Multiscale Kinetic Knowledge Propagation - Combustion Chemistry of Small Hydrocarbons

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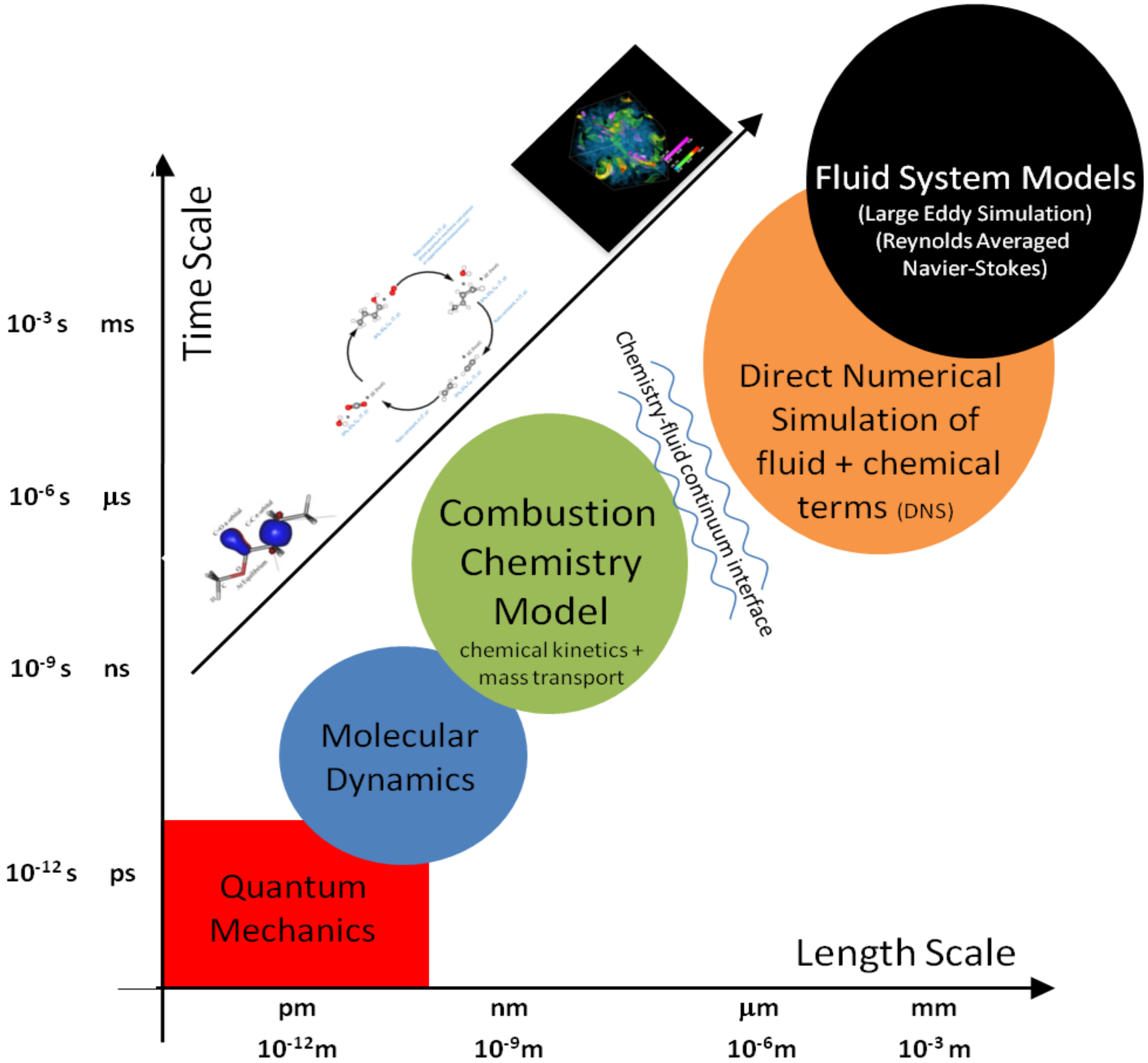
Combustion in the Nation's Energy Future

- Optimal utilization of alternative synthetic and biofuels requires advances in combustion science and engineering.

