The Effects of Parametric Uncertainties in Simulations of a Reactive Plume using Lagrangian Stochastic Models

Tilo Ziehn
Nick S. Dixon
Alison S. Tomlin

School of Process, Environmental and Materials Engineering
Energy and Resources Research Institute
University of Leeds

HARMO 12, 6 October 2008

Outline

1. Introduction
2. Model Validation and Uncertainty Analysis
3. Extended Chemical Mechanism
4. Global Sensitivity Analysis
5. Results
6. Conclusions

Outline

1. Introduction
2. Model Validation and Uncertainty Analysis
3. Extended Chemical Mechanism
4. Global Sensitivity Analysis
5. Results
6. Conclusions

Outline

1. Introduction
2. Model Validation and Uncertainty Analysis
3. Extended Chemical Mechanism
4. Global Sensitivity Analysis
5. Results
6. Conclusions

Outline

1. Introduction
2. Model Validation and Uncertainty Analysis
3. Extended Chemical Mechanism
4. Global Sensitivity Analysis
5. Results
6. Conclusions

Outline

1. Introduction
2. Model Validation and Uncertainty Analysis
3. Extended Chemical Mechanism
4. Global Sensitivity Analysis
5. Results
6. Conclusions

Outline

1. Introduction
2. Model Validation and Uncertainty Analysis
3. Extended Chemical Mechanism
4. Global Sensitivity Analysis
5. Results
6. Conclusions

Modelling of Atmospheric Plumes

Lagrangian Stochastic (LS)-Models

- LS-models are widely used to model concentrations and concentration fluctuations in atmospheric plumes
- Here: combined LS-model with a micro-mixing sub-model (Dixon & Tomlin 2007) using the interaction by exchange with the mean (IEM) mechanism
- Can also be applied for predicting concentration fluctuations in urban areas
- Based on the marked particle model which uses the formulation of Thomson (1987) for inhomogeneous turbulent flows

Model Validation

Wind Tunnel Experiment

- Wind tunnel experiment by Brown & Bilger (1996) is used to provide the first case study
- Nominal mean axial velocity of the flow: $\bar{U} = 0.5\,\mathrm{m\,s^{-1}}$
- Background O_3 concentration: $2.45 \cdot 10^{13}\,\mathrm{mol\,cm^{-3}}$
- Source NO concentration: $1.26 \cdot 10^{16}\,\mathrm{mol\,cm^{-3}}$
- Reaction: $NO + O_3 \longrightarrow NO_2 + O_2$
- No photolysis reactions included due to the absence of ultraviolet light

Uncertainty Analysis (1)

Structure Function Coefficient c_0

- c_0 determines diffusion in the velocity space and plays an important role in Lagrangian modelling
- Wide range of values can be found in the literature, e.g.
  - Rizza et al. (2006): $c_0 = 4.3 \pm 0.3$
  - Du (1997): $c_0 = 3.0 \pm 0.5$
  - Lien and D'Asaro (2002): $c_0 = 5.5$ and $c_0 = 6.4$
  - Reynolds (1998): $c_0 = 5.0 \pm 0.5$
- Low value of $c_0 = 3$ is questioned by Reynolds (1998)
- Therfore, it is of interest to explore the impact of the uncertainties in c_0
- Based on the studies we use $c_0 = [3 \ldots 6]$ (nominal value $c_0 = 5$)

