2-1, 2-2
Model uncertainties, sensitivity analysis and optimisation

Intro

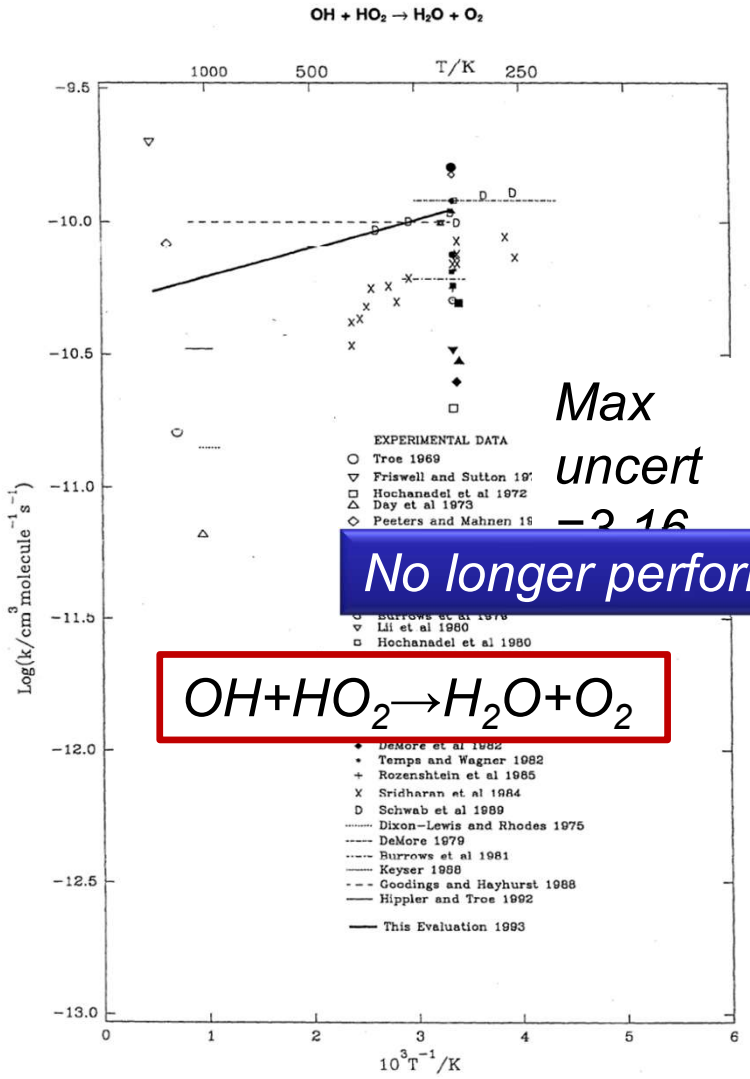
- Presenting uncertainties in experimental data is far more common than for modelling results.
- But we need to know the **robustness** of our models to use them in design and decision making e.g. for use of new fuels, design of new gas turbines and marine engines with low carbon fuels.
- Need methods for **uncertainty quantification**.
  - Emerging field of UQ in combustion modelling.
  - Need first to estimate uncertainties in
    - Input parameters
    - Model structure.
  - Efficient methods for **propagating** through model to provide uncertainties on target outputs.

Sources of model input data and uncertainty information

- Kinetic evaluations (e.g. Baulch, Tsang, Atkinson).
- NIST data base.
- Output from theoretical studies.
- Trawling literature for individual papers on rate coefficients etc.…. time consuming!
- Detailed statistical studies
  - Active tables for thermodynamic data (Ruscic and coworkers).
  - Optimisation studies for reaction mechanisms (Nagy and coworkers).
  - New approaches to AMG using ML with uncertainty quantification.
- What to do about estimated parameters?

Evaluated data (Baulch et al., 1994)

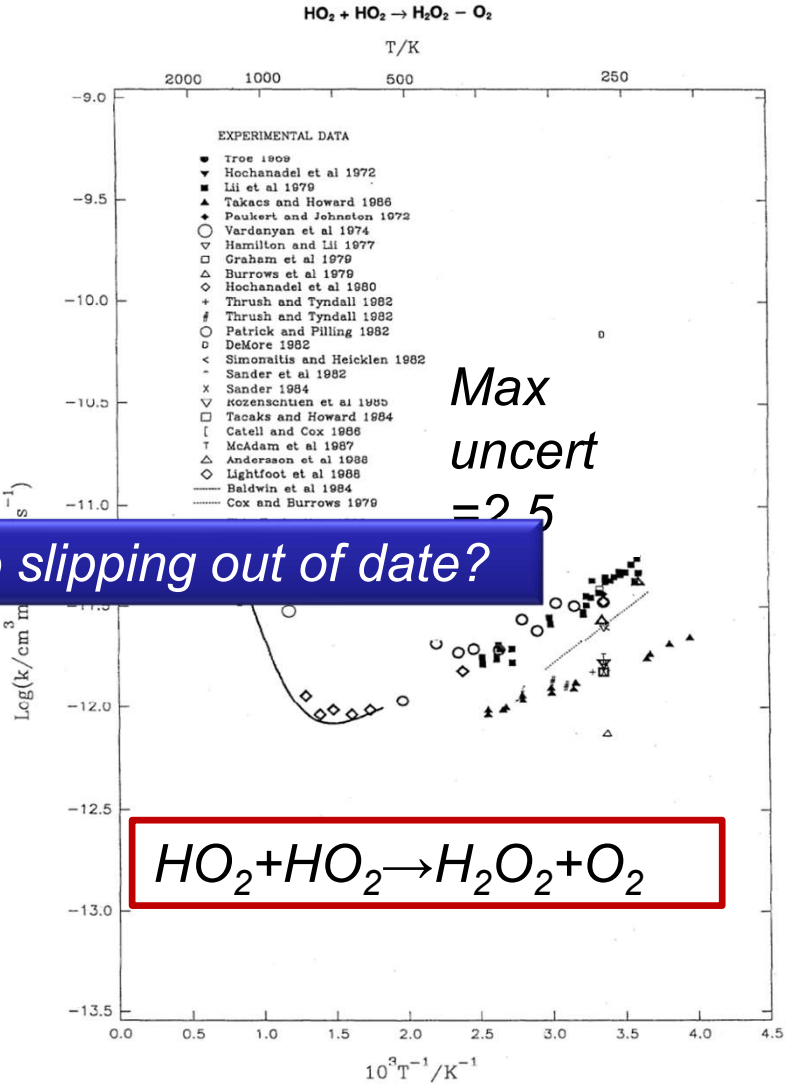
Δlogk = ±0.2 (300K) ±0.5 (2000K)



Max uncert =3.16

$OH+HO_2 \rightarrow H_2O+O_2$

Δlogk = ±0.15 (550-800K) ±0.4 (1250K)



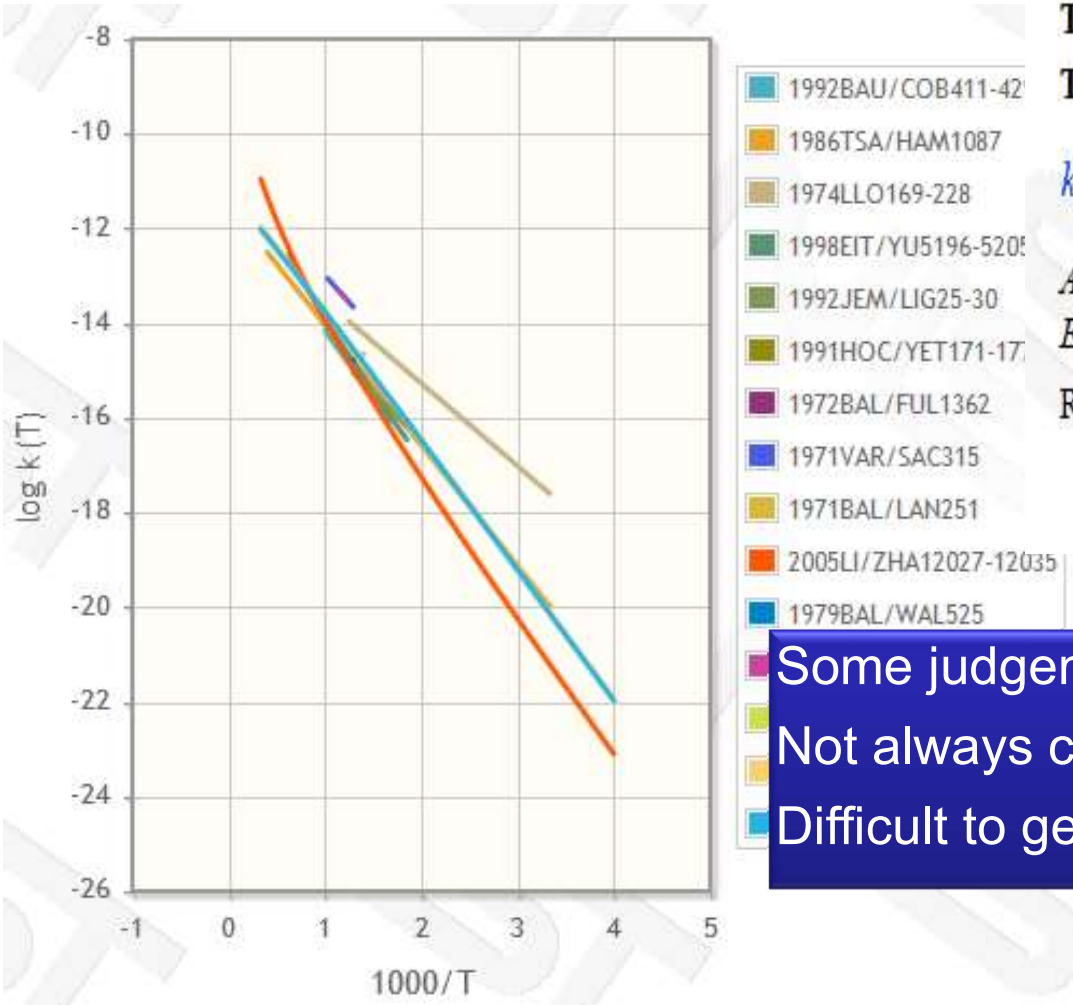
Max uncert =2.5

$HO_2+HO_2 \rightarrow H_2O_2+O_2$

No longer performed so slipping out of date?

