Chemical Kinetic and Transport Model Uncertainty Analyses and Mechanism Reduction for Propulsion Systems

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Rolls Royce

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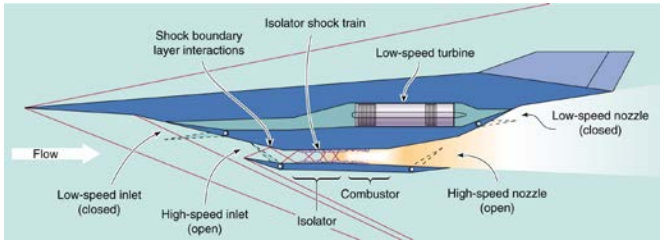
Objectives

- **Reduced kinetic models for hypersonic reacting flow simulations**
  - endothermic fuels containing H2, CH4, C2H2, C2H4, C3H6, …
  - predict critical induction/flame holding limits
  - goal: turbulent mixing-chemistry interactions



- **Reduced kinetic models for soot formation/oxidation under high-pressure conditions (gas turbine engines)**
  - non-premixed and premixed high-pressure combustion
  - goal: soot data and uncertainties from GC/MS, SMPS, TEM, LII
  - soot modeling? (PrIME?)
- **Main focus of this presentation: <u>Uncertainty analyses</u>**
  - detailed kinetic model parameters (A-factors)
  - transport coefficients
  - Metamodels (PrIME?)

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Model Reduction

Skeletal and Reduced Reaction Models:

- **PCAS** based method (Vajda 1985, Brown 1997) applied for **ethylene-air:**

$$Q(\mathbf{P}) = \sum_{j=1}^{q}\sum_{i=1}^{m}\left[\frac{f_i(x_j,\mathbf{P}) - f_i(x_j,\mathbf{P^0})}{f_i(x_j,\mathbf{P^0})}\right]^2$$

$$Q(\mathbf{P}) \approx q(\mathbf{P}) = (\Delta\mathbf{P})^T\,\mathbf{S}^T\mathbf{S}\,(\Delta\mathbf{P})$$

$$q(\mathbf{P}) = \sum_{k=1}^{r}\lambda_k\,(\Delta\Psi_k)^2$$

  - *Esposito and Chelliah. Combustion and Flame 158 (2011)*
  - combined sensitivities of ignition, propagation, and extinction
  - **37-38 species combined skeletal model** from USC Mech II* (2009)
- **QSSA** for reduced reaction model for ethylene-air:
  - **20-step reduced reaction model**
  - *Zambon and Chelliah, Combust. and Flame (2007)*
- **NIST Chemical Kinetic Model Database, PrIME?**

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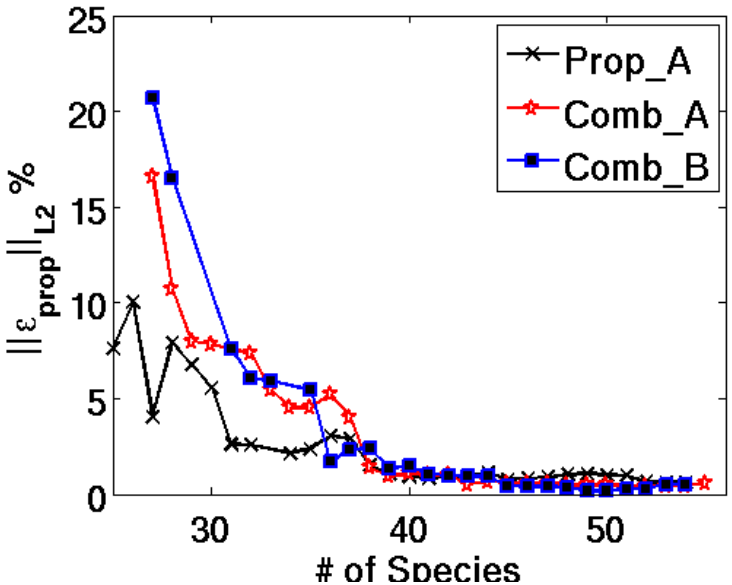
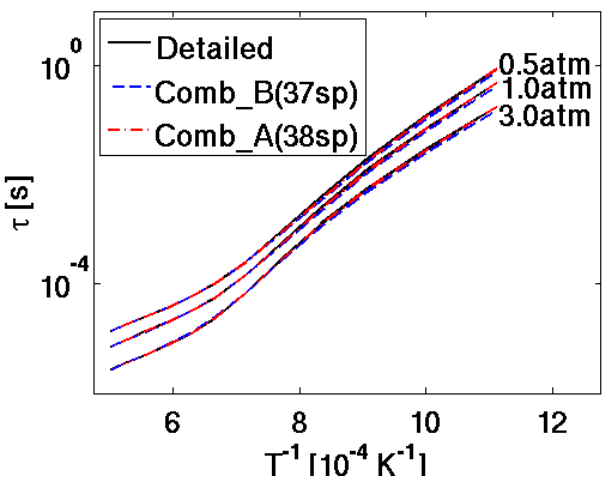
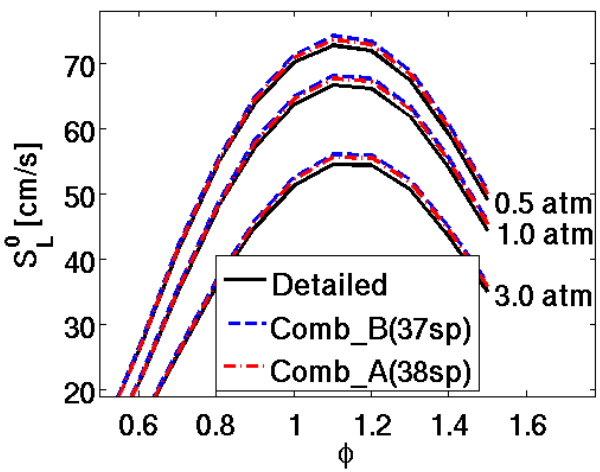
Model Reduction

