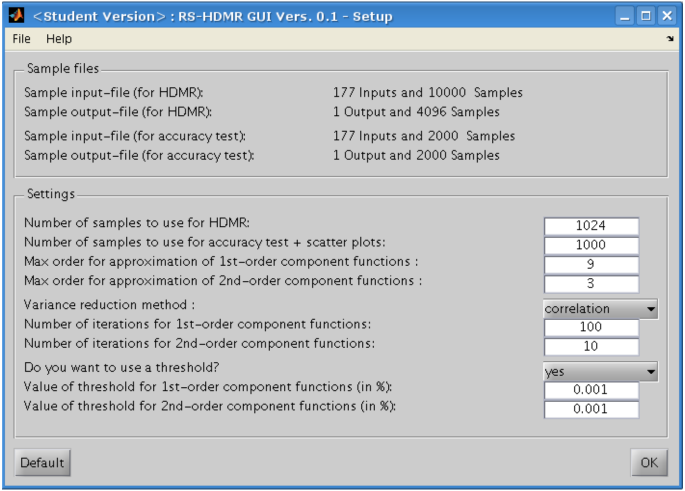


Graphical User Interface for RS-HDMR



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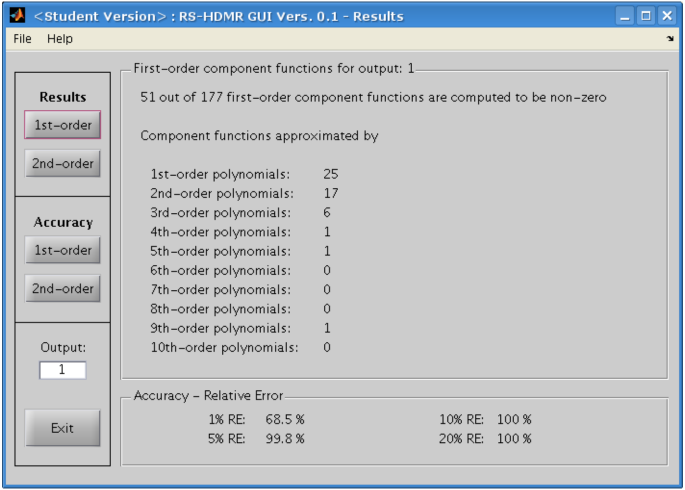

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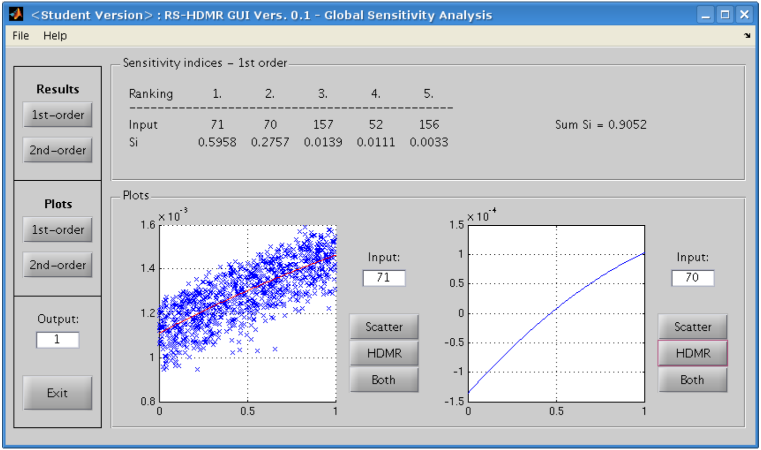

Conclusions

- RS-HDMR method provides straightforeward approach for global sensitivity analysis
- However, extension to existing HDMR tools necessary to explore large number of input parameters
- Optimisation method in combination with excluding component functions via a threshold is one useful extension
- Variance reduction method (correlation method, Li et. al 2003) useful to further improve accuracy
- Generally no screening method necessary in order to reduce the number of parameters, but using one can in certain cases reduce the computational effort
- Final ranking of the important parameters is critically dependent on the input ranges chosen