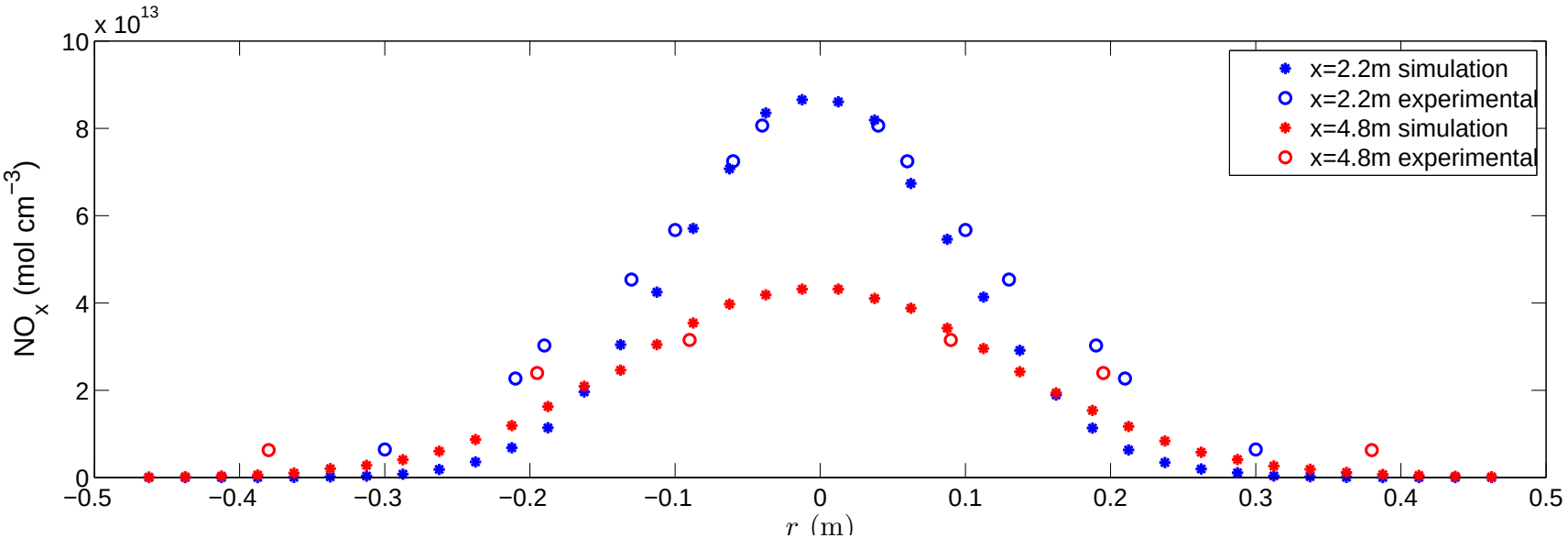
Uncertainty Analysis (2)

Mixing Time Scale Coefficient α and Source Size σ

- Simple scheme is used in the LS-model to relate mixing time scale t_m to turbulence timescale: $t_m = \alpha \frac{k}{\epsilon}$
- Review of the range of values for α is given in Cassini et al. (2005)
- Based on this we use $\alpha = [0.6 \ldots 3]$ (nominal value $\alpha = 0.75$)
- Shortcoming of the IEM model: mean concentration field is influenced by the micro-mixing (further explored using sensitivity analysis)
- Gaussian distribution is assumed for the release from the source
- Range for σ is estimated to represent the uncertainty in the effect of near field mixing on the initial source width
- Here we use $\sigma = [0.008 \ldots 0.016]$m (nominal value $\sigma = 0.008\,$m)