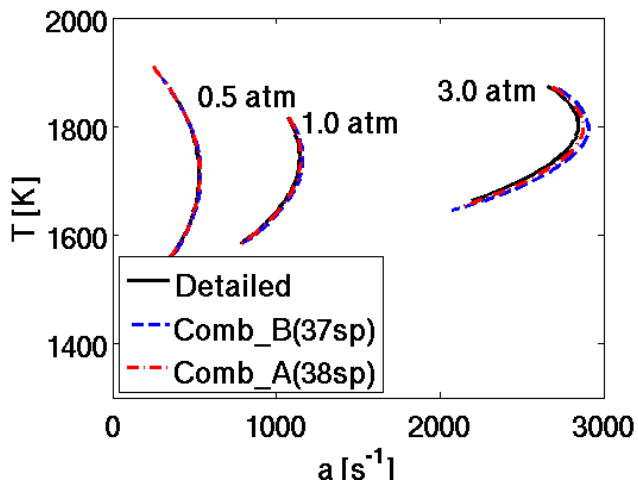
- **L2 norm-error from PCAS:**
  - **Integrated over range of conditions**

$$||\epsilon_{ign}||_{L_2} = \left[\int_{\phi_a}^{\phi_b} \int_{T_a}^{T_b} \left(\frac{\tau_{ign,skel} - \tau_{ign,det}}{\tau_{ign,det}} \right)^2 \mathrm{d}T\mathrm{d}\phi \right]^{1/2}$$









- Esposito and Chelliah, Combustion and Flame158 (2011)

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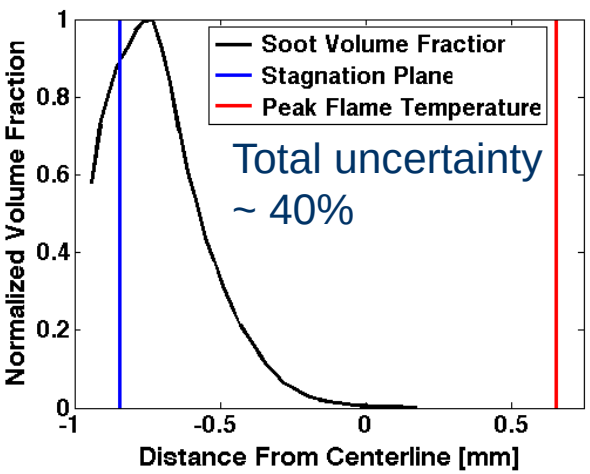
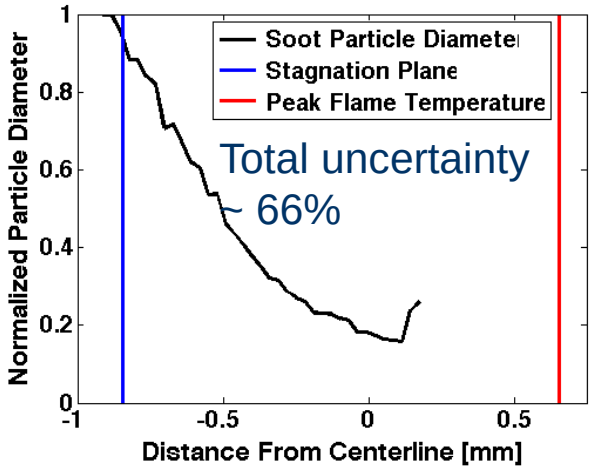
Some Experimental Data

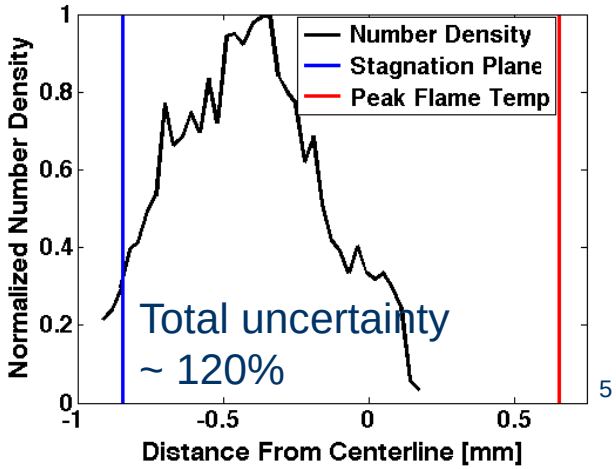
- Non-premixed flame extinction limits with 2σ uncertainties (Sarnacki et al., CNF 2012, Esposito et al. CTM 2012)
- Model predictions using USC Mech II for HC's and JetSurf 2.0 for H2

Fuel-air system	Local Extinction Strain Rate (s^{-1})	Model Predict. (s^{-1}) with finite U's
Methane	380±21	456-470
Ethylene	1284±48	1223-1232
Propylene	617±34	606-624
n-Butane	499±38	544-550
diluted Hydrogen	2542 ± 112	2644