- Advances in combustion technology can substantially stretch the petroleum supply and reduce GHG emissions.

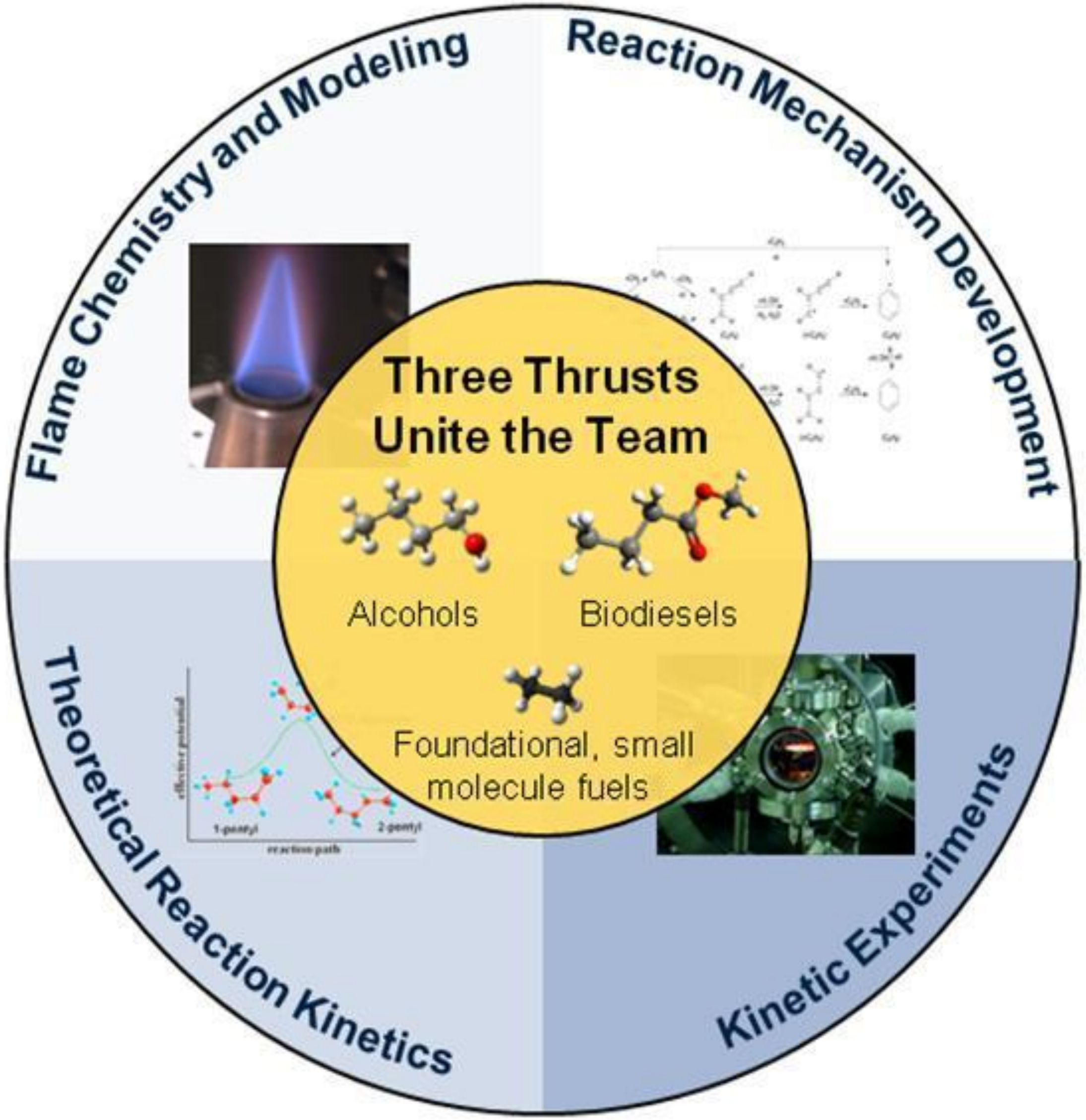
Combustion – a Multiscale, Multiphenomenon Challenge