NIST data base

$HCHO+HO_2 \rightarrow HCO+H_2O_2$



Fit of Arrhenius parameters to set:

Temperature range: 250 - 3000 K

Two-parameter fit:

$k(T) = A\exp(-E_a/RT)$

A = 9.25E-12 [cm^3/molecule s]

E_a = 52.17 [kJ]

RMSD = 4.3

Three-parameter fit:

$k(T) = A(T/T_{ref})^n \exp(-E_a/RT)$

A = 1.51E-11 [cm^3/molecule s]

n = -0.3

T_{ref} = 298 [K]

E_a = 53.48 [kJ]

RMSD = 4.2

Some judgement has to be made about outliers

Not always completely up to date

Difficult to get temperature dependant uncertainties

Representations of Uncertainty

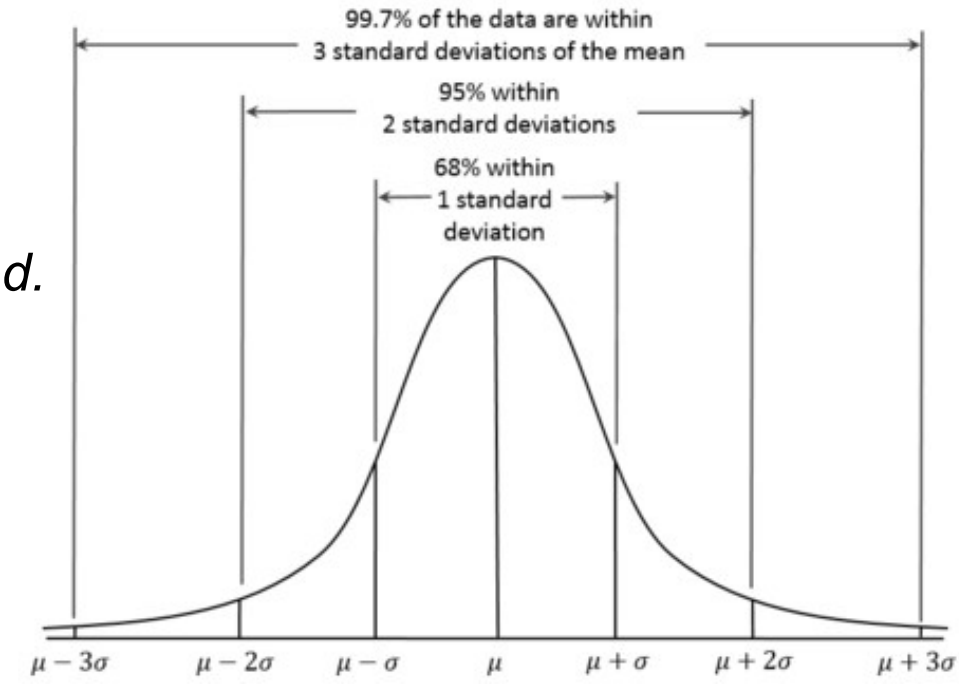
- Depends of level of knowledge about a particular parameter.
- If evaluation available then *f* value may be given.

$$f = \log_{10}\left(\frac{k^0}{k^{\min}}\right) = \log_{10}\left(\frac{k^{\max}}{k^0}\right) \qquad \frac{k^{\max}}{k^0} = 10^f$$

- k_0 recommended value of rate coefficient. k_{min}, k_{max} extreme values.

$\sigma^2(\ln\{k\}) = ((f \ln 10)/m)^2$

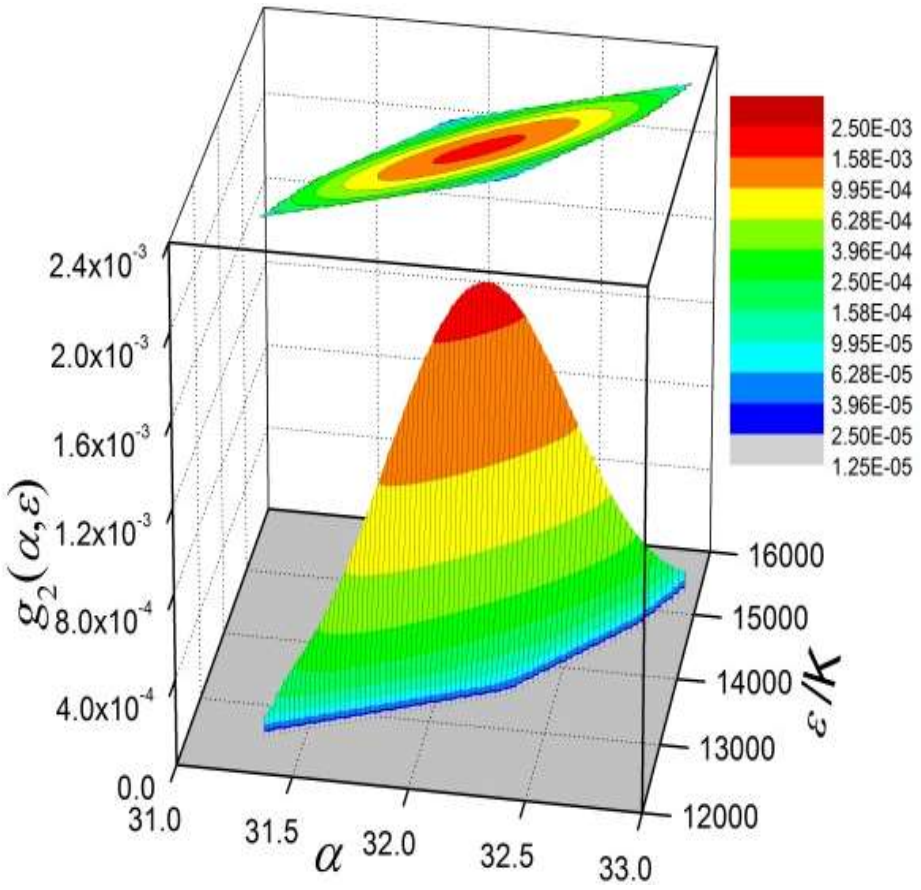
where m is the level of uncertainty suggested.
m = 2, 2σ deviation or 95 %ile,
m= 3, 3σ deviation or 99.7 %ile



Examples of levels of uncertainty

uncertainty parameter *f*	multiplication factor of 3σ uncertainty limits	σ($\log_{10}$ k)	σ(ln k)	multiplication factor of 1σ	multiplication factor of 2σ
0.1	1.26	0.03	0.08	1.09 (9%)	1.17
0.3	2.00	0.10	0.23	1.33 (33%)	1.67
0.5	3.16	0.17	0.38	1.72	2.44
0.7	5.01	0.23	0.54	2.34	3.67
0.9	7.94	0.30	0.69	3.31	5.63
1.0	10.00	0.33	0.77	4.00	7.00

Statistical methods

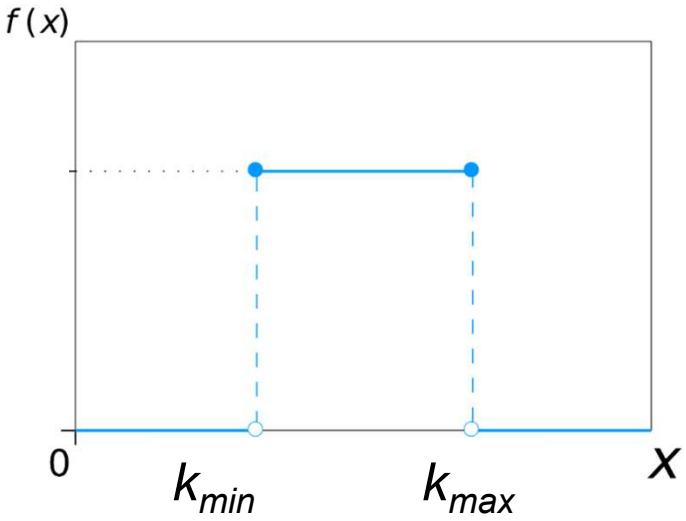


- All available current information on a system is used fit a joint pdf of parameters
  - ➢ e.g. 2,3 parameter Arrhenius
- Provides highly detailed data on parameter correlations etc.

For reaction kinetics has been carried out for only a few simple systems e.g. H_2, wet CO oxidation, methanol (Nagy et al., 2011).

Estimated parameters

- For estimated parameters 10^f is likely to be a **guestimate** e.g. factor of 2 or a factor of 5.
- A **uniform distribution** used since no probabilistic information likely to be available.
- In future – would be useful to provide estimates based on **reaction classes** from AMG packages like RMG etc.
  - What is the uncertainty for a particular reaction class based on available data?
  - Or even for individual reactions based on the machine learning approaches discussed earlier.
  - How does the uncertainty change as e.g. the number of carbons grows?



Active Tables

- New paradigm to develop accurate, reliable, and internally consistent thermochemical values for stable, reactive, and transient chemical species by utilizing to the fullest all available experimental measurements as well as state-of-the art theoretical data.
- **ATcT** is based on constructing, analysing, and solving the underlying Thermochemical Network (TN).
- Brings together both experimental and theoretical studies (**see earlier**) to reduce uncertainties in data (Burcat & Ruscic, 2005).
- Network of Computed Reaction Enthalpies to Atom-Based thermochemistry **(NEAT) (***Csaszar and Furtenbacher, 2010)*
- **Results in highly correlated parameters – be careful of the effects of neglecting such correlations!**

Species Name	Formula	$\Delta_f H°(0\ K)$	$\Delta_f H°(298.15\ K)$	Uncertainty	Units	Relative Molecular Mass	ATcT ID
Dihydrogen	H2 (g)	0	0	exact		2.01588 ± 0.00014	1333-74-0*0
Helium	He (g)	0	0	exact		4.0026020 ± 0.0000020	7440-59-7*0
Heptane	C7H16 (l)	-201.46	-223.91	± 0.74	kJ/mol	100.2019 ± 0.0057	142-82-5*500
Octane	C8H18 (l)	-226.61	-249.73	± 0.79	kJ/mol	114.2285 ± 0.0065	111-65-9*500
2,2,4-Trimethylpentane	(CH3)2CHCH2C(CH3)3 (l)	-224.4	-258.9	± 1.5	kJ/mol	114.2285 ± 0.0065	540-84-1*500

Part of a thermo-chemical network showing the basic ideas

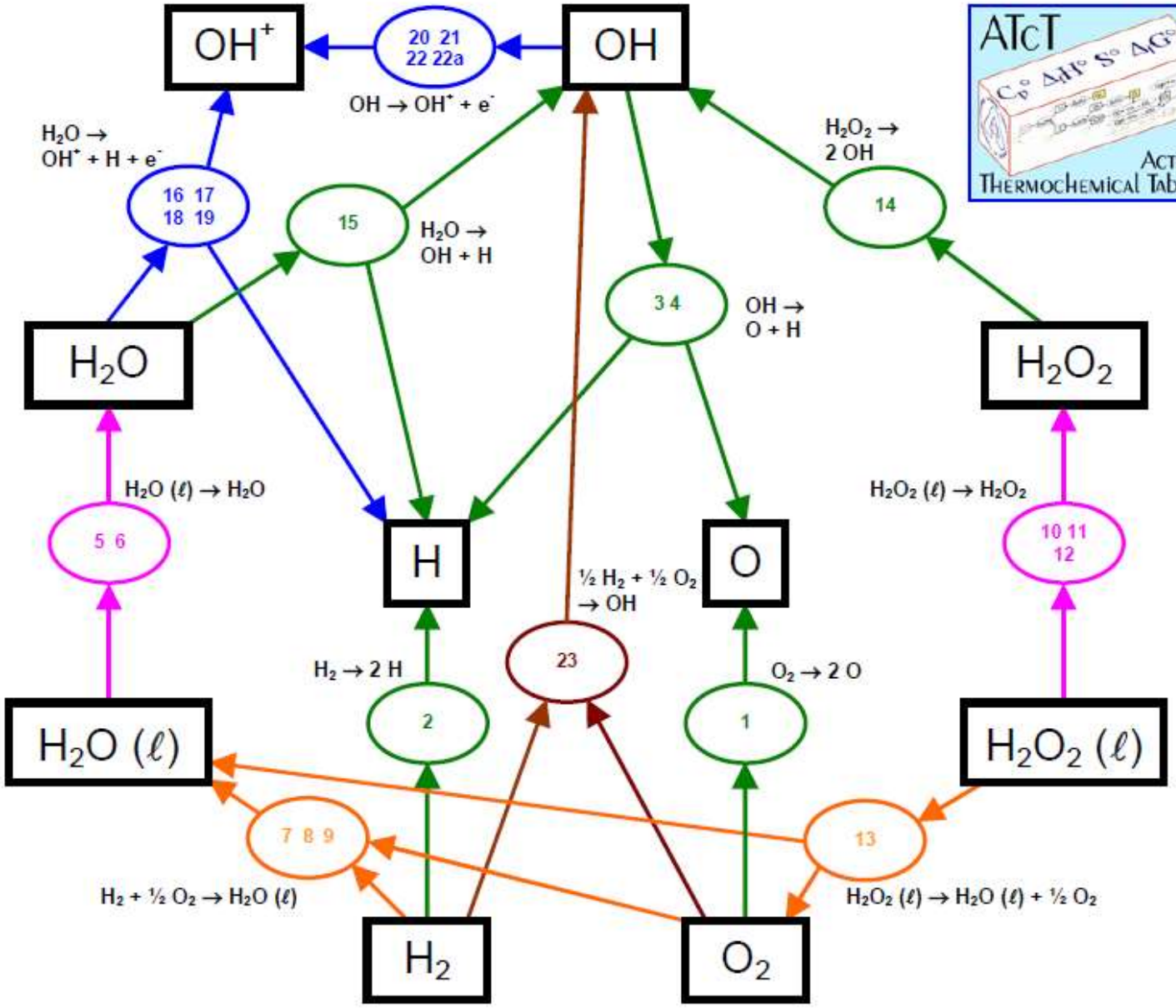
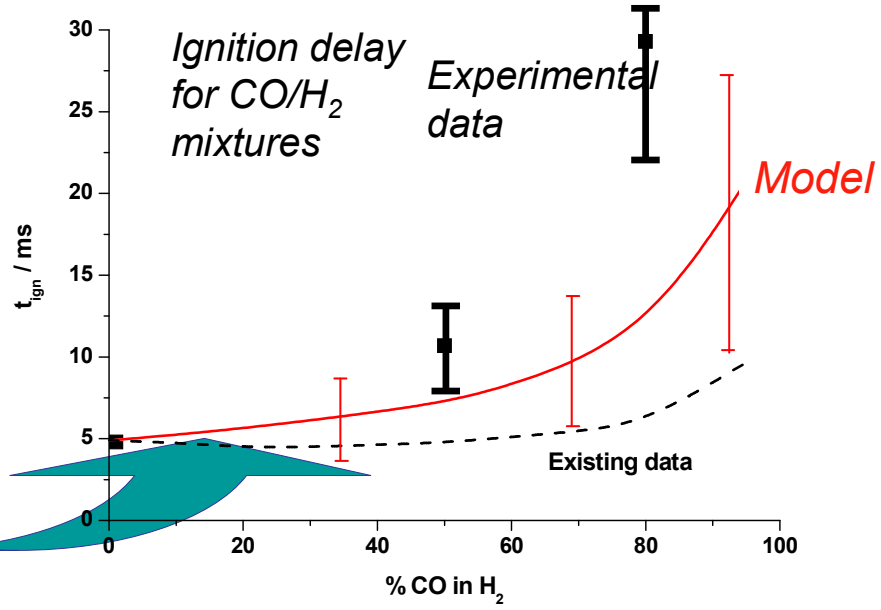