- Soot characteristics in non-premixed flames, p=1 to 8 atms







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Uncertainty analyses

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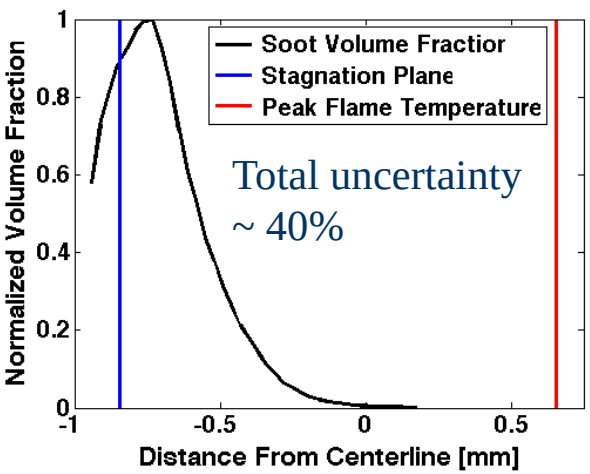
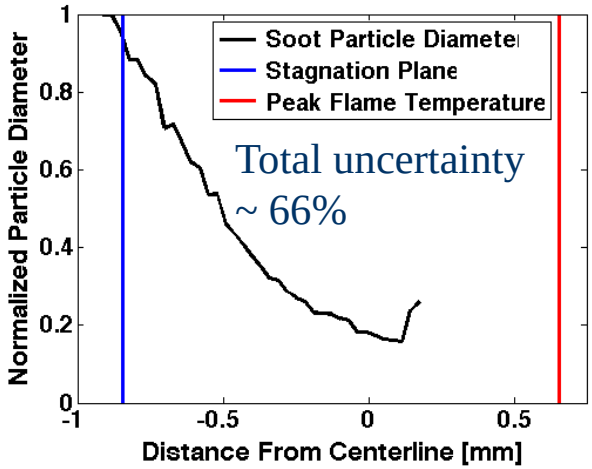
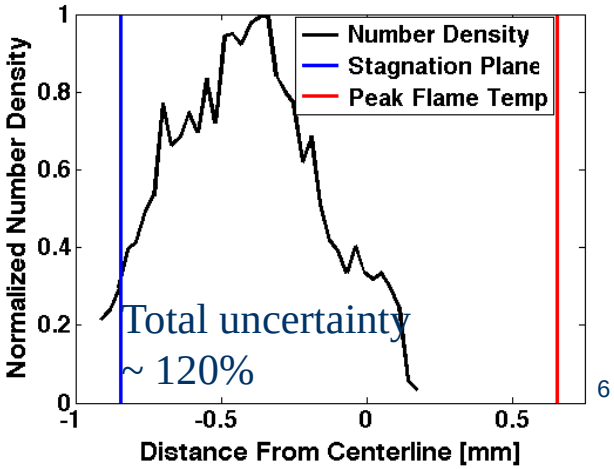


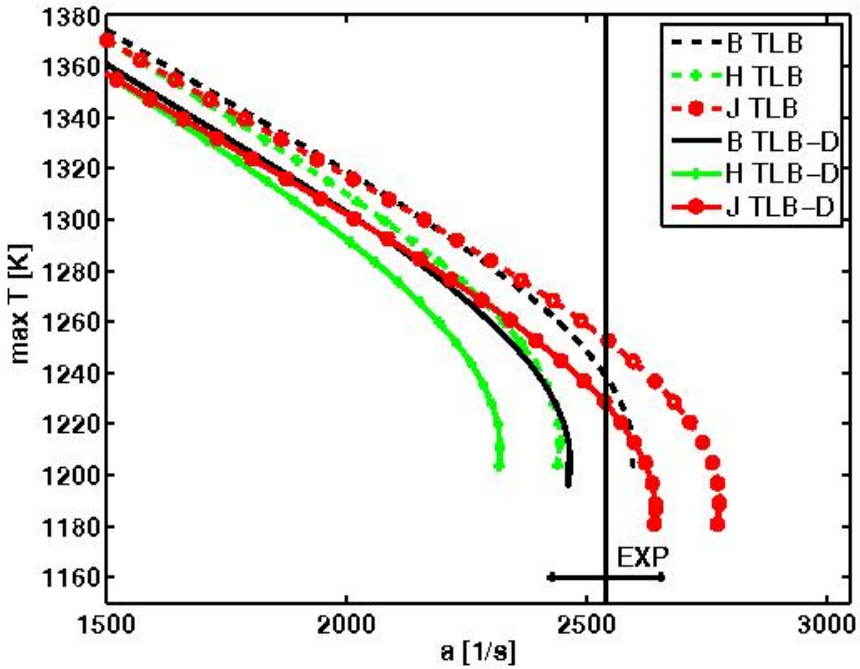




Variability of Tuned Kinetic Models

- Detailed kinetic models continue to evolve/tuned/optimized, <u>including H2</u>
- Uncertainty reduction of kinetic models has received considerable attention
- Uncertainty of simulations is due to two key parameters in flames used for model optimizations:
  - Chemical kinetic parameters
  - Transport coefficients (eg. binary diffusion coefficients)
- Find the relative importance of the uncertainty of both CK and diffusion parameters
  - Search for conditions that minimize diffusion coefficient influence on target flame property predictions

H2/N2 (25%/75%)-Air



Uncertainty of extinction limit using three latest H2/O2/N2 chemical kinetic models and diffusion coefficients (G Esposito, BG Sarnacki, HK Chelliah, CTM 2012)

(B=Burk et al. 2012, H=Hong et al. 2011, J=JetSurf 2012, D=Middha & Wang 2002)

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Chemical Kinetic and Transport Uncertainty factors

- CK Uncertainties:
  - Complied by USC, NIST

Reactions (H2)	Uf
H+O2=O+OH	1.2
HO2+H=2OH	2.0
OH+H2=H+H2O	1.3
O+H2=H+OH	1.3
OH+HO2=H2O+O2	2.0

Reactions (C2H4)	Uf
C2H3+H=C2H2+H2	3.0
C2H3+H=H2CC+H2	5.0
HCO+M=CO+H+M	4.0
HCO+H=CO+H2	2.0
C2H3+O2=CH2CHO+O	3.0

- Diff. Coeff. Uncertainties:
  - From various literature (Brown, PCES 2010)

Binary Diff Coeff (H2)	Uf (%)
D(H2,N2)	3%
D(H,N2)	8%
D(O2,N2)	12%
D(H2O,N2)	12%
D(H2,H2O)	12%
D(H2,He)	5%