Multi-Scale Tools

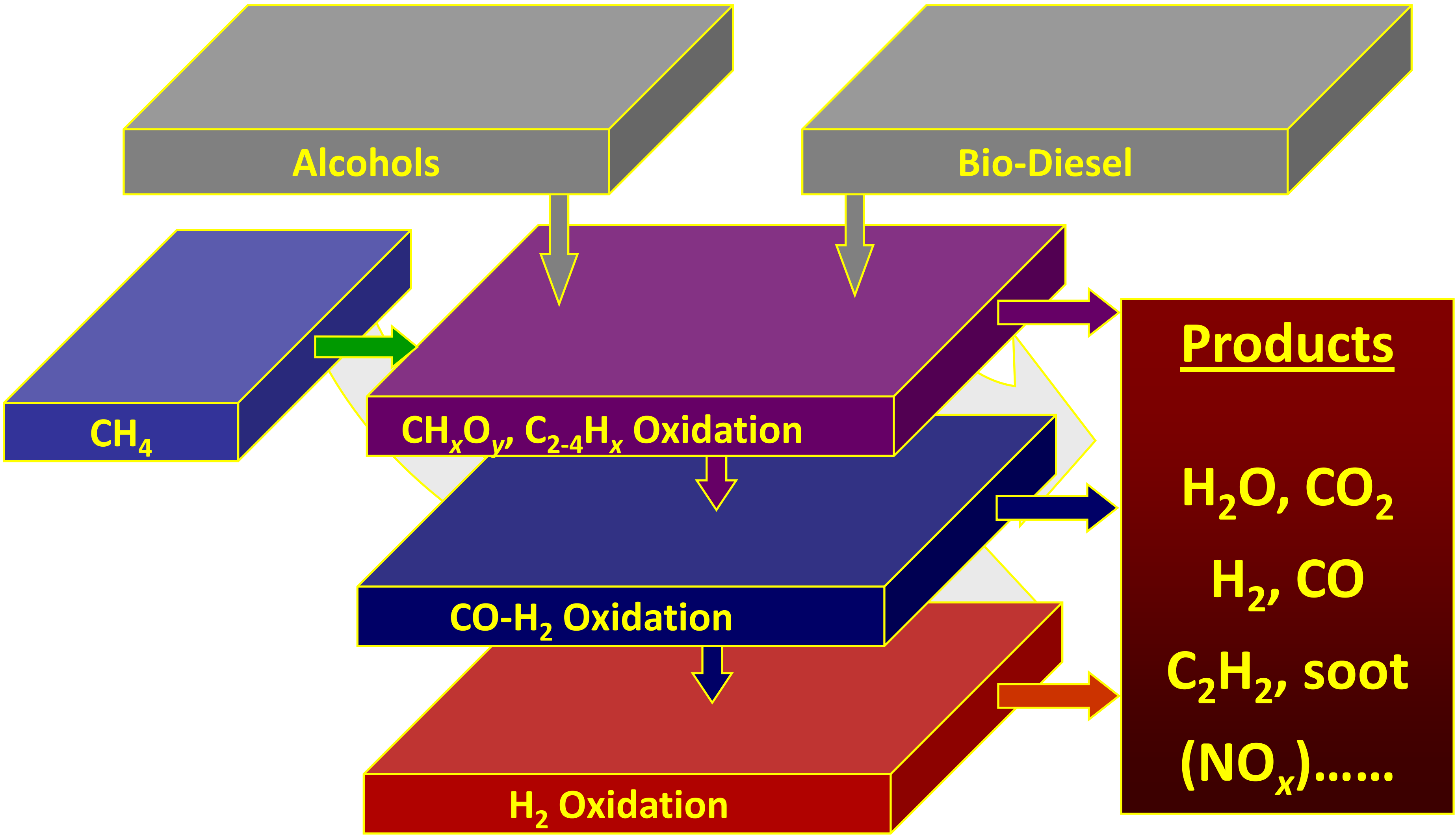
- Rapid deployment for combustion engine design.
- Rapid adaptation and utilization of emerging synthetic or biofuels.
- Science-based fuel design and blending strategies.