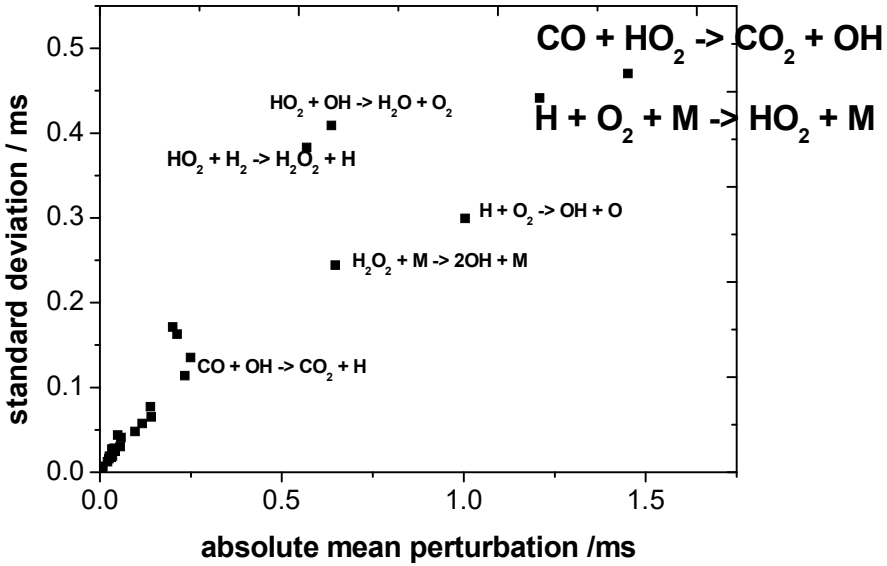
Figure 1. A small subsection of the current Core (Argonne) Thermochemical Network. The full network currently contains >600 primary vertices and >3200 secondary vertices. See text for further details

Sensitivity and uncertainty analysis

• **Uncertainty quantification (UQ)** estimates the overall predictive uncertainty of a model given the state/or lack of knowledge about its input parameters.

• UQ puts error bars on predictions.

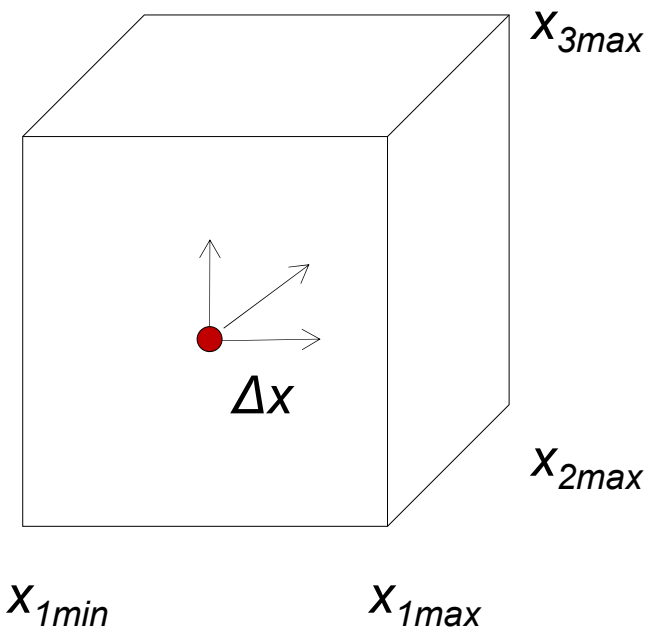




Sensitivity analysis (SA) determines how much each input parameter contributes to the output uncertainty (usually expressed as variance).

Local sensitivity coefficients

- *nominal values*



Local first-order sensitivity coefficient

$$S_{ij} = \frac{\partial Y_i}{\partial x_j}$$

Normalised first-order sensitivity coefficient

$$S'_{ij} = \frac{x_j}{Y_i}\frac{\partial Y_i}{\partial x_j}$$

Commonly incorporated into codes such as Chemkin, Cantera using finite difference methods.

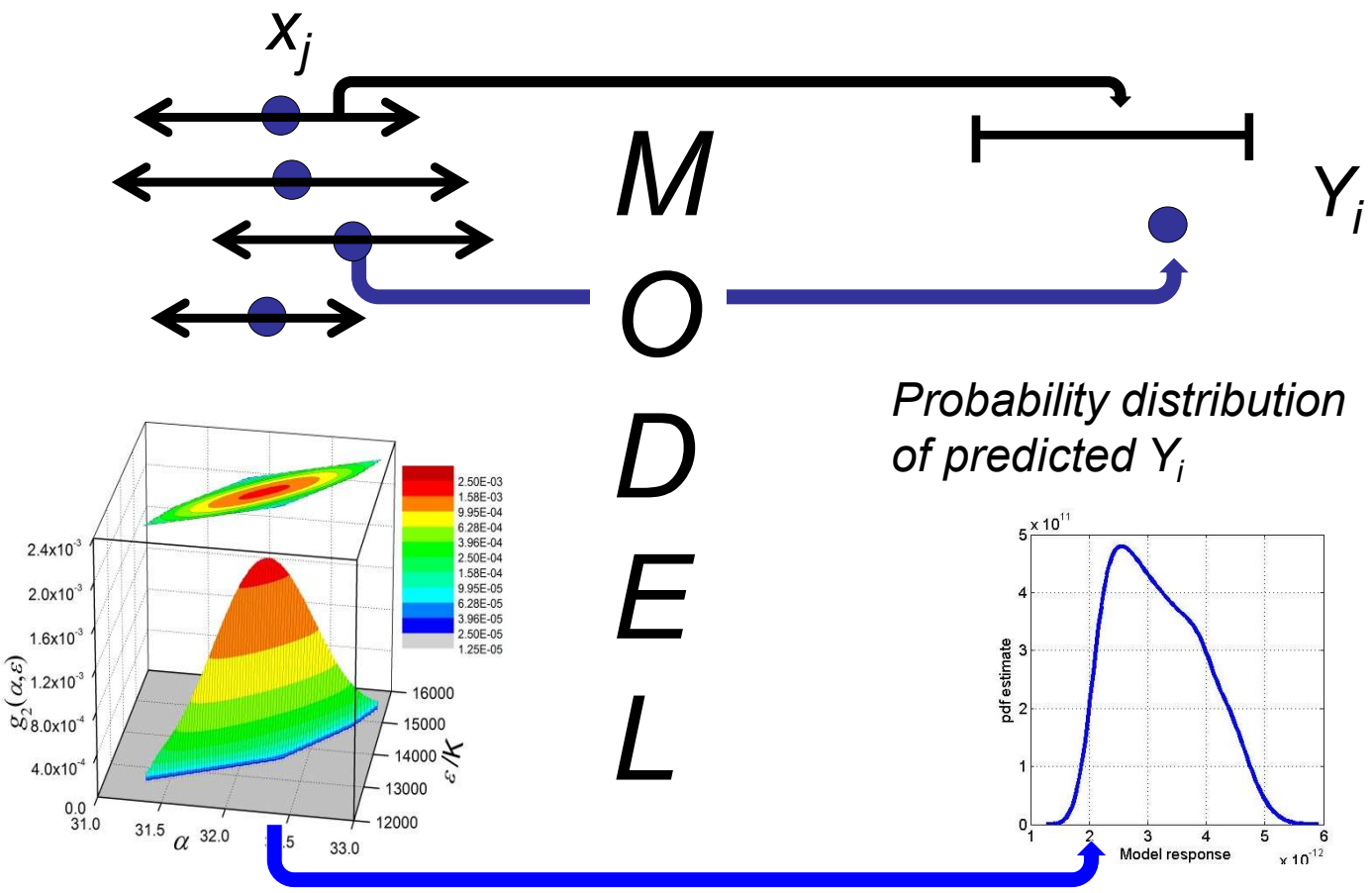
Contributions to uncertainty?

- Really we want to know, not just sensitivity, but also how much a parameter contributes to model uncertainty.
- Some parameters have **high sensitivity**, but are very **well quantified**.
- Others may have **lower sensitivity** but are **poorly known** and therefore drive potential errors in models.
- If $\sigma(x_j)$ are known – or estimated - then we can estimate overall uncertainty:

$$\sigma^2(\mathbf{Y}) = \sum_j (\mathbf{S}'_j)^2 \frac{\sigma^2(x_j)}{x^2{}_j}$$

- The **fractional contribution** of each parameter to this **uncertainty** can be estimated.
- Gives a better measure of parameter importance than S'_{ij} alone.
- Tells us how better quantification of each parameter could reduce overall modelling uncertainty.