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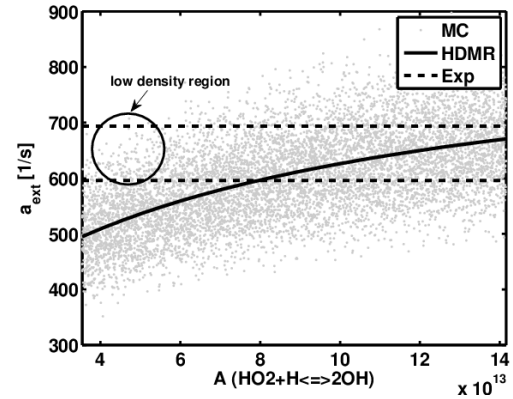
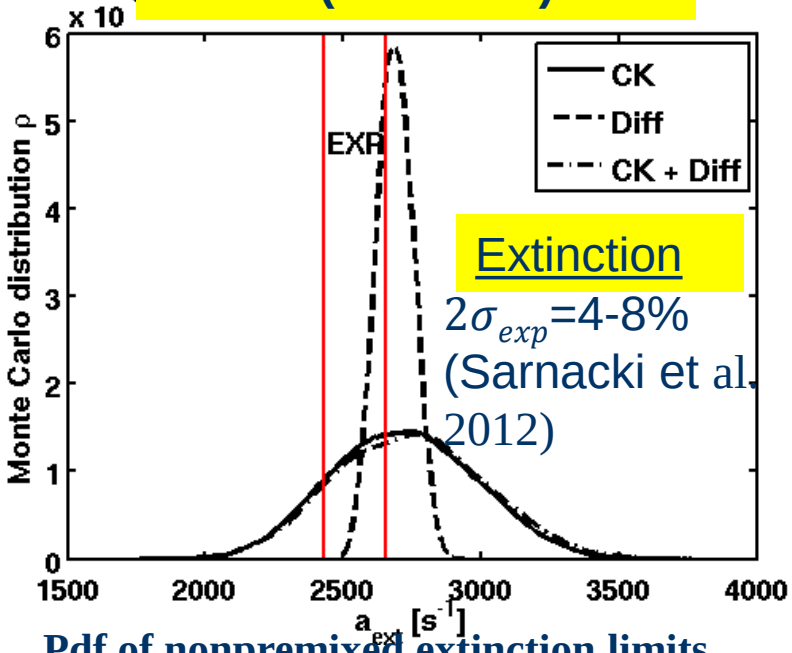
Uncertainty Propagation and HDMR

- 2^{Np} Monte-Carlo simulations of N_p parameters bounded by experimentally determined uncertainty factors
- ~15,000 simulations for each phenomenon: ignition, flame propagation, and extinction
- Embarrassingly parallel simulations with distributed computing
- Use untuned CK parameters (JetSurf 2.0 H2 sub-mechanism)
- HDMR decomposition (Tomlin GUI):
  - Chemical parameters only
  - Binary diffusion coefficients only
  - Simultaneous analysis of chemical and diffusion parameters



H2/N2 (25%/75%) - Air



Extinction

$2\sigma_{exp}$=4-8% (Sarnacki et al. 2012)

Pdf of nonpremixed extinction limits
(Esposito, et al., CTM 2012)

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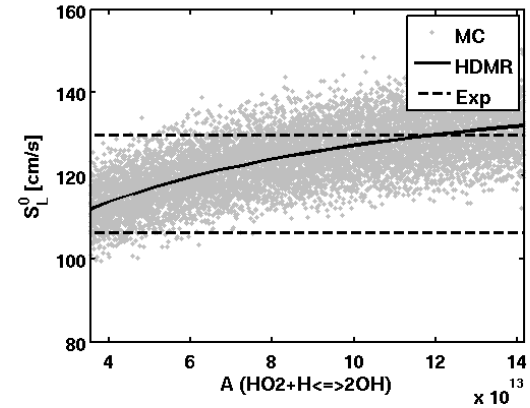
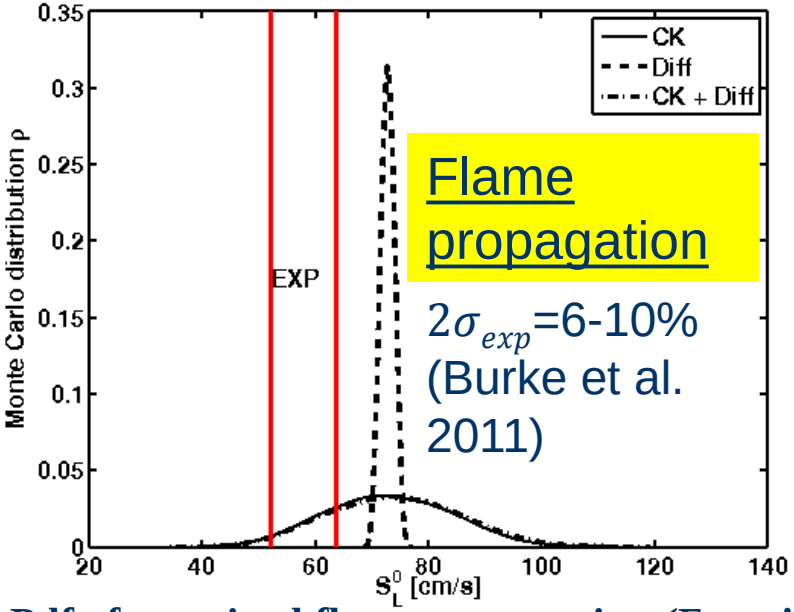


H2/O2/He (0.14/0.07/0.79)



Flame propagation

$2\sigma_{exp}$=6-10% (Burke et al. 2011)

Pdf of premixed flame propagation *(Esposito, et al. in press, CTM 2012)*

Comparison of Propagated Uncertainties on Global Property Predictions

- Uncertainty of CK much larger than experimental ($2\sigma_{exp}$)
- Uncertainty of Diff smaller than $2\sigma_{exp}$ for laminar flames, but comparable to $2\sigma_{exp}$ in non-premixed flames
- High-pressure data may be ideal for optimization of some reactions that are important at low pressure!
  - Ex: HO2+H=2OH