Radial Profile Conserved Scalar NO_x

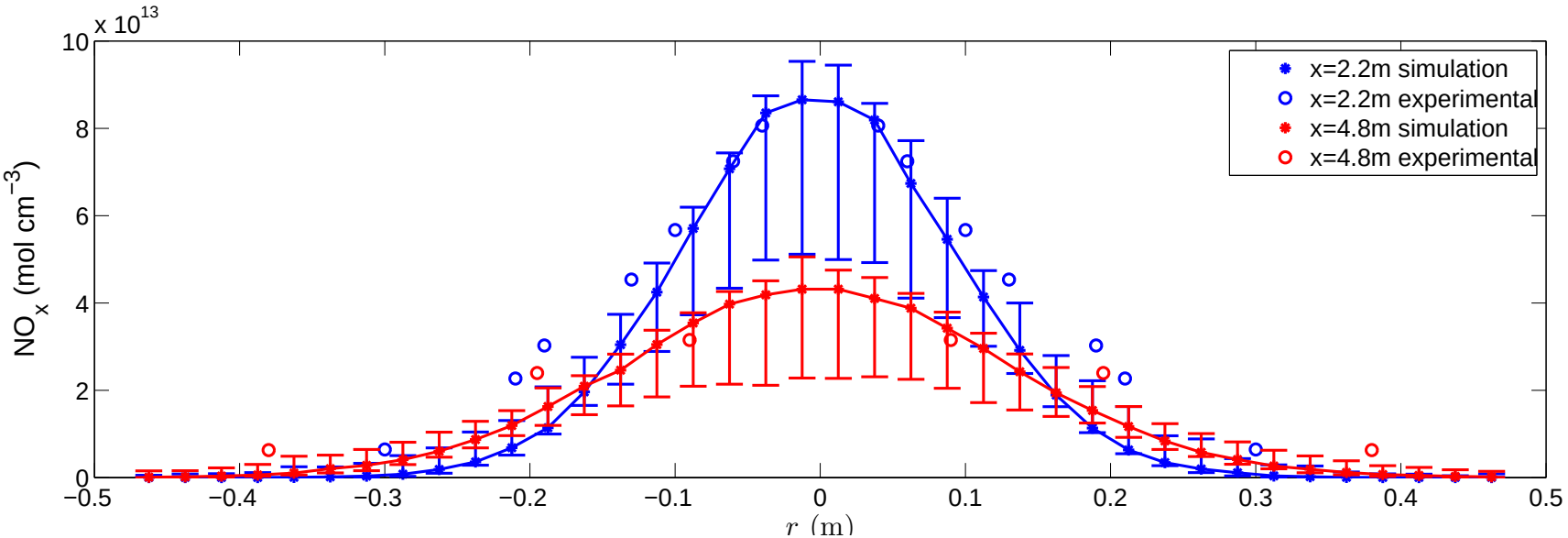
- Using the nominal values LS-model gives a good representation of the radial profile, however simulated profile is slightly narrower
- 400 simulations to produce error bars using random values within the uncertainty ranges for the 3 parameters (c_0, α, σ)



- Range of predictions is large $\longrightarrow$ high degree of sensitivity to the input parameters (further explored using sensitivity analysis)

Radial Profile Conserved Scalar NO_x

- Using the nominal values LS-model gives a good representation of the radial profile, however simulated profile is slightly narrower
- 400 simulations to produce error bars using random values within the uncertainty ranges for the 3 parameters (c_0, α, σ)



- Range of predictions is large $\longrightarrow$ high degree of sensitivity to the input parameters (further explored using sensitivity analysis)

Chemical Mechanism

Reaction Scheme

- Eight reactions between the species NO, O, $\mathrm{O_3}$ and $\mathrm{NO_2}$
- Photolysis included for $\mathrm{O_3}$ and $\mathrm{NO_2}$

$$\begin{aligned}
&\text{R1:} \quad \mathrm{O} \xrightarrow{k_1} \mathrm{O_3} && \text{R5:} \quad \mathrm{O} + \mathrm{NO_2} \xrightarrow{k_5} \mathrm{NO} \\
&\text{R2:} \quad \mathrm{O} \xrightarrow{k_2} \mathrm{O_3} && \text{R6:} \quad \mathrm{NO} + \mathrm{O_3} \xrightarrow{k_6} \mathrm{NO_2} \\
&\text{R3:} \quad \mathrm{O} + \mathrm{O_3} \xrightarrow{k_3} 2\mathrm{O_2} && \text{R7:} \quad \mathrm{O_3} \xrightarrow{k_7} \mathrm{O} \\
&\text{R4:} \quad \mathrm{O} + \mathrm{NO} \xrightarrow{k_4} \mathrm{NO_2} && \text{R8:} \quad \mathrm{NO_2} \xrightarrow{k_8} \mathrm{NO} + \mathrm{O}
\end{aligned}$$

