

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

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Outline

- 1 Introduction
- 2 HDMR - High Dimensional Model Representation
- 3 Application and Results
- 4 HDMR Software

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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}

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

- 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 Sampling (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 Sampling (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 Sampling (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.

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

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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 $\Delta H_f = \pm 10 \text{KJ}$ for all 24 parameters (assume equal uncertainties)
 - 2 Ranges according to Burcat table if available ($\Delta H_f \text{ SN} = \pm 105 \text{KJ}$) (<http://technicon.ac.il/~aer0201>)
 - 3 Ranges according to Burcat table, but $\Delta H_f \text{ SN} = +13 \text{KJ}$ (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, $\Delta H_f^{\text{SN}} = +13\text{KJ}$)

- 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, $\Delta H_f \text{ SN} = +13\text{KJ}$)

- 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, $\Delta H_f \text{ SN} = +13\text{KJ}$)

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, $\Delta H_f \text{ SN} = +13\text{KJ}$)

Accuracy - variance

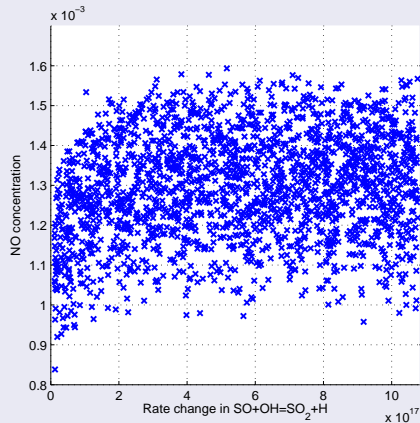
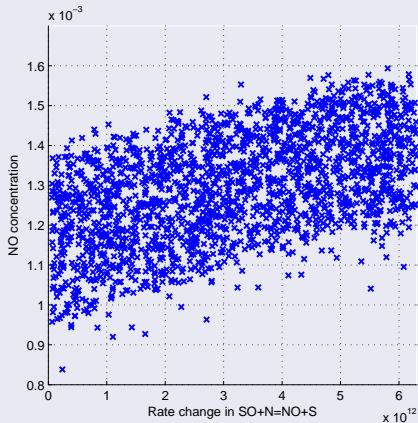
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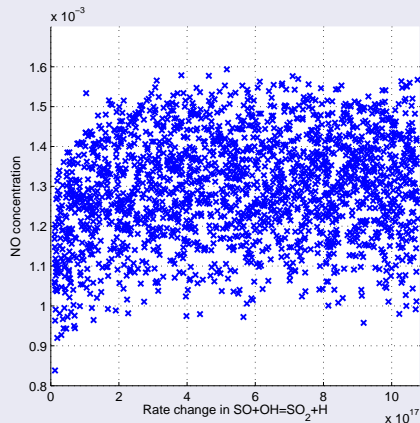
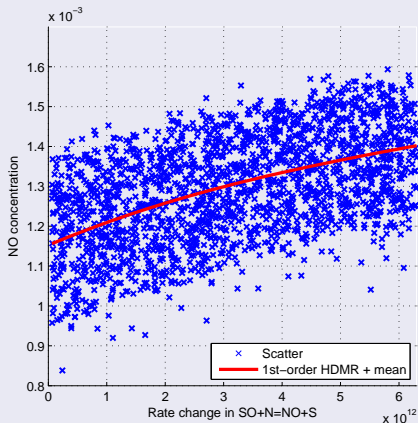
Results - Scenario 3 (Burcat table, ΔH_f SN = +13KJ)

Scatter plots



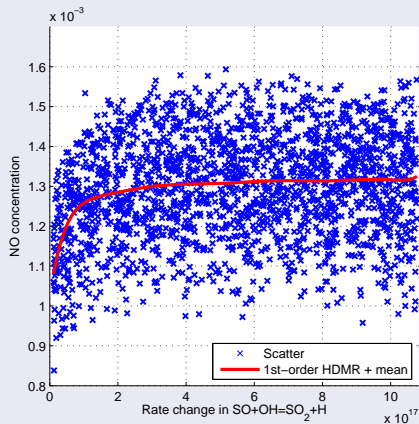
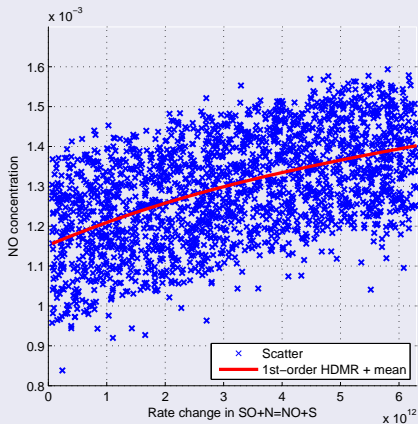
Results - Scenario 3 (Burcat table, ΔH_f SN = +13KJ)