Laminar Flame Speed (1 atm)						
ϕ	1.0		1.74		0.85	
Model	Diff	CK	Diff	CK	Diff	CK
ϵ	0.03	0.13	0.03	0.12	0.03	0.15
Laminar Flame Speed (15 atm)						
ϕ	1.0		1.74		0.85	
Model	Diff	CK	Diff	CK	Diff	CK
ϵ	0.03	0.31	0.02	0.33	0.05	0.41
Extinction strain rate (1 atm)						
Case	A		B		C	
Model	Diff	CK	Diff	CK	Diff	CK
ϵ	0.08	0.28	0.06	0.22	0.08	0.19

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Sensitivity of Reactions (S_i) from Uncertainty Analysis

Laminar Flame Speed (1 atm)				
Reactions	U_f	S_i		
		ϕ= 1.0	ϕ=1.74	ϕ=0.85
HO2+H=2OH	2.0	0.471	0.447	0.488
OH+H2=H+H2O	1.3	0.241	0.086	0.255
O+H2=H+OH	1.3	0.084	0.033	0.084
H+O2=O+OH	1.2	0.065	0.247	0.043

Laminar Flame Speed (15 atm)				
Reactions	U_f	S_i		
		ϕ= 1.0	ϕ=1.74	ϕ=0.85
HO2+H=2OH	2.0	0.502	0.539	0.497
H+O2=O+OH	1.2	0.140	0.218	0.131
OH+H2=H+H2O	1.3	0.170	0.031	0.105
H+OH+M=H2O+M	2.0	0.087	0.072	0.036

- Global Sensitivity Indices S_i (portion of the uncertainty of the target flame property due to single reactions, from variances V_i/V)
- HO2+H=2OH most uncertain (U_f=2.0) and highest contribution ~ 50%
- CK contribution significantly higher at high *p* (see ϵ).
- Second most important reactions:
  - OH+H2=H+H2O at 1 atm
  - H+O2=O+OH at 15 atm
  - Decrease of their U_f may make these flames even more suitable to improve the evaluation of HO2+H=2OH

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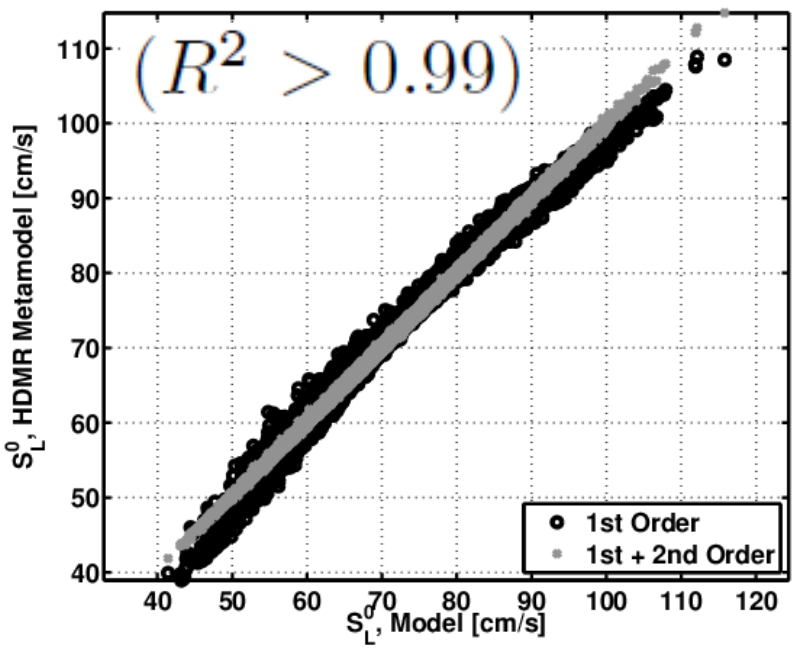
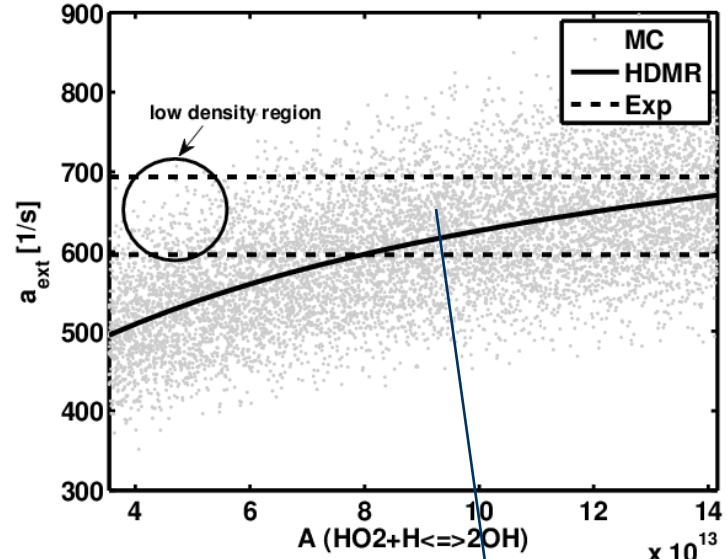
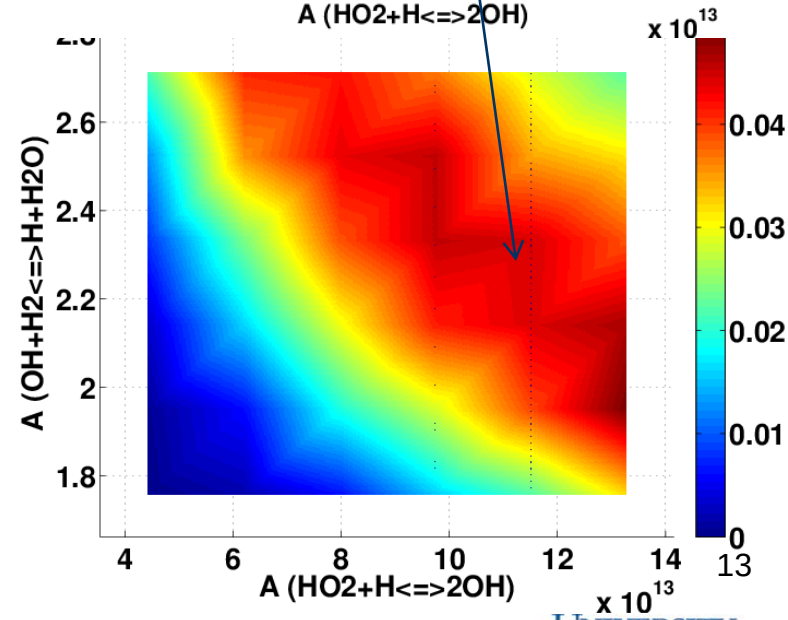
Other Benefits of Uncertainty Based MC Simulations