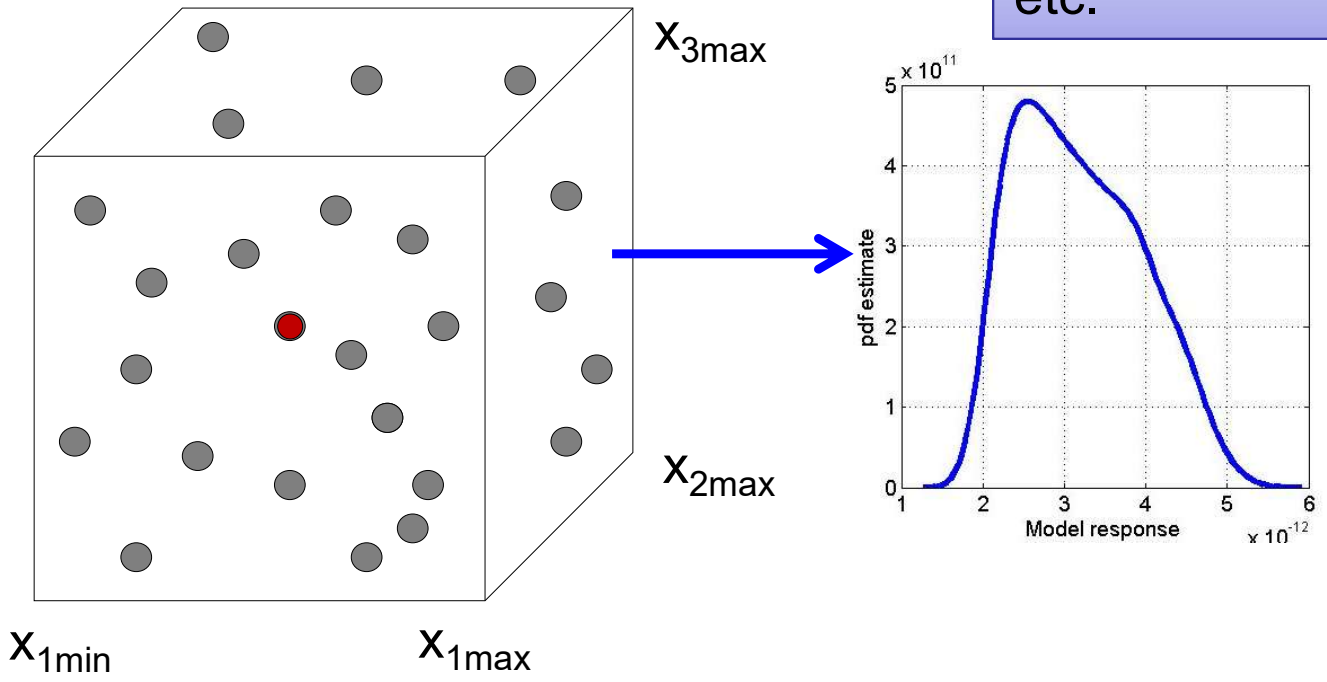
Local vs global methods



Global sensitivity/uncertainty methods

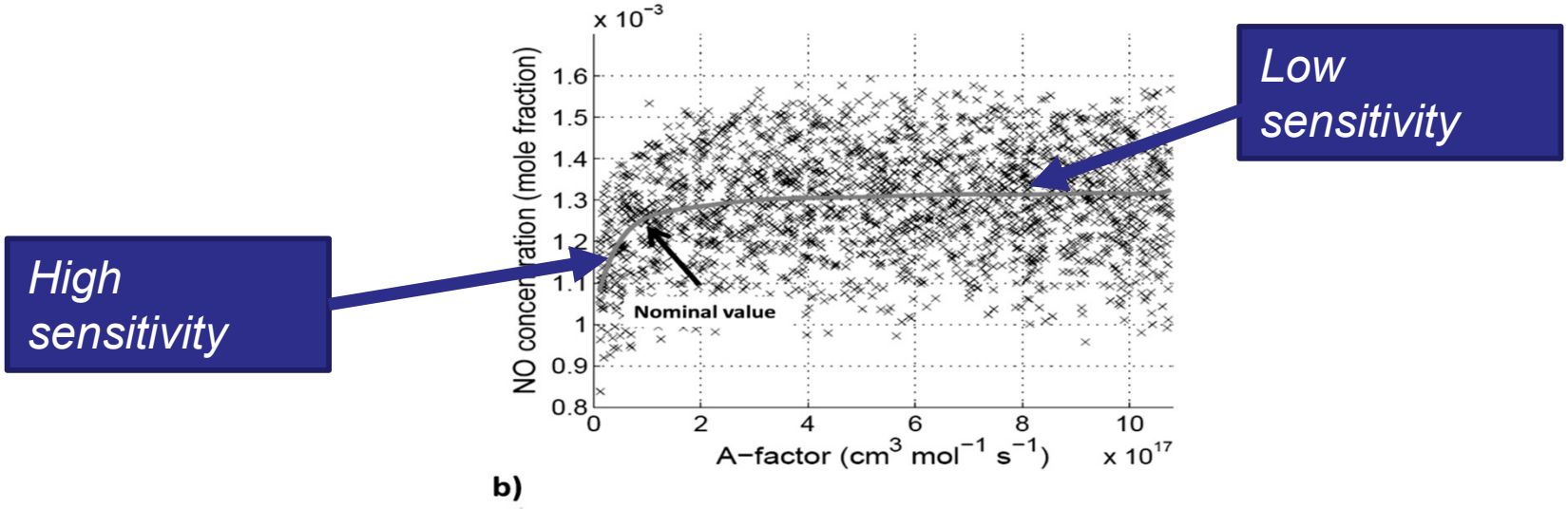
Global - attempts to cover whole input space using a sampling method.

Najm, Wang, Frenklach, Sheen, Tomlin, Turányi etc.



Why use global methods?

- Local sensitivity and uncertainty methods are usually based on a single (best estimate) value of the parameters.
- If the sensitivity of the output changes depending on the values of the parameters then local methods could be inaccurate.
- Particularly important for **non-linear models** and models with **large uncertainties**.



Disadvantages of global methods

- In order to cover the regions of parameter uncertainty, ***sampling based methods*** need to be used and therefore a ***large number of model runs*** is needed instead of the single run required for local sensitivity analysis using e.g. finite difference methods.
- The methods also require **prior knowledge of the input parameter distributions**.
- Methods are then required to interpret the data from a large number of samples to determine the sensitivity indices.
- For large parameter systems sample sparsity can be an issue.
- Screening methods are therefore often first applied to identify **unimportant** parameters which do not need to be varied in the full global approach.

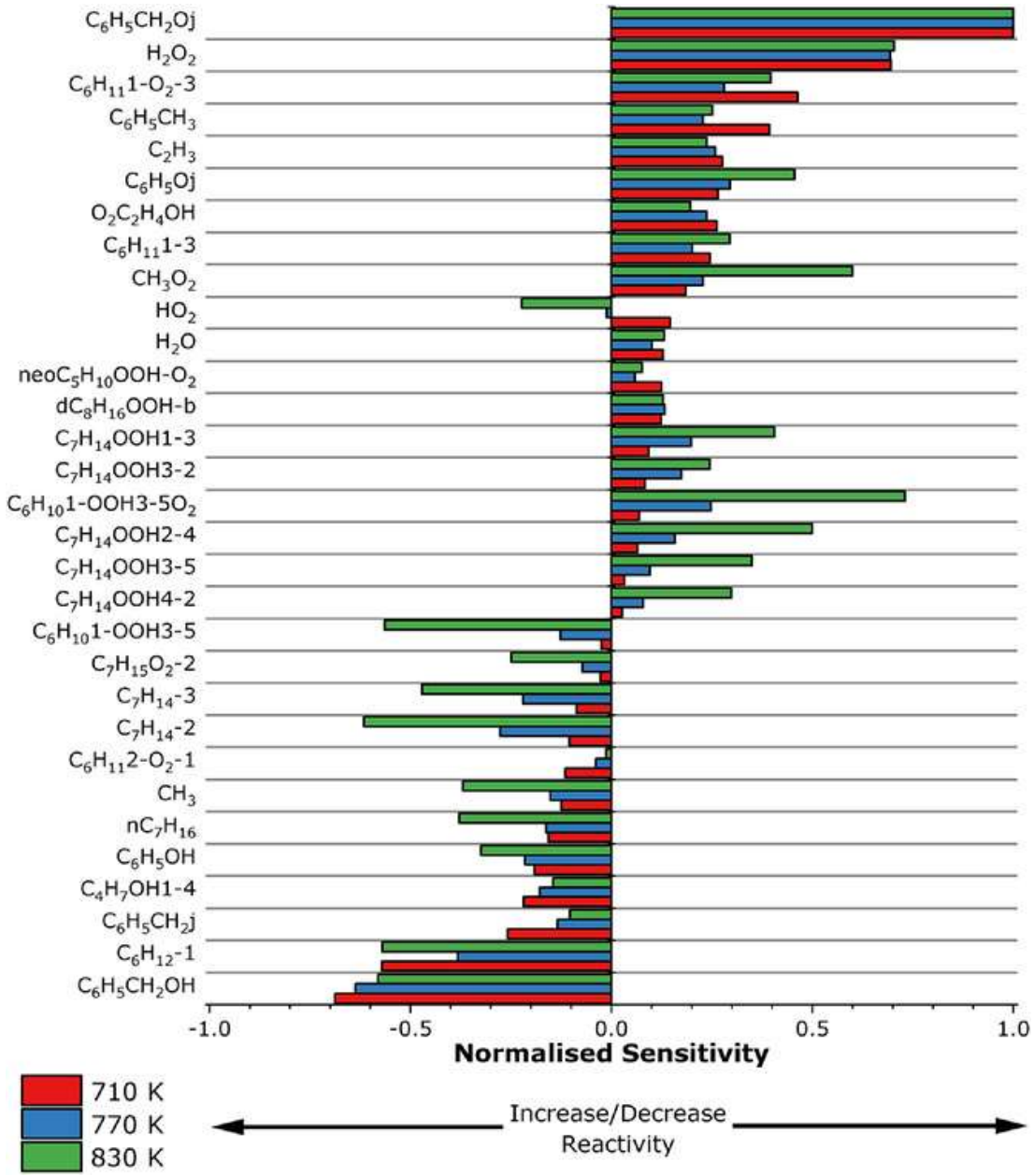
Screening methods.

- Can be based on **local sensitivity coefficients**.
- Problematic unless it is applied at various values of the nominal parameters e.g. recommended value, rv, rv x 2, rv x 0.5.
- Can be automatically calculated using simple finite differences approaches in Chemkin/Cantera for simple outputs such as peak *T*, peak [OH].
  - Adds to model run time but not significantly.
- Can be run as **Brute Force Method**, changing each parameter in scheme once and assessing sensitivity of target outputs.
  - Very expensive for large models, useful for targets such as IDTs.
- **Morris method** also used for screening and applies a one at a time method, changing one parameter by a fixed amount for each run of the model starting with a random seed.
- Several random seeds used at different points in parameter space.
  - Probably 10 times more expensive than even the Brute Force Method.

Brute Force Methods

- Useful for target outputs which do not have an obvious functional relationship to inputs e.g. ignition delay times.
- Each parameter of interest is modified in turn from the chosen nominal value by a small %, the model is re-run and the % change in output calculated.
- The relative output change gives a ranking of parameter importance.
- Cost is proportional to number of parameters *n*.
- Can be run for different chosen nominal values – pushing cost to 2*n*, 3*n* etc.
- Method does not consider non-linear interactions between parameters.
- Still, a straightforward way to screen out unimportant parameters before full global study.

