SAND2015-6017D

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Global Sensitivity Analysis for Chemical Kinetics of Hydrocarbon Combustion

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**Sandia National Laboratories, CA* *July 29, 2015*

Abstract

For reliability purposes, numerical predictions of combustion need to account for uncertainties in model parameters. Global sensitivity analysis (GSA) informs the reduction of stochastic dimensionality for uncertainty quantification analysis. The goal of this study was to perform GSA in order to examine the relationship between dimensionality and the number of rate-determining steps in a reaction mechanism. Using the Konnov hydrogen mechanism, it is shown that with high dimensionality, only a few parameters have a significant effect on uncertainty in ignition delay times, which means that GSA can potentially save time and effort in high-dimensional problems.

Introduction

Temperature dependence of rate coefficient *k*:

A + B + … $\xrightarrow{k}$ C + D + … is given by the Arrhenius model as

$$k = AT^n \exp(-\frac{E}{RT})$$

For now, assume that k is constant, which means all uncertainty in k is due to uncertainty in A. (In general, there is uncertainty in A, n, and E.)

Dimensionality is proportional to the number of reactions:

- Konnov hydrogen mechanism – 33 dimensions, 30 reactions
- Tomlin n-butane mechanism – 1127 dimensions, 1111 reactions

Investigating 0-D Ignition:

- Quantity of interest: ignition-delay time for 0-D ignition
- Factors: initial conditions (temperature, pressure, mole fractions, mechanism); number of samples; uncertainty factors

Method

Local sensitivity analysis:

- emphasizes the local impact (at a point) of the factors on the model
- differential analysis falls under this category
- not useful in this case because the model is nonlinear over a range of parameters

Global sensitivity analysis:

- emphasizes the contribution of each factor to the overall uncertainty in the output
- sampling approach
- Monte Carlo analysis, method of Sobol', and Fourier amplitude sensitivity test (FAST) are some examples

Use TChem (open source tool to analyze reaction kinetic models) and CVODE (tool to numerically integrate initial value problems) to simulate ignition, obtain *A* coefficients and ignition-delay times

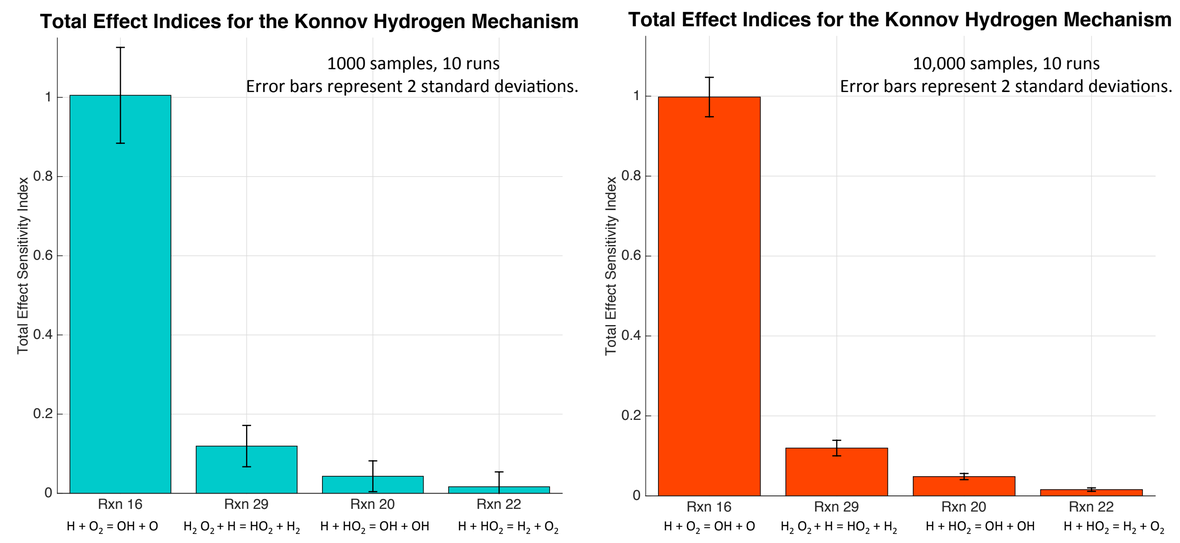
Calculate total effect indices, derived from method of Sobol':

- total order sensitivity indices, or total effect indices, measure an input variable's contribution to the variance of the output
- here, the total effect index, S_j^T, for the jth factor: $S_j^T = \frac{E_{x_{\sim i}}(V(y \mid x_{\sim j}))}{V(y)}$

  where x = (x1,…,xn), y = f(x)
- estimate using Monte Carlo sampling: $S_i^T = \frac{1}{2V}\left(\frac{1}{N}\sum_{j=1}^{N}\left(f(x^j) - f(x^j_{-i} \cup x_i''^j)\right)^2\right)$

Results

Mechanism	Konnov	Tomlin
Fuel	hydrogen, H_2	n-butane, n-C_4H_{10}
Reactions	30	1111
Dimensions	33	1127
Samples	1000; 10,000	500
Initial Conditions	T = 1000K, P = 1 atm, stoich. mix	T = 1000K, P = 1 atm, stoich. mix
Number of species	12	177
Computational expense	**0.020 sec** to run one ignition	**9.70 sec** to run one ignition
Important reactions	Rxn 16: $H + O_2 = OH + O$ (Chain branching) Rxn 29: $H_2O_2 + H = HO_2 + H_2$ (Chain termination)	In progress; results to be obtained.



The reactions not shown have total effect indices close to 0.

Conclusions and Future Work

In this study, we attempt to characterize the relationship between dimensionality and the number of important reactions in a combustion reaction mechanism. For the Konnov hydrogen mechanism, there were 4 significant parameters out of 33, which shows that with high dimensionality, few parameters have a large impact on the uncertainty in ignition delay times. In the Konnov mechanism, reaction 16 is clearly the most important, followed by reaction 29. Thus for high-dimensional problems, one could potentially save a lot of computational effort and time by only considering a few parameters for uncertainty analysis. The authors are currently analyzing Tomlin's n-butane mechanism in a similar manner.

In the future, we would like to check the validity of the above conclusions in a larger dimensional context, i.e. for more complex hydrocarbon fuels. For complex hydrocarbons like n-butane, there are on the order of thousands of rate parameters, and therefore global sensitivity analysis would be an essential step in reducing the dimensionality for uncertainty quantification purposes. Sampling such systems becomes computationally expensive, and thus a study examining the effect of ensemble size on the accuracy of the resulting sensitivity indices is imperative. Some other possible studies are to perform more extensive GSA on a subset of the parameters deemed important from the initial analysis, examine the effect of different initial conditions on sensitivity indices, and study different quantities of interest such as species' concentration profiles and flame speed.

U.S. DEPARTMENT OF ENERGY
National Nuclear Security Administration

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SAND No. 2011-XXXXP