- Uncertainty back-propagation (can also include binary diffusion coefficient uncertainties)
- Can identify feasible regions of multidimensional parameter space
- More accurate meta-models for optimizations?

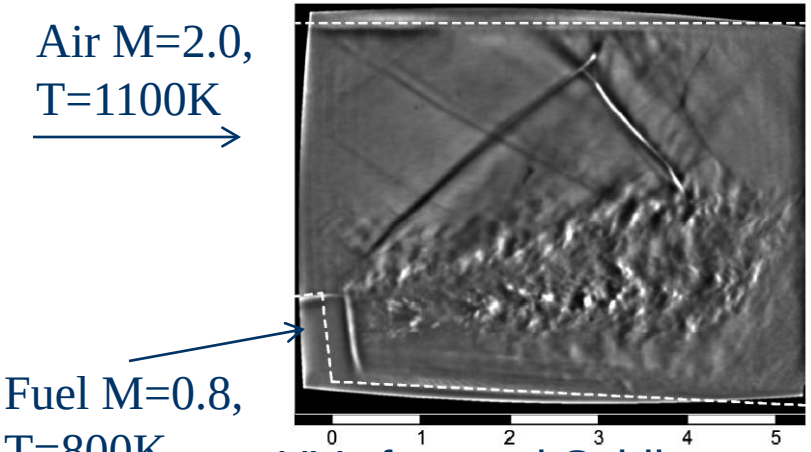




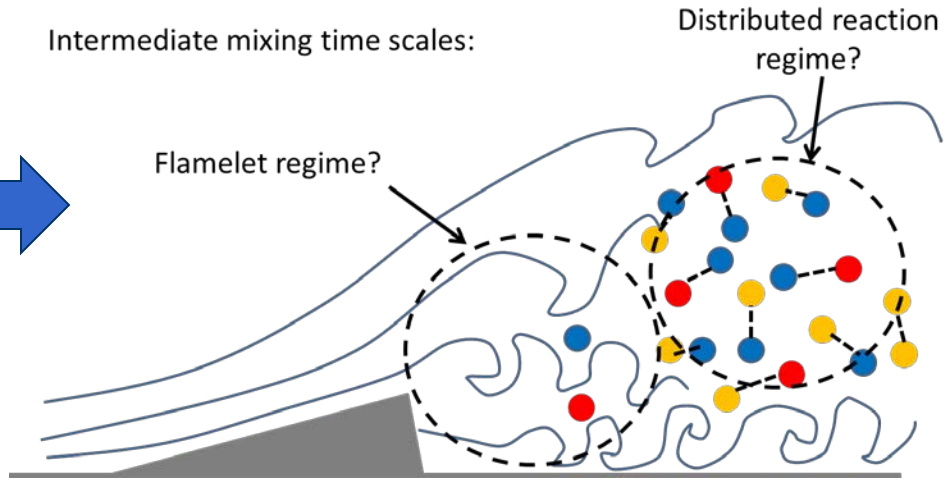


Turbulent Mixing Effects on Reduction?

- What is the relevant combustion mode or regime for high-speed reacting flows?