Example



- Normalized sensitivity coefficients of IDTs on species enthalpy of formation, for a 5-component gasoline surrogate. Rapid compression machine,
  P_C = 20 bar, Φ = 1

Morris Method

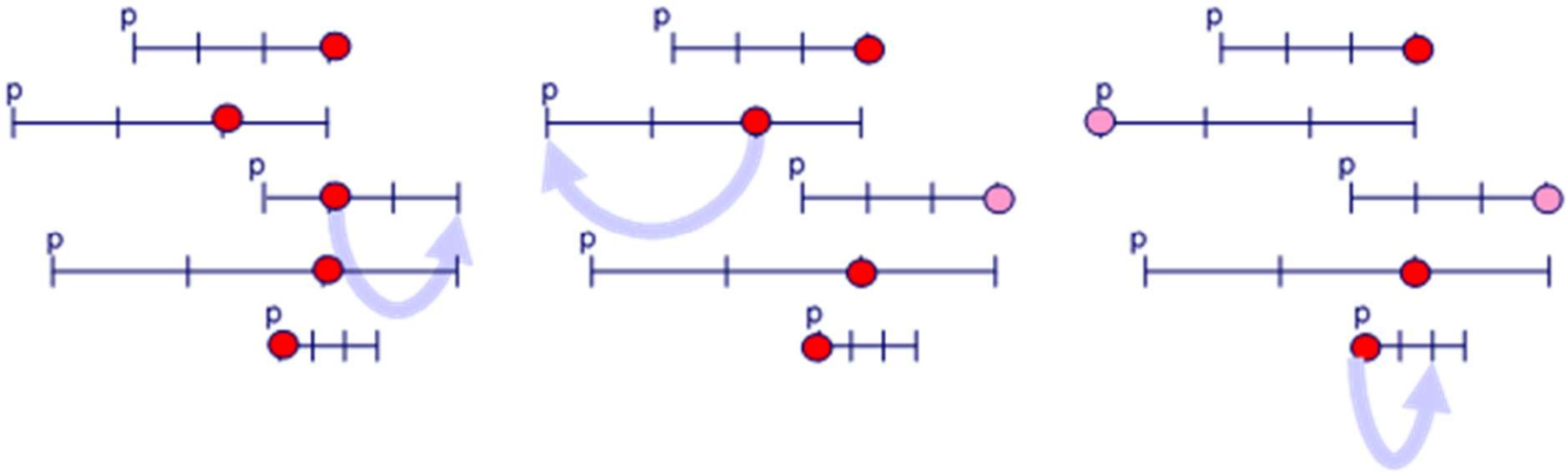
- Classified as global sensitivity method since entire parameter space is covered.
- Can also rank **degree of nonlinearity** of response to changes in each parameter via the standard deviation of the parameter effect across all parameter sets.
- Does not however provide functional dependency of output on individual parameters or parameter pairs.
- Mainly used to screen out unimportant parameters prior to the use of more informative global methods.
- The relative computational cost of the Morris Method vs. fully global sampling based methods depends on the number of important parameters in the system.

MM cont.

- A series of parameter sets are generated so that the next one differs from the previous in the value of a single parameter only, which is randomly chosen.
- The value of each parameter x_j is modified within the range $[x_j^{min}, x_j^{max}]$ by a fixed amount Δ determined as follows.
- A vector $\left\{0, \frac{1}{q-1}, \frac{2}{q-1}, \frac{3}{q-1}, \ldots, 1\right\}$ is generated using a small even number q selected by user.
- 0 and 1 are assigned to x_j^{min}, x_j^{max} respectively.
- All other parameter values are scaled according to the vector above.
- The first parameter set is randomly selected from the values determined by the vector.
- The next parameter set is identical to the previous one, except for the value of a single parameter which is moved randomly to another possible value.

MM cont.

- Next parameter set obtained by changing another parameter etc. with a random order of parameter selection.
- By the end, the algorithm has changed the value of each parameter exactly once, and hence *m* + 1 parameter sets are generated.
- The method is also a **one-at-a-time** method together with brute force linear sensitivity analysis.
- However, here, the full uncertainty range of the input parameters can be covered, whereas in the BF method, all parameters were each changed from their nominal values.



MM continued

- Measure d_{ij} shows the effect of changing parameter x_j on model result Y_i at arbitrary values of all other parameters:

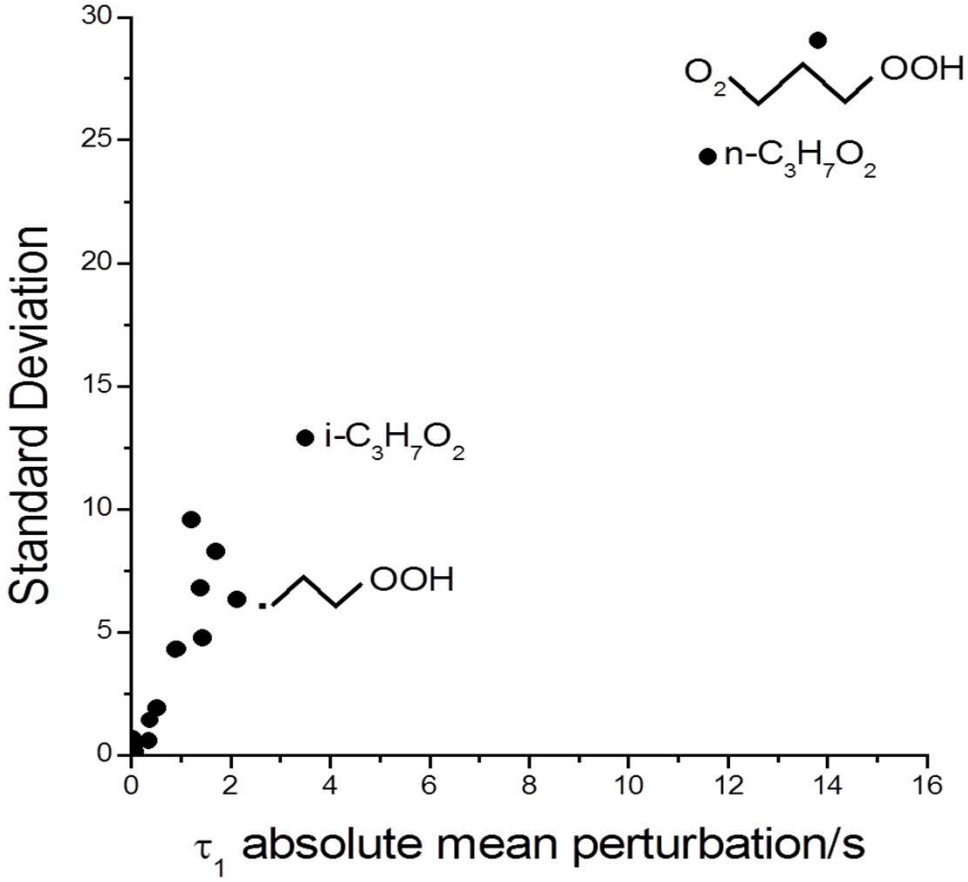
$$d_{ij} = \frac{Y_i\left(x_1^z, x_2^z, \ldots, x_j^z + \Delta, \ldots, x_m^z\right) - Y_i\left(\mathbf{x}^{z-1}\right)}{|\Delta|}$$

- In the z-th parameter set, the value of parameter x_j was changed by Δ.
- The calculation above is repeated several (r=10-20) times, always starting from randomly selected different parameter sets.
- The total computational effort required is therefore $r(m + 1)$, where r is the number of repeated parameter sets.

Analysis of results, MM

- Usually the results of the Morris method are presented graphically as a plot of $\sigma(d_{ij})$ (variance) against $\mu(d_{ij})$ (mean effect) calculated across the runs.
- Parameters with high and linear influence are in the lower right corner of the plot.
- Ones with high and nonlinear influence are in the upper right corner.
- Non-influential parameters are in the lower left corner, near the origin and are likely to be parameters that could be neglected in subsequent global sensitivity analysis.
- For parameters in the upper right corner, their influence varies according to the position of the parameter sample within the input space, implying either a strong interaction between these and other parameters in the model, or a highly nonlinear sensitivity index for individual parameters.

Examples of application of Morris screening method

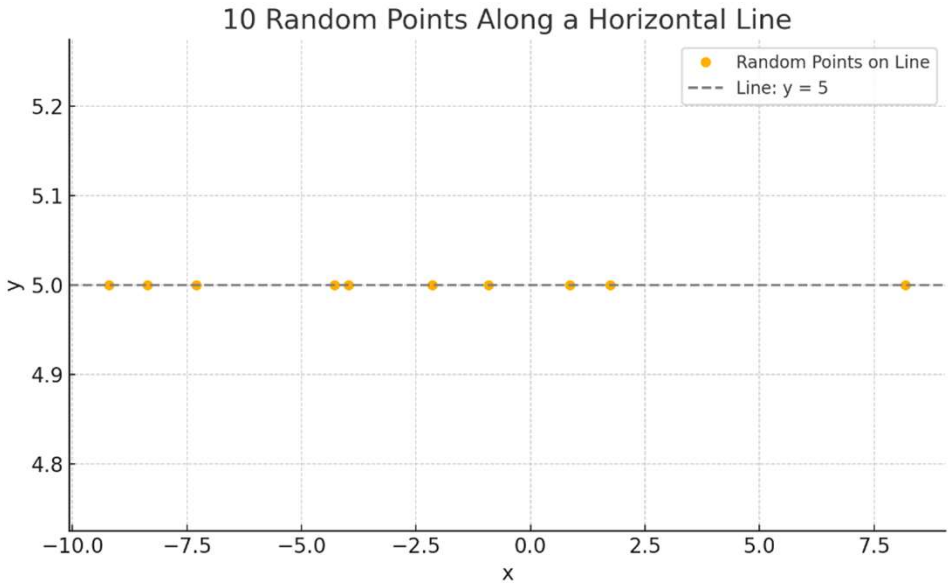


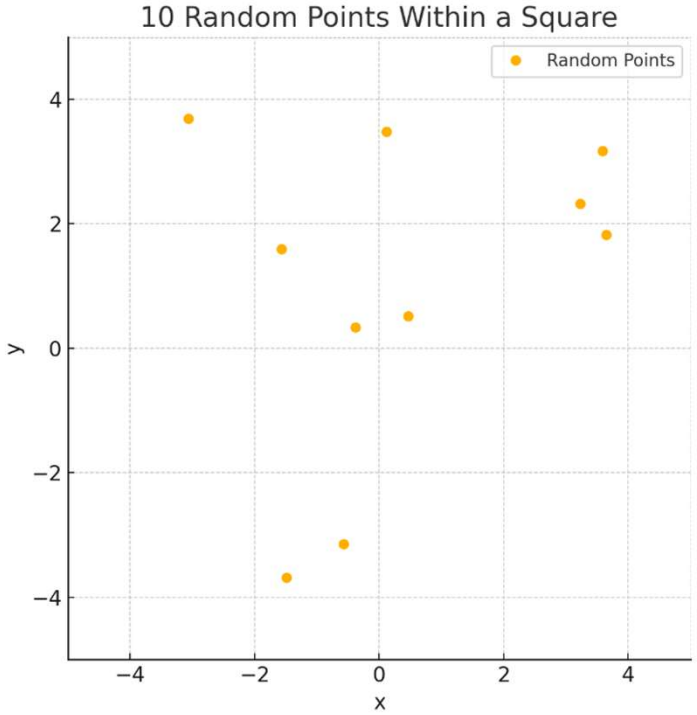
Morris analysis for species ΔH_f^o with respect to ***time to cool flame for propane oxidation****. T= 593 K, equimolar C_3H_8+O_2 at 53.4 kPa, diluted by N_2 to 101.3 kPa (Hughes et al., 2006)*