- Rate equations are implemented in the Lagrangian stochastic model

Setup

Uncertain Parameters

- Study of the combined effects of uncertainties in physical and chemical parameters
- 22 parameters assumed to be uncertain
- Physical parameters: σ, α and c_0 using the uncertainty ranges introduced earlier
- Chemical reaction rate parameters for each reaction: A-factor, activation energy E and temperature coefficient for Arrhenius parameters or photolysis rate
- Temperature: [273 . . . 293] K
- NO fraction in the source: 100 % to 80 %
- Ozone concentration: $[7.35 \cdot 10^{11} \ldots 1.225 \cdot 10^{12}]\,\mathrm{mol\,cm^{-3}}$

High Dimensional Model Representation (HDMR)

Random Sampling (RS)-HDMR

- HDMR was developed to express the input-output relationship of a complex model with a large number of parameters
- Allows the investigation of the effects caused by individual parameters ($f_i(x_i)$) and parameter interactions (e.g. $f_{ij}(x_i, x_j)$) upon the model output
- Requires only one set of random or quasi-random samples

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

Conserved Scalar $\mathrm{NO_x}$ (1)

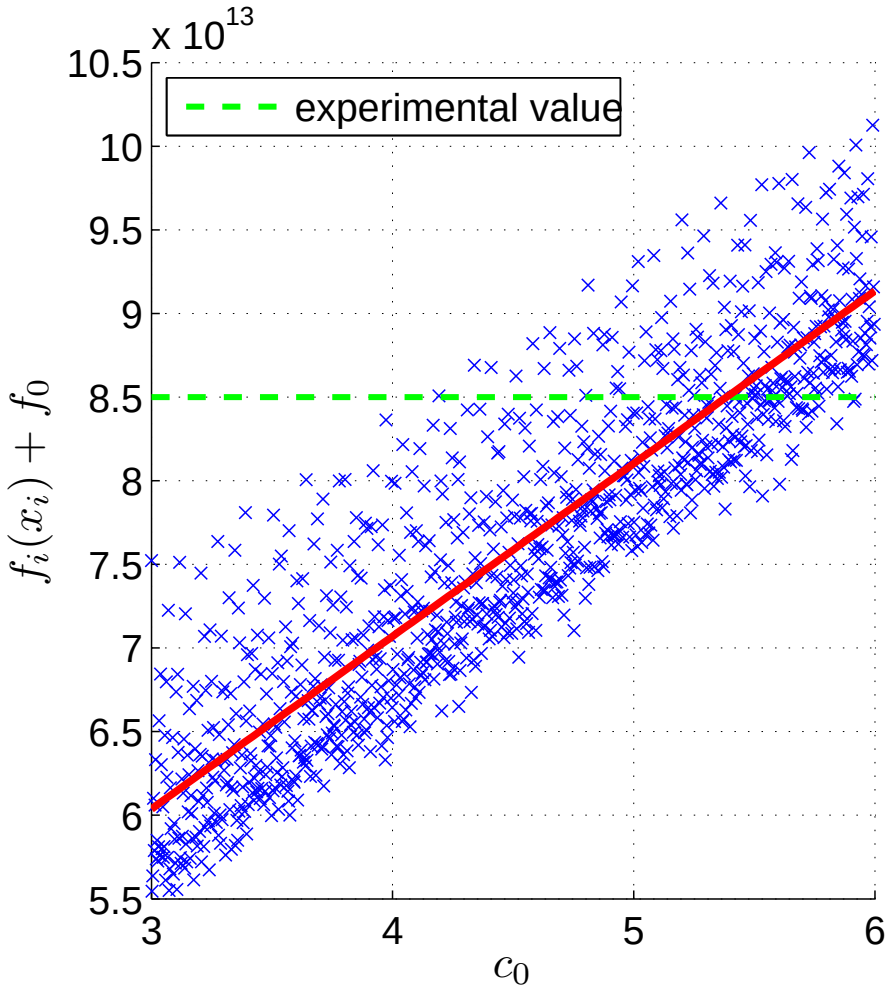
RS-HDMR using $N = 2048$ quasi-random samples

- Output of interest: Concentration of the conserved scalar $\mathrm{NO_x}$ at the plume centre