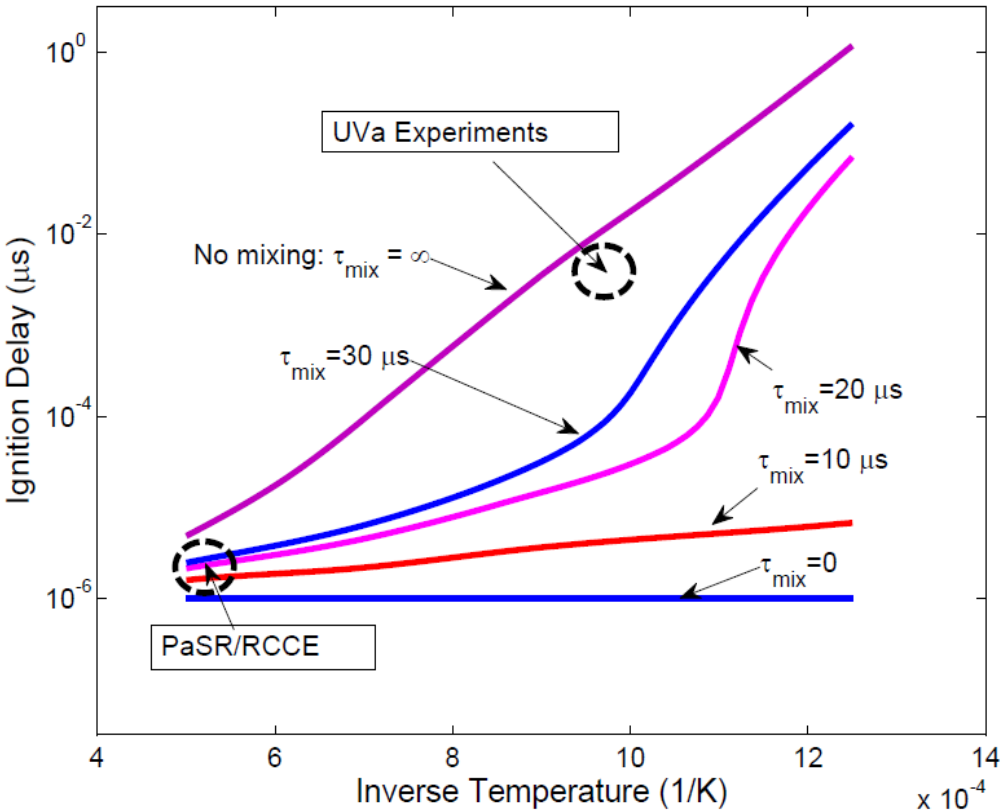
UVa focused Schlieren

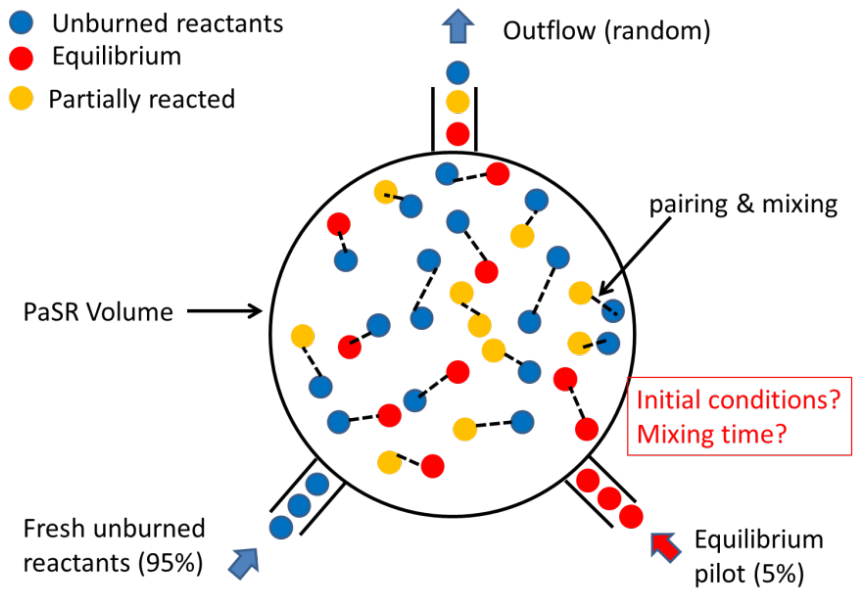


- Preliminary SPIV indicates $\tau_{mix} \approx$ 60-100μs and $\tau_{res} \approx$ 350μs
  - Hybrid RANS-LES also predict $\tau_{mix} \approx$ 100 μs
- PaSR to test model reduction (Correa, Chen, Pope in 1990's)

Model Reduction with Turbulent Mixing

- In PaSR, reactant streams consisting of fresh stream of premixed stoichiometric ethylene-air (95%) and pilot stream of equilibrium mixture (5%)