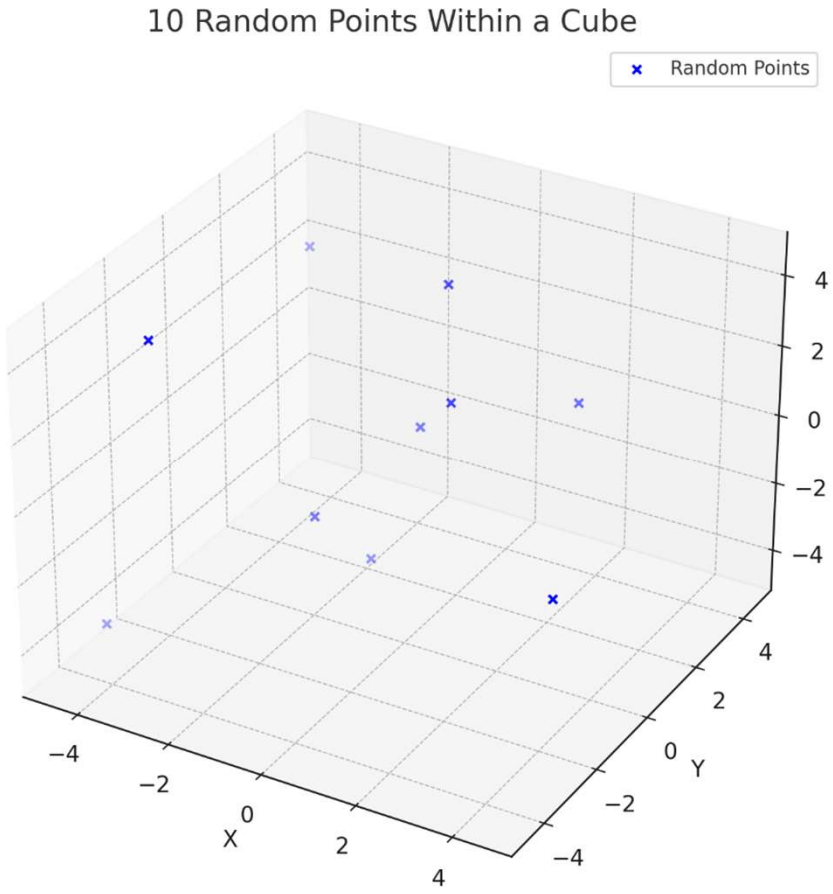
- *Note the high standard deviation of the outputs compared to the mean.*
- *Very nonlinear responses requiring large sample size to converge.*

Global uncertainty and sensitivity methods

- Once screening tests have been completed a fully global sampling based method can be applied to the parameters deemed to be of importance.
- All other parameters will be retained at their nominal values during the global sampling.
- Depending on the complexity of the model up to 50 parameters may be carried forward to a full global analysis.
- This would mean sampling from a 50 dimensional hyper space. **Hard to imagine…**
- Imagine 10 random points on a line, now place them randomly in a square,…, now place them in a cube.
- As the dimension goes up the sparsity increases massively!



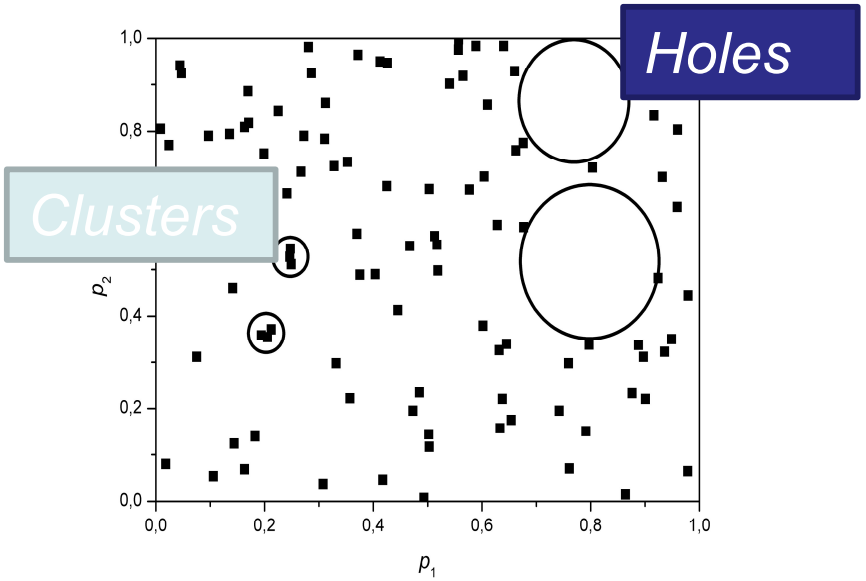


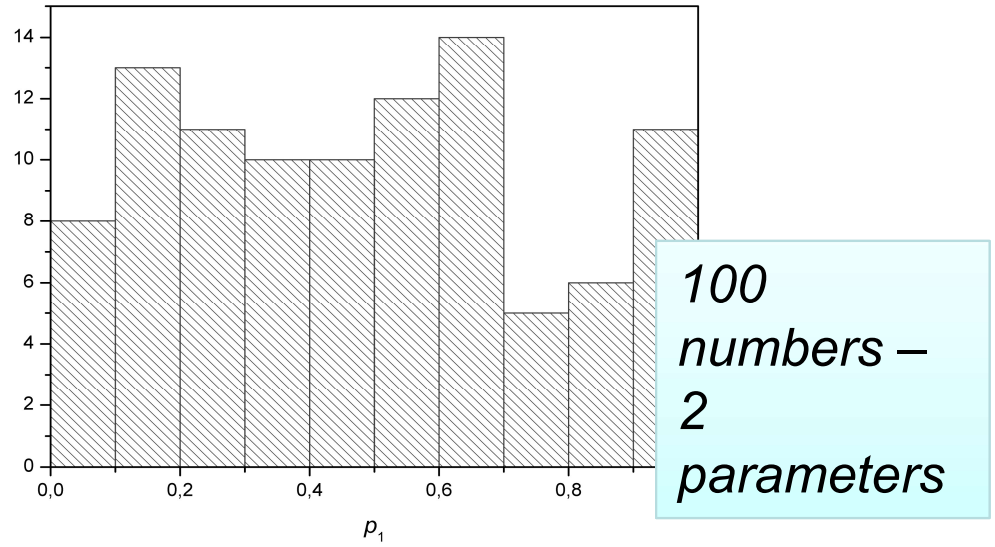


As the dimension goes up the sparsity increases massively!

Global sampling methods
Monte Carlo

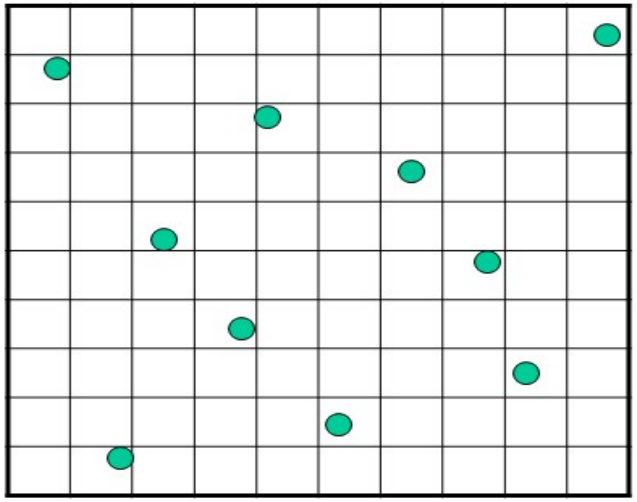
- For a global sampling method it is important to get good coverage of the input parameter space – which may be high dimensional.
- Typical random sampling methods can lead to clustering and holes.





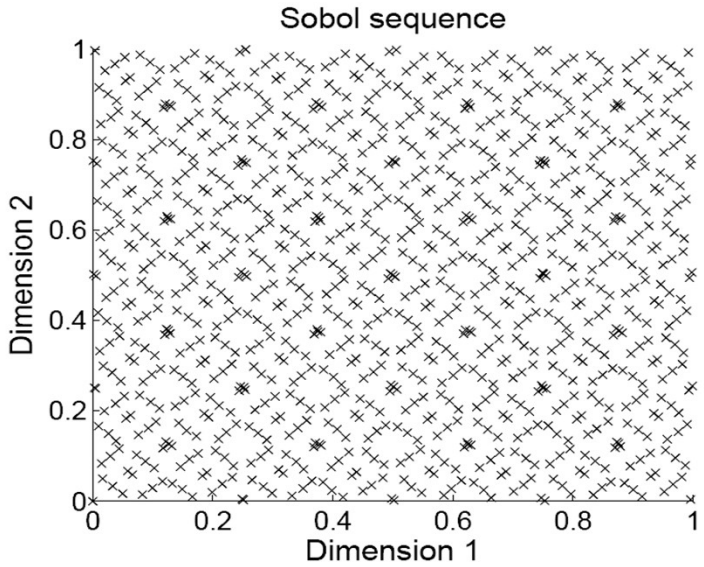
Structured sampling

Latin Hypercube



- Points generated by Latin hypercube sampling according to a uniform distribution.
- Each horizontal and vertical stratus contains a single point, while the location of the point is random in the corresponding small square.
- Quickly gets expensive for large *n*.

Low discrepancy sequences



- Successive sample points are added to positions as far away as possible from existing sample points so that clustering can be avoided.
- Scales well for large parameter systems.

Sobol sequence

Sobol sequences use a base of two to form successively finer uniform partitions of the unit interval and then reorder the coordinates in each dimension.