Parameter	S_i $(x = 2.2\,\mathrm{m})$	S_i $(x = 4.8\,\mathrm{m})$	S_i $(x = 5.8\,\mathrm{m})$
Structure function coefficient c_0	0.7997	0.8039	0.8023
Mixing time scale coefficient α	0.1868	0.1864	0.1876
$\sum S_i$	0.9866	0.9903	0.9900

- Mixing time scale coefficient has significant effect on $[\mathrm{NO_x}] \longrightarrow$ IEM model breaks one of the desireable properties of a mixing model: the mean concentration should be uneffected by the mixing model

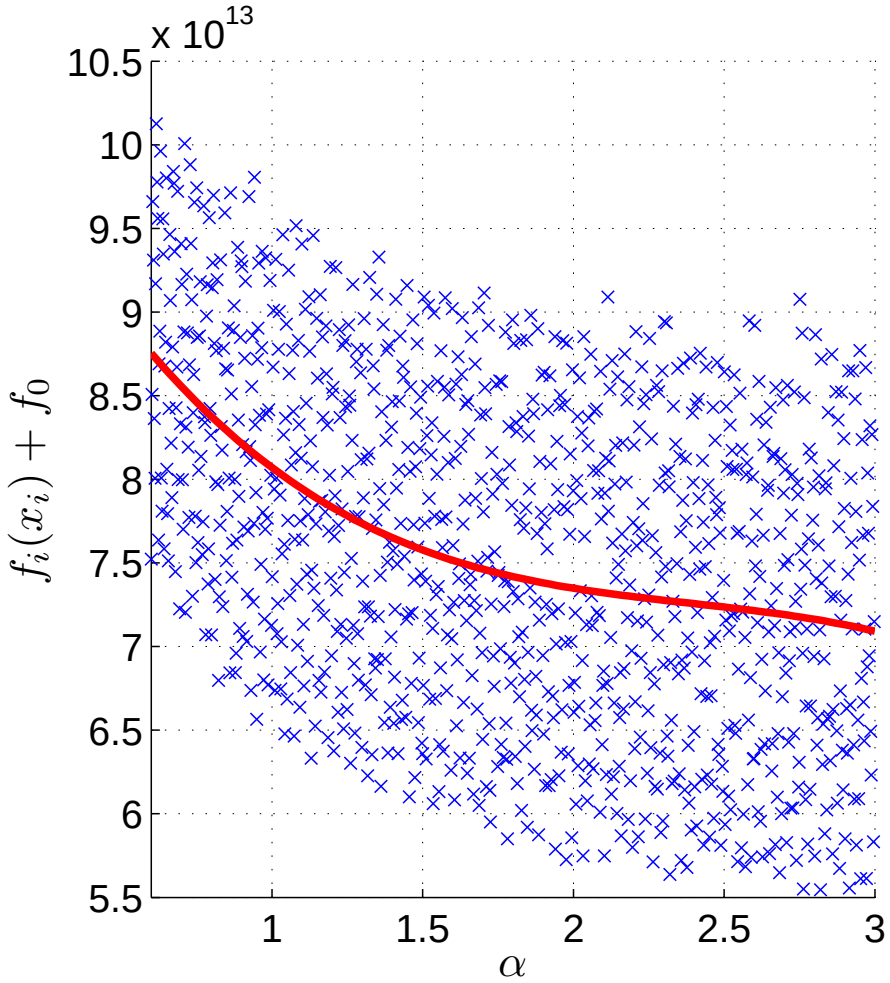
Conserved Scalar NO_x (plume centre, x=2.2m)



Results

- Strong positive linear relationship
- Most important parameter, causes around 80 % of the overall variance
- Experimental value can not be recovered with a value of $c_0 < 4$, even taking into account the uncertainties in the other parameters (in agreement with Reynolds (1998))

Conserved Scalar NO_x (plume centre, x=2.2m)



Results

- Negative non-linear relationship
- Causes nearly 18 % of the overall variance
- High sensitivity for values of $\alpha < 1.5$