Scatter plots + first-order RS-HDMR component function



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Scatter plots + first-order RS-HDMR component functions



Results - Scenario 3 (Burcat table, $\Delta H_f \text{ SN} = +13\text{KJ}$)

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 |
| $\Delta H_f \text{ SO}$ | 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 = H ₂ + S | SH + NH = NS + H ₂ | 0.0015 |
| S + CS ₂ = CS + S ₂ | HS ₂ + H + M = H ₂ S ₂ + M | 0.0005 |
| H ₂ S + 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 | Scenario 2 | Scenario 3 |
|-------------------------------|--|--|--|
| | ($\Delta H_f = \pm 10\text{KJ}$) S_i (Rank) | ($\Delta H_f \text{ SN} = \pm 105\text{KJ}$) S_i (Rank) | ($\Delta H_f \text{ SN} = +13\text{KJ}$) 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) |
| ΔH_f SO | 0.3082 (1) | 0.0101 (5) | 0.0140 (4) |
| SH + NH = NS + H ₂ | 0.0035 | 0.0019 | 0.0111 (5) |
| ΔH_f SO ₂ | 0.2874 (2) | 0 | 0 |
| ΔH_f 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, ΔH_f SN = ± 105 KJ)

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, ΔH_f SN = ± 105 KJ)

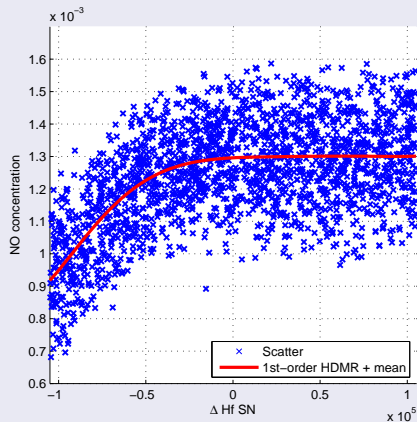
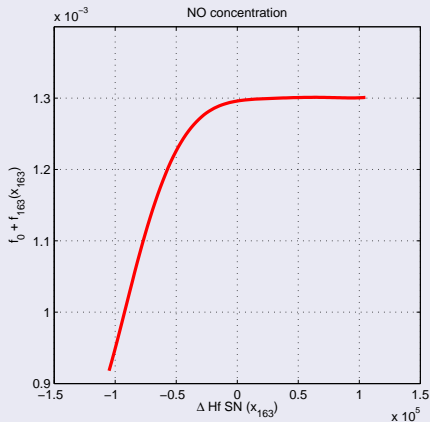
First-order sensitivity indices

- 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)

| Parameter | RS-HDMR | | Screening + RS-HDMR | |
|-------------------------------|---------|--------|---------------------|--------|
| | S_i | (Rank) | S_i | (Rank) |
| ΔH_f 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) |
| ΔH_f SO | 0.0086 | (5) | 0.0087 | (5) |
| $\sum S_i$ | 0.9524 | | 0.9590 | |
| S | 0.9695 | | 0.9755 | |

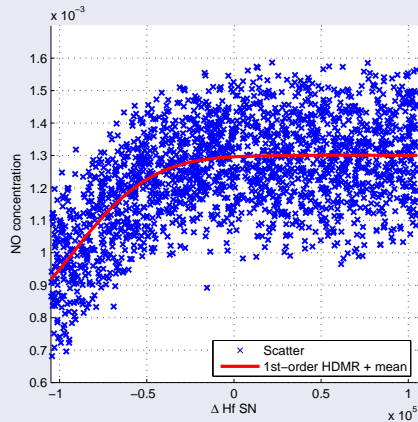
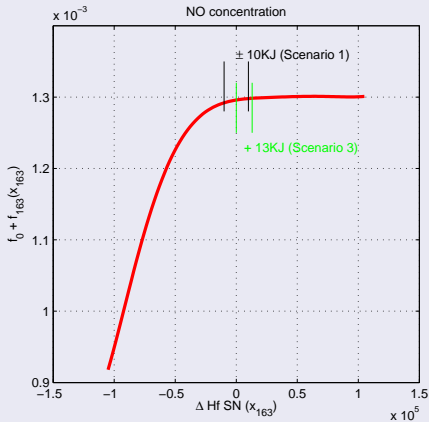
Results - Scenario 2 (Burcat table, $\Delta H_f \text{ SN} = \pm 105 \text{ KJ}$)