Combustion – a Multiscale, Multiphenomenon Challenge

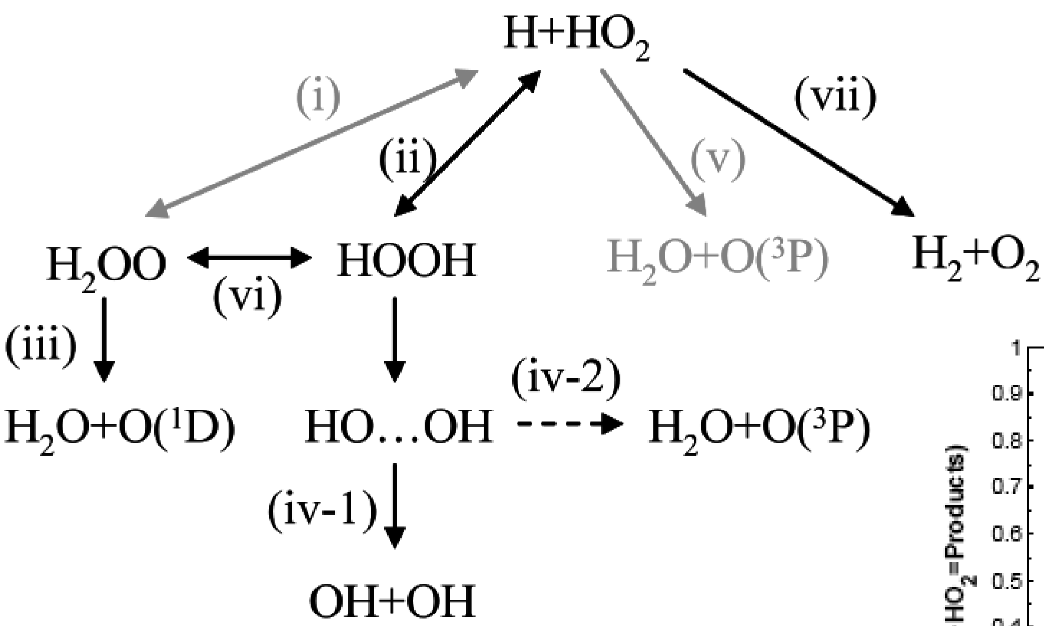


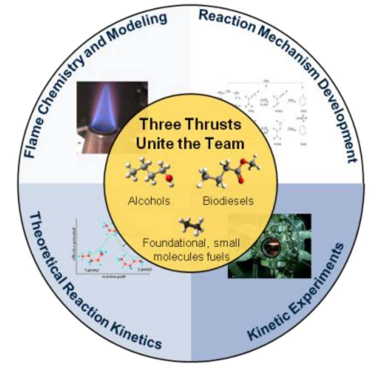
- Highly nonlinear problems
- Large thermo-dynamic condition space
- Mathematically ill-defined