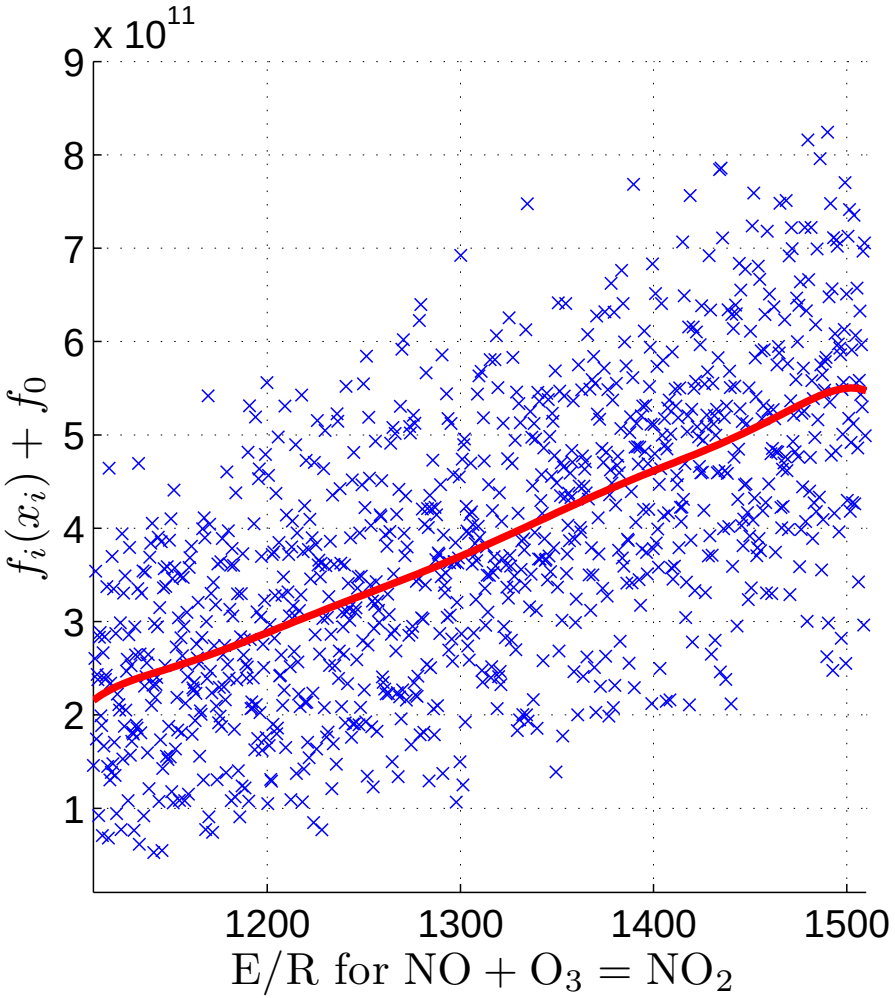
Ozone concentration (1)

RS-HDMR using $N = 2048$ quasi-random samples

- Output of interest: Ozone concentration $[O_3]$ at the plume centre

Parameter	S_i ($x = 2.2\,\mathrm{m}$)	S_i ($x = 4.8\,\mathrm{m}$)	S_i ($x = 5.8\,\mathrm{m}$)
E/R for $NO + O_3 \longrightarrow NO_2$	0.4086 (1)	0.4302 (1)	0.4330 (1)
Mixing time scale coefficient α	0.3030 (2)	0.2693 (2)	0.2594 (2)
Background concentration for O_3	0.1057 (3)	0.0749 (4)	0.0708 (4)
Fraction of NO in total $[NO_x]$	0.0591 (4)	0.0776 (3)	0.0834 (3)
Temperature	0.0335 (5)	0.0359 (6)	0.0362 (6)
Structure function coefficient c_0	0.0296 (6)	0.0363 (5)	0.0373 (5)
A factor for $NO + O_3 \longrightarrow NO_2$	0.0276 (7)	0.0292 (7)	0.0295 (7)
$\sum S_i$	0.9745	0.9643	0.9620