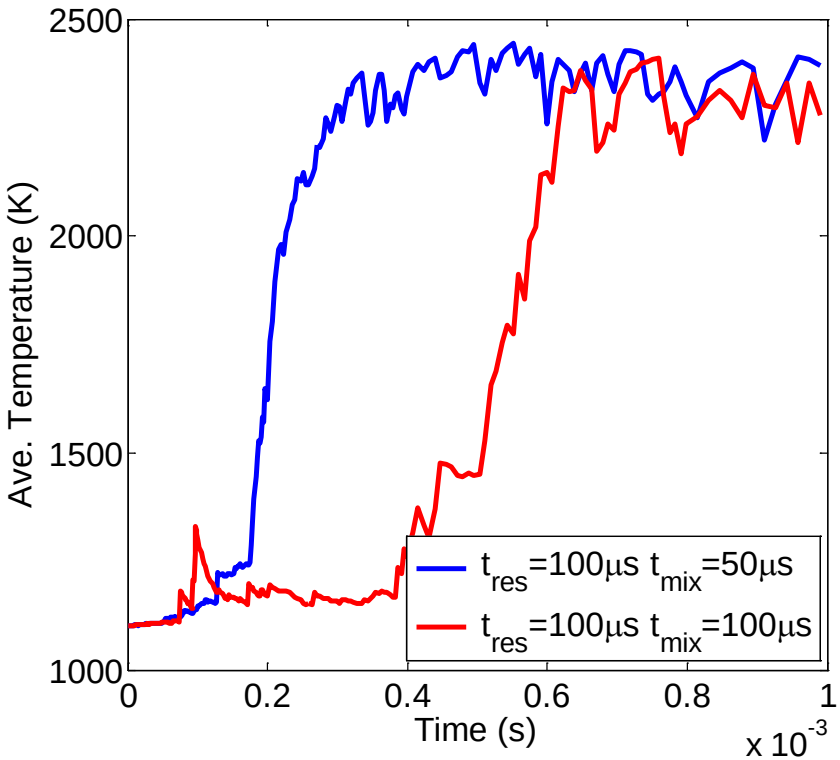




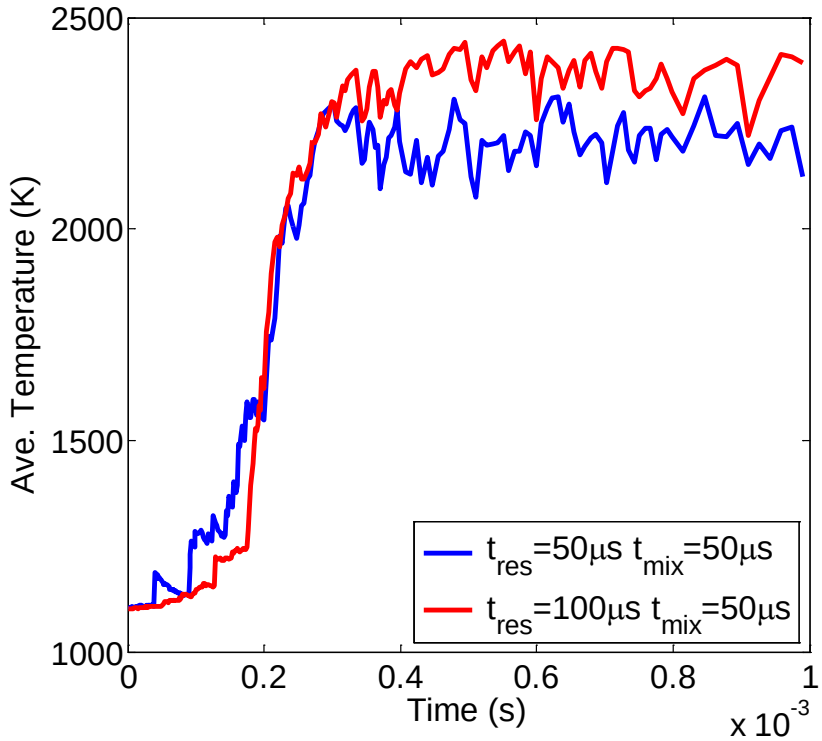
Comparison of previous PaSR analysis regime vs. actual experimental conditions (AIAA Jan. 2012)

Model Reduction with Turbulent Mixing

- Fresh mixture of stoichiometric ethylene-air (95%) mixing with a pilot stream of equilibrium mixture (5%)



Effect of mixing time variability on evolution of average temperature



Effect of residence time variability on evolution of average temperature

- As Da_{mix} and Da_{res} approach unity, significant sensitivity of PaSR results to kinetics, consistent with Chen 1997

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Model Reduction with Turbulent Mixing

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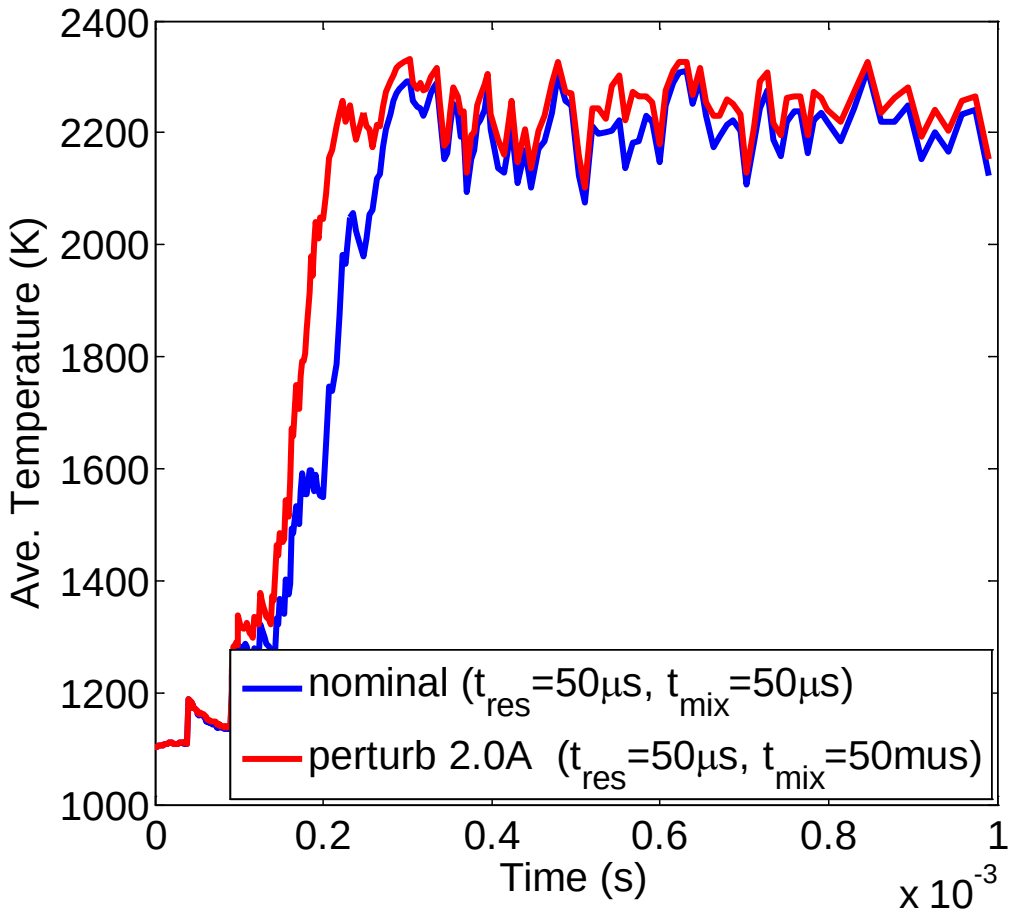

- Sensitivity of PaSR results to perturbation of of H+O2=OH+O reaction
- Need to construct sensitivity matrix in order to apply PCAS
- Test reduction under realistic turbulent reacting flow conditions using PCAS, QSSA, RCCE, …

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Summary

- Uncertainty analysis of both kinetics and transport parameters, based on Morris Method for screening active parameters, followed by Monte Carlo simulations is computationally feasible using massively distributed computing
- HDMR analyses successfully applied to hydrogen-air system and ethylene-air system to extract:
  - global sensitivities
  - feasible parameters, and
  - meta models
- **<u>FUTURE WORK:</u>**
- Extend the analysis tools to C3 and higher fuels
- For turbulent reacting flows, use PaSR and its variants to test model reduction via PCAS, QSSA, RCCE, etc.
  - ➢ Need to know realistic turbulent mixing times, residence times, etc.

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