First-order RS-HDMR component functions and scatter plot

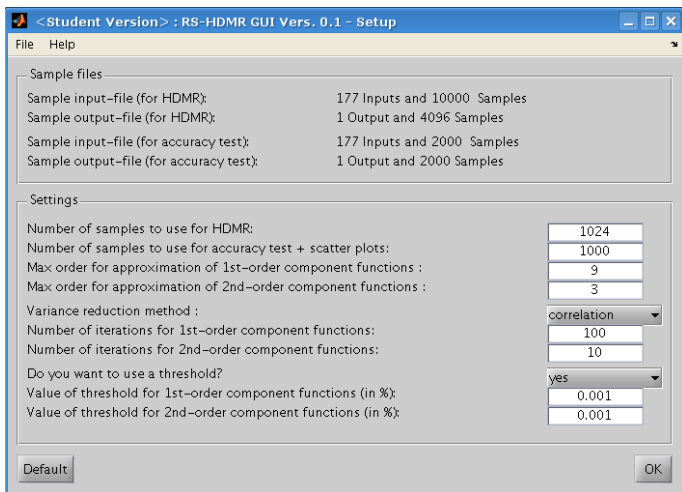


Results - Scenario 2 (Burcat table, $\Delta H_f \text{ SN} = \pm 105\text{KJ}$)

First-order RS-HDMR component functions and scatter plot



Graphical User Interface for RS-HDMR



Graphical User Interface for RS-HDMR

<Student Version> : RS-HDMR GUI Vers. 0.1 - Results

File Help

Results

1st-order

2nd-order

Accuracy

1st-order

2nd-order

Output:

1

Exit

First-order component functions for output: 1

51 out of 177 first-order component functions are computed to be non-zero

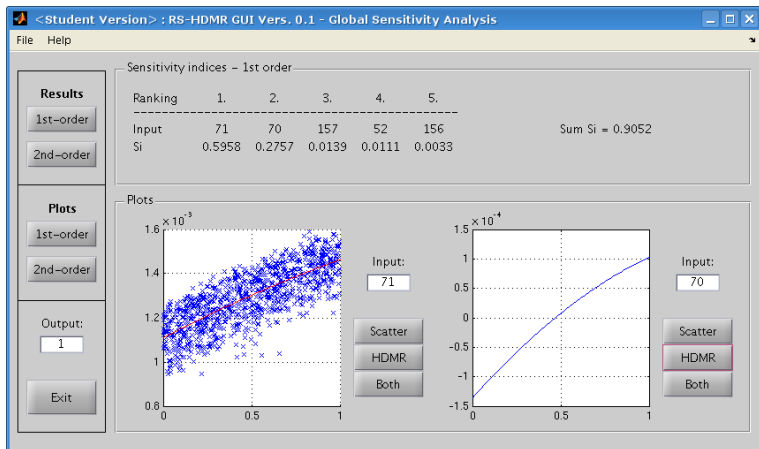
Component functions approximated by

1st-order polynomials: 25
 2nd-order polynomials: 17
 3rd-order polynomials: 6
 4th-order polynomials: 1
 5th-order polynomials: 1
 6th-order polynomials: 0
 7th-order polynomials: 0
 8th-order polynomials: 0
 9th-order polynomials: 1
 10th-order polynomials: 0

Accuracy - Relative Error

| | | | |
|--------|--------|---------|-------|
| 1% RE: | 68.5 % | 10% RE: | 100 % |
| 5% RE: | 99.8 % | 20% RE: | 100 % |

Graphical User Interface for RS-HDMR



Conclusions

- RS-HDMR method provides straightforward 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