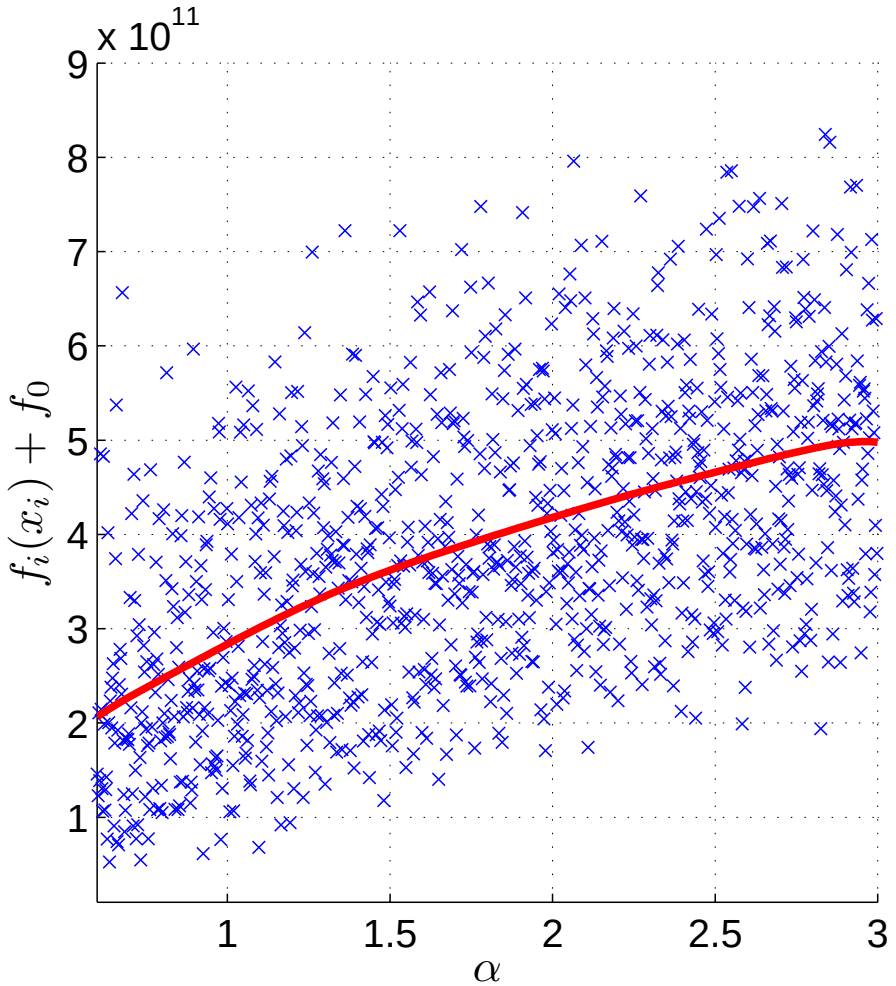
Ozone concentration (plume centre, x=2.2m)



Results

- Positive, nearly linear relationship
- Most important parameter, causes more than 40 % of the overall variance
- Importance of the parameter increases with growing distance from the point source

Ozone concentration (plume centre, x=2.2m)



Results

- Positive, non-linear relationship
- Second most important parameter, causes around 30 % of the overall variance
- Importance of the parameter decreases with growing distance from the point source

Conclusions and Future Work

Conclusions

- Variance in the mean concentration of the conserved scalar $\mathrm{NO_x}$ at the plume centre is as expected only influenced by the turbulence parameters (structure function coefficient c_0 dominates and causes 80 % of the variance)
- Variance in the mean Ozone concentration is influenced by both physical and chemical parameters
- Relative importance of α increases when the chosen target output is a secondary species rather than a conserved scalar

Future Work

- Investigation of the reactive plume over a larger range
- Application of the reactive plume model to an urban area

GUI-HDMR - a Tool for Global Sensitivity Analysis

- GUI-HDMR was developed to combine existing RS-HDMR tools and RS-HDMR extensions and make them easily available for a wide range of users
- Software is written in Matlab and requires the basic Matlab package