0.000e+00	0.000e+00	0.000e+00
5.000e-01	5.000e-01	5.000e-01
7.500e-01	2.500e-01	7.500e-01
2.500e-01	7.500e-01	2.500e-01
3.750e-01	3.750e-01	6.250e-01
8.750e-01	8.750e-01	1.250e-01
6.250e-01	1.250e-01	3.750e-01
1.250e-01	6.250e-01	8.750e-01
1.875e-01	3.125e-01	3.125e-01
6.875e-01	8.125e-01	8.125e-01
9.375e-01	6.250e-02	5.625e-01
4.375e-01	5.625e-01	6.250e-02
3.125e-01	1.875e-01	9.375e-01
8.125e-01	6.875e-01	4.375e-01
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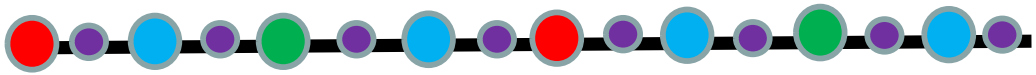
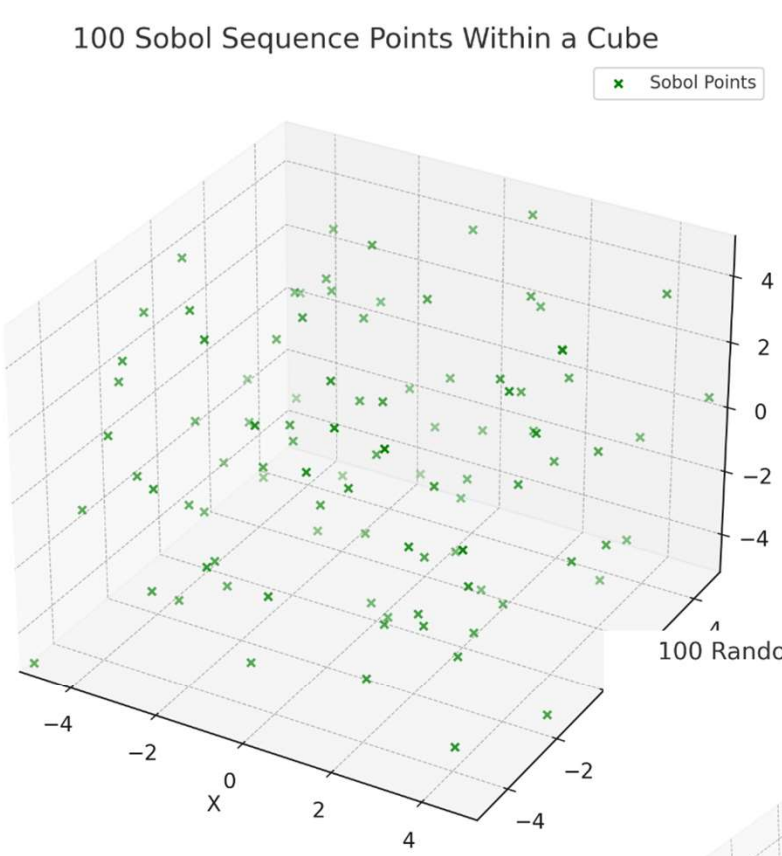
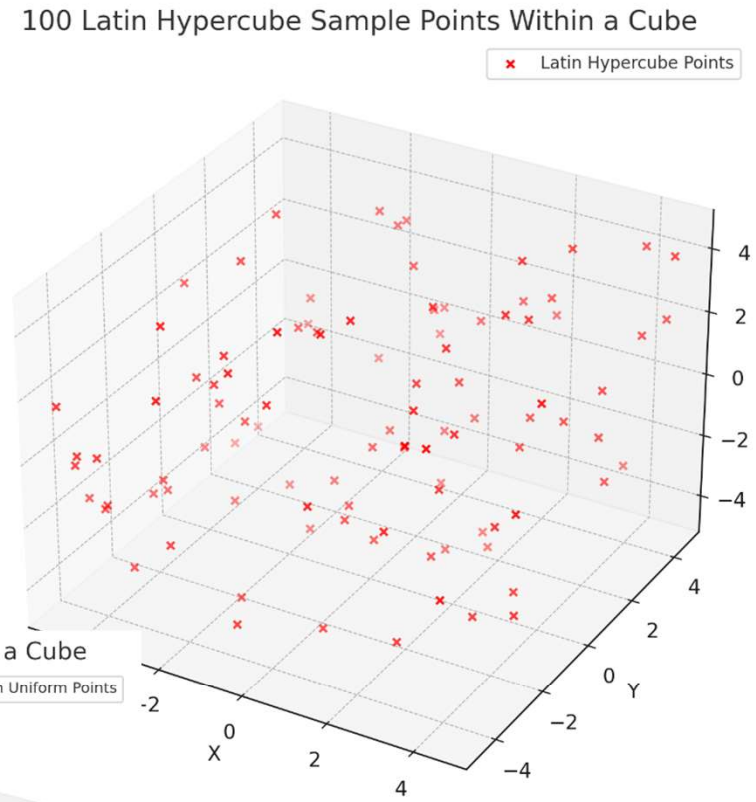
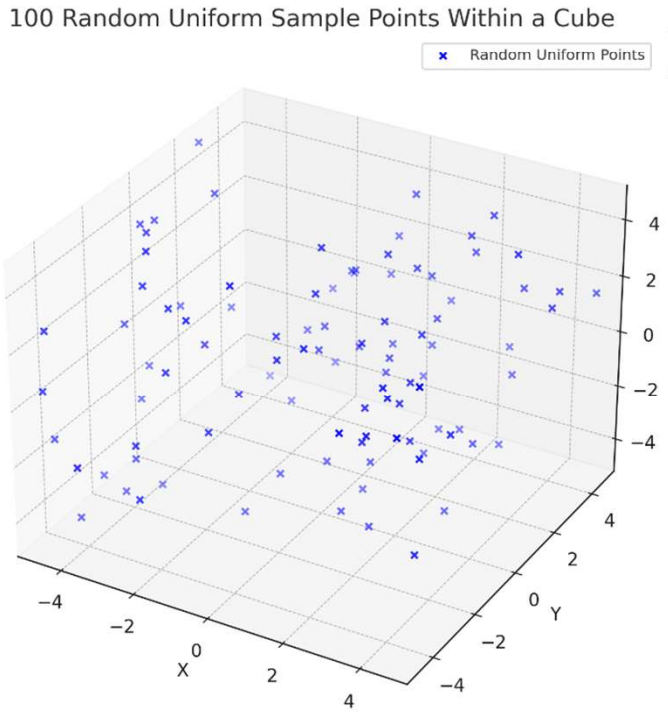


The Sobol sequence is designed to have the best convergence properties and hence can lead to savings in sampling based sensitivity and uncertainty analysis because smaller sample sizes are needed to get equivalent accuracy in the results.

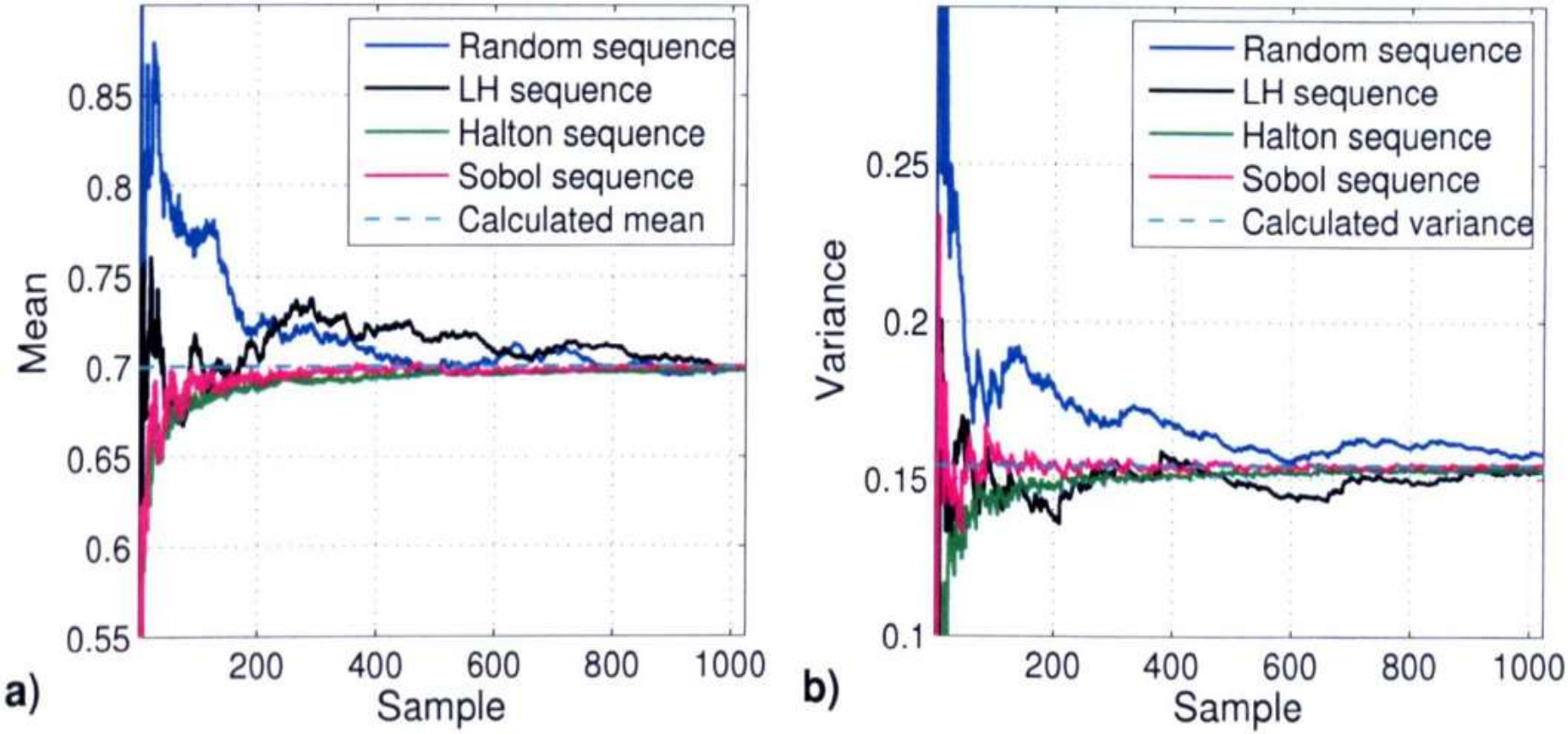
Sobol vs Random vs Latin Hypercube







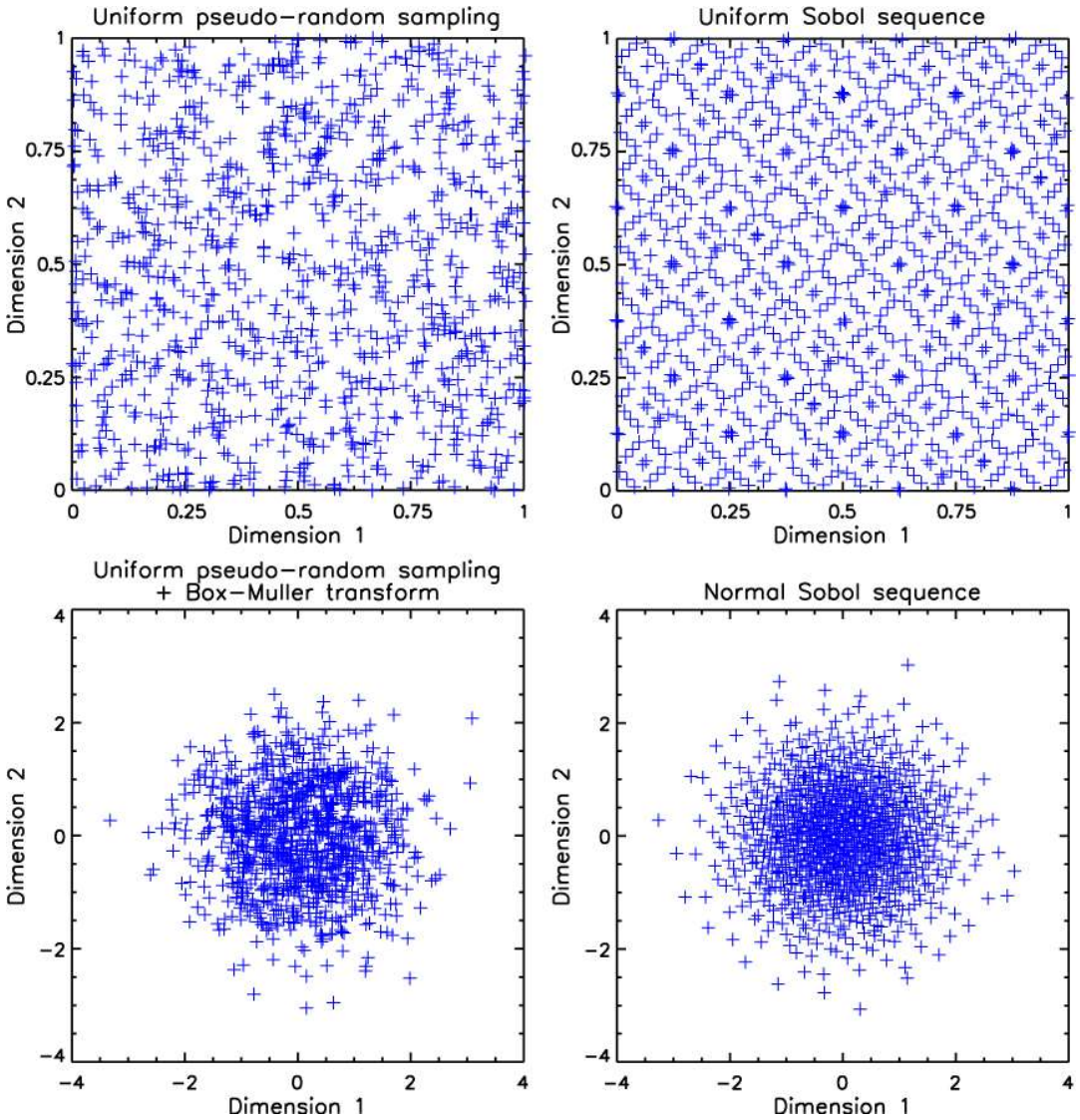
Comparison of convergence properties of different sampling strategies for a simple test model: $f(x) = x_1 + x_2^4$



Probabilistic sampling

If probabilistic information known then we may wish to sample from this distribution e.g. **a normal distribution** based on 2σ uncertainties (Hébrard et al., 2015)

- 2-parameter samples, N = 1000.
- Box-Muller transformation applied to an uniform pseudo-random sample (bottom left)
- Normal inverse cumulative function of a Sobol's quasi-random sequence sample (bottom right).