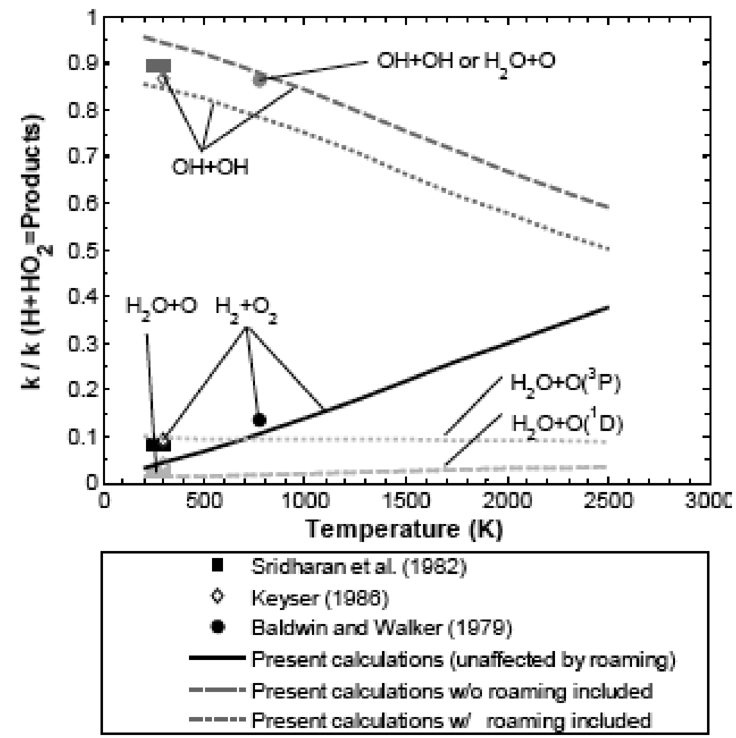
Combustion Reaction Model Hierarchy



Ab initio & reaction rate theory predictions of elementary reaction rate constants



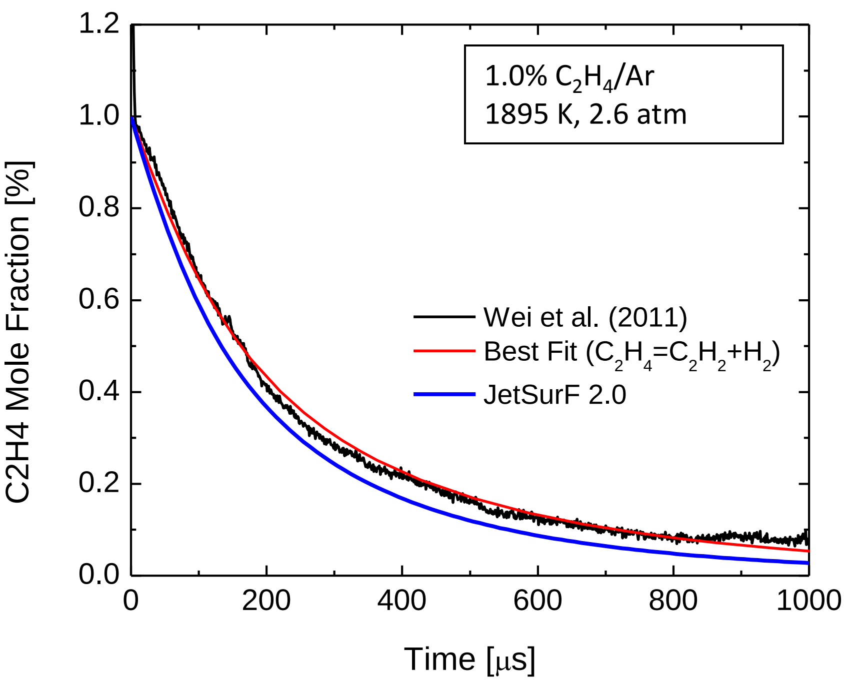




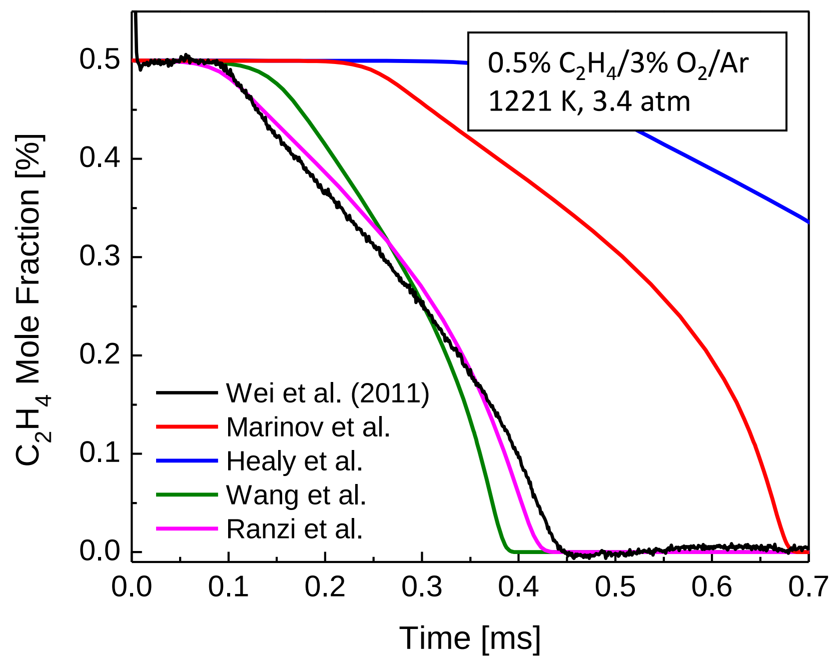
Klippenstein/ANL

Shock tube/laser absorption measurements: Ethylene time-Histories

Ethylene Pyrolysis
Rate Determination: $C_2H_4 \rightarrow C_2H_2 + H_2$

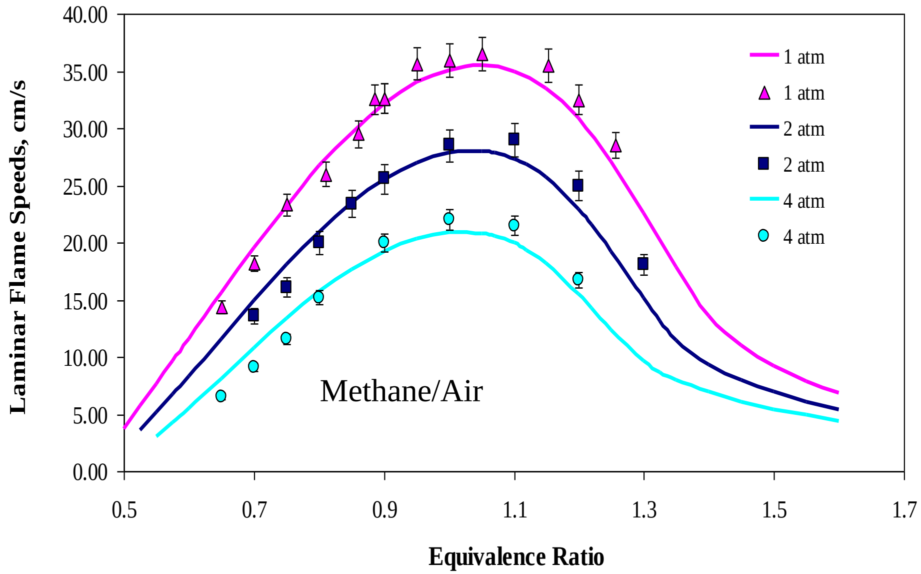


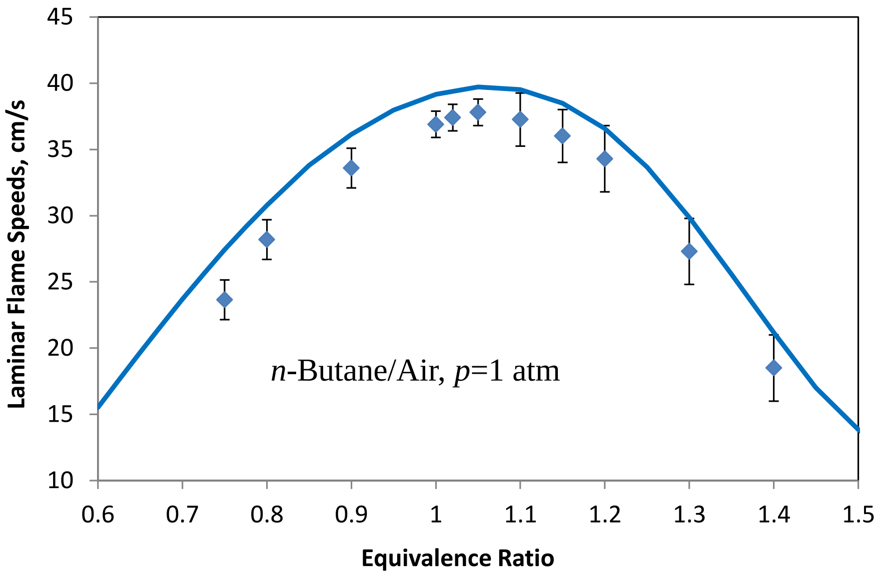
Ethylene Oxidation



Davidson & Hanson/Stanford

Measurement and model validation: laminar flame speeds

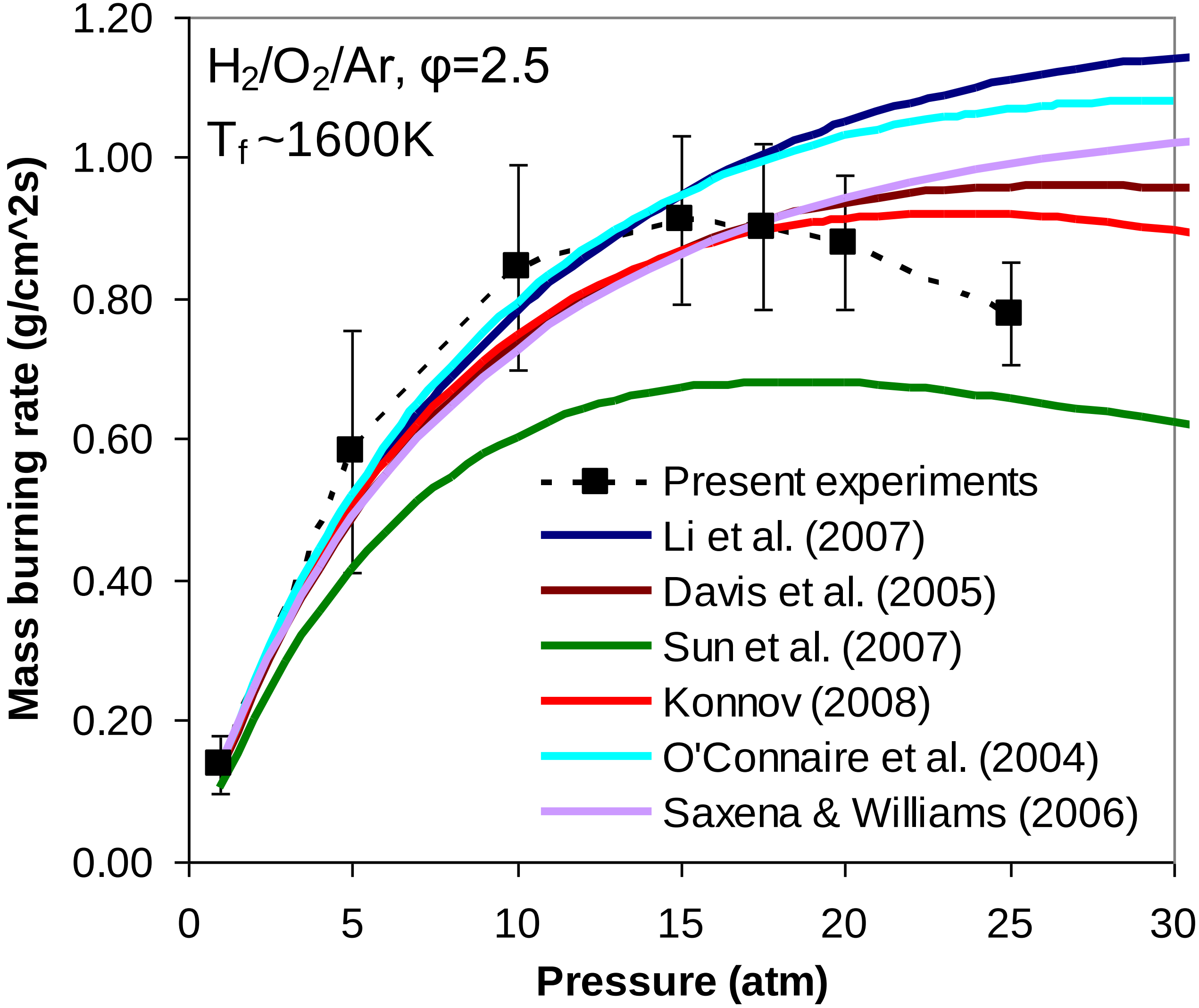




Symbols: experimental data
Lines: USC-Mech II predictions

Egolfopoulos/USC

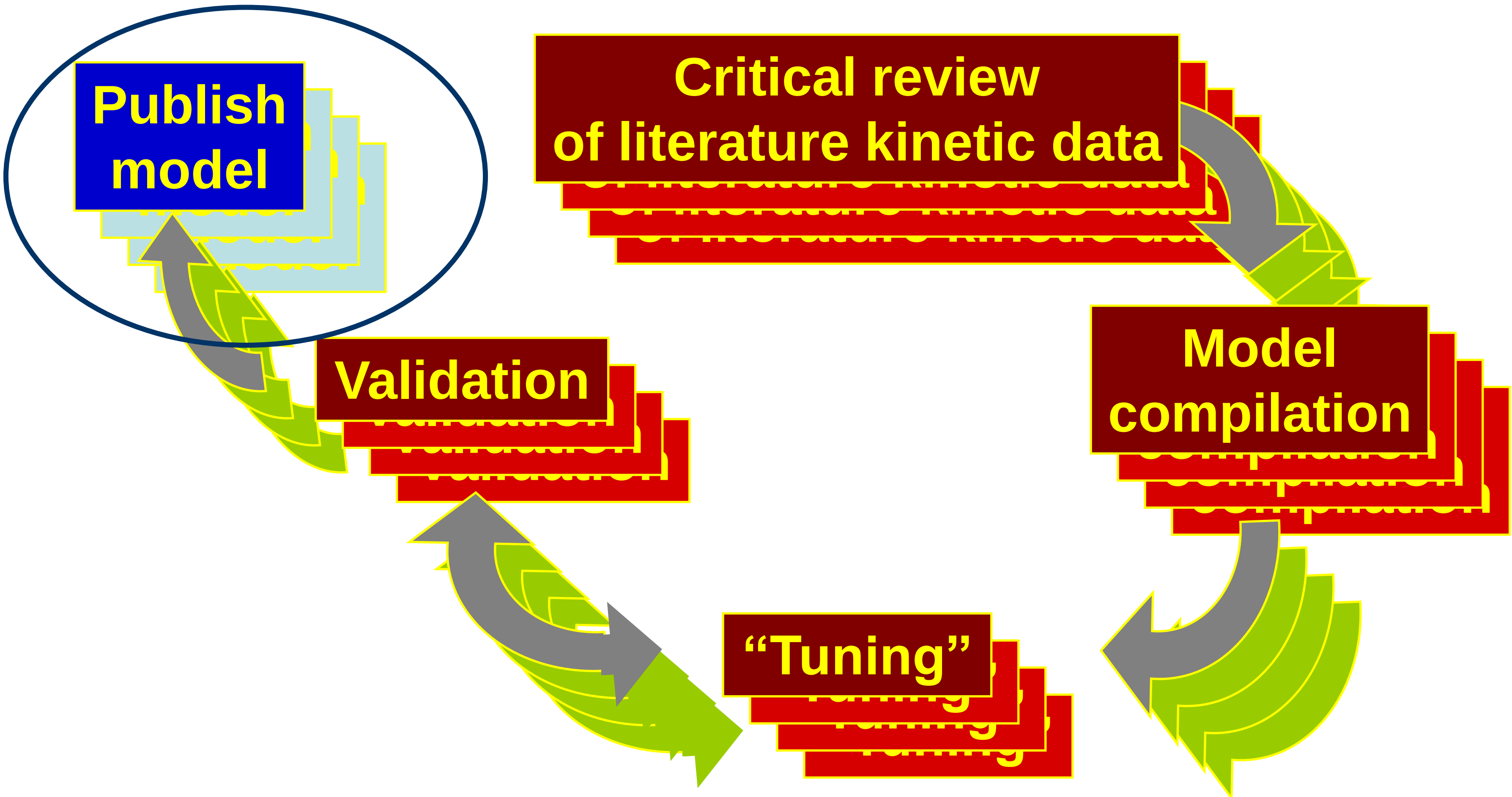
Measurement and model validation: mass burning rate of hydrogen