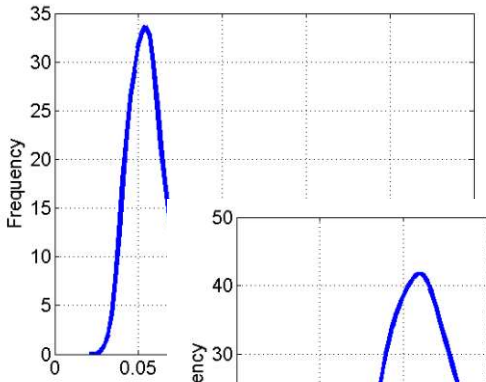


What parameters to include?

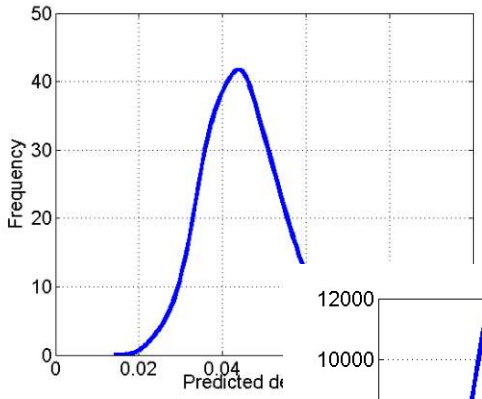
- ***In an ideal world, just for fundamental devices:***
  - All Arrhenius parameters
  - Thermodynamic parameters - used to calculate reverse reaction rates
  - Species transport data
  - Other potential model errors
    - Temperature profile
    - Heat transfer coefficients
    - Residence times
    - Loss rates to the walls of the reactor vessel
- In reality many of these are often ignored and a most common approach is to simply look at the *A*-factors for each forward reaction.
  - **Tells us something about the important reactions but does not give a full picture of uncertainties.**

Examples of outputs

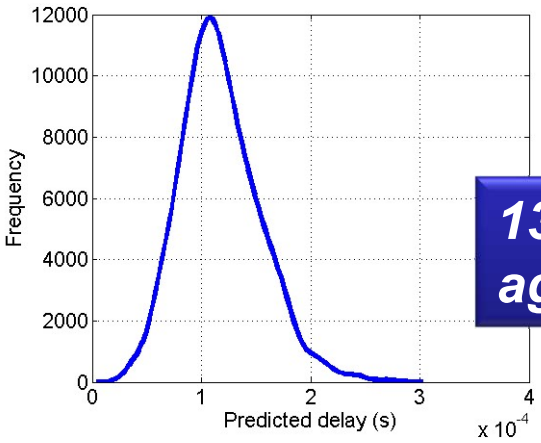
Ignition delays:
Predicted output distributions (butane model)



700 K – narrow distribution but long tail



900 K - factor of 2 uncertainty



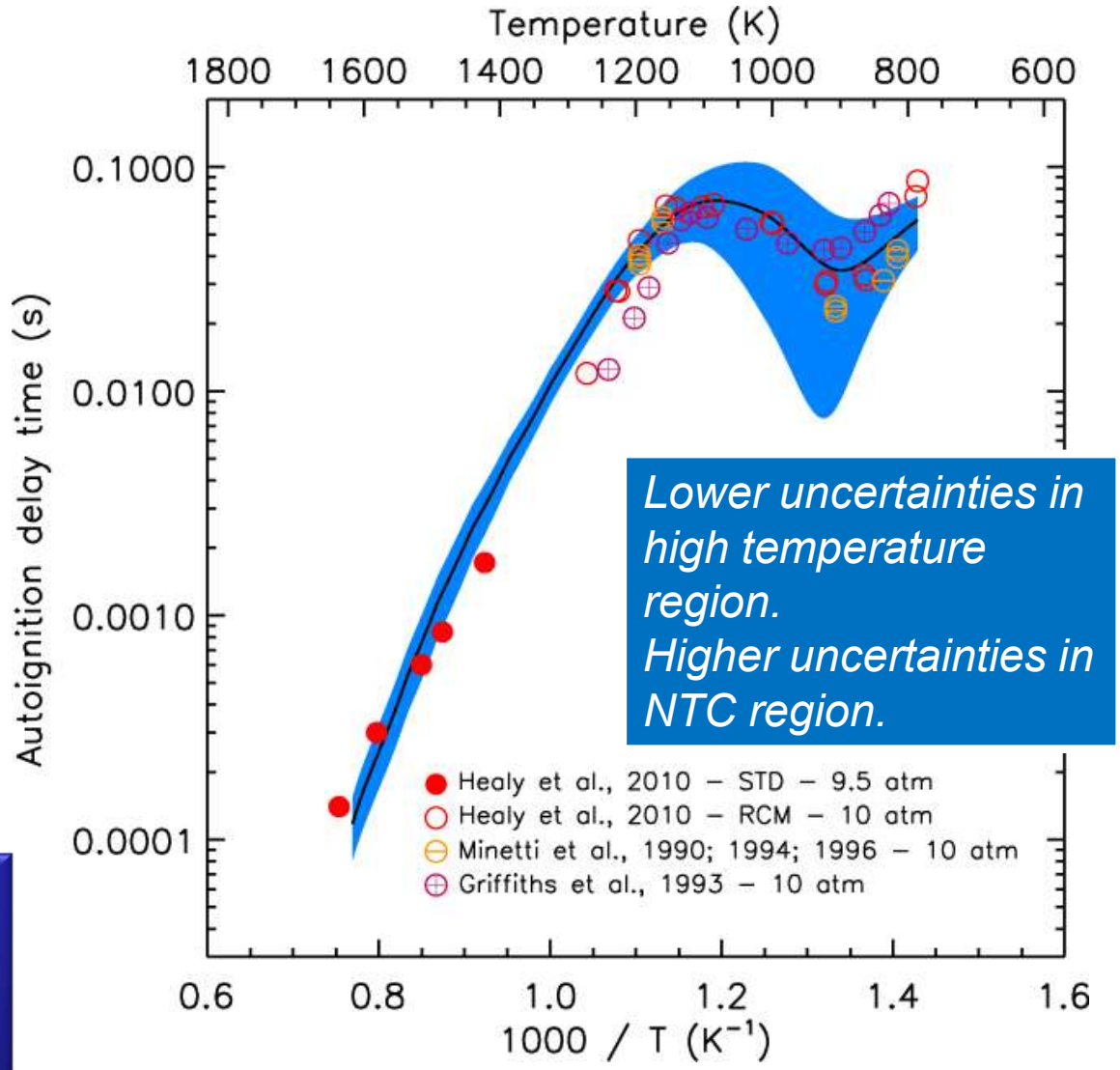
1300 K – narrows again

- *1σ used to represent output error bars.*

(Hébrard et al., 2015)

Interpreting output distributions

- Example from simulations of ignition delay time for a butane oxidation system.
- The blue shaded area represents 1σ of the outputs based on a sampled normal distribution of the input rate parameters.
- Hébrard et al. (2015)



Lower uncertainties in high temperature region.
Higher uncertainties in NTC region.

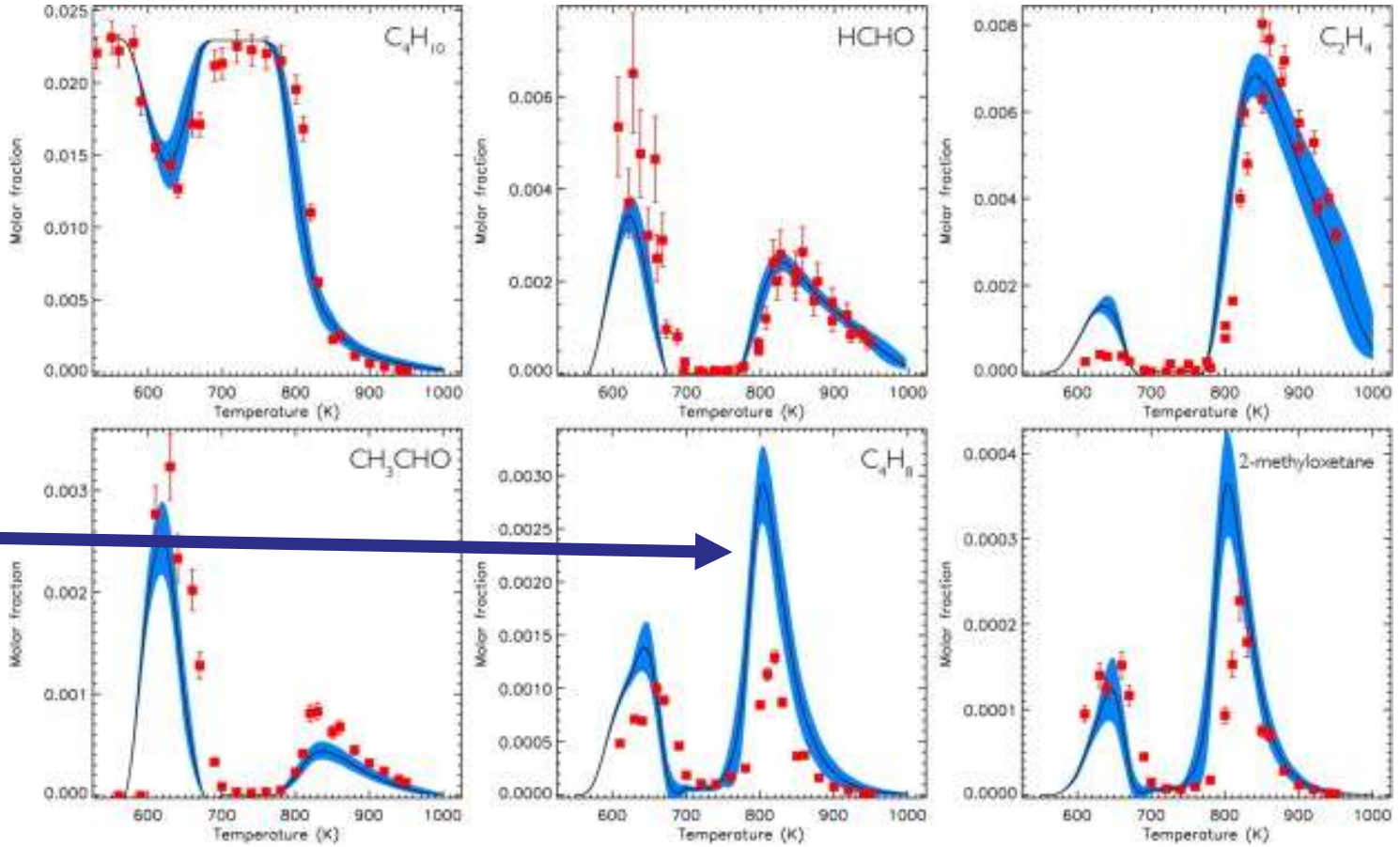
Reasonable agreement between model and shock tube and RCM data if uncertainties are taken into account.

JSR data

Some discrepancies between model and experimental data even when accounting for estimated uncertainties.

Missing reaction steps?

Other uncertainties not identified?



We learn about missing parts of the model.

(Hébrard et al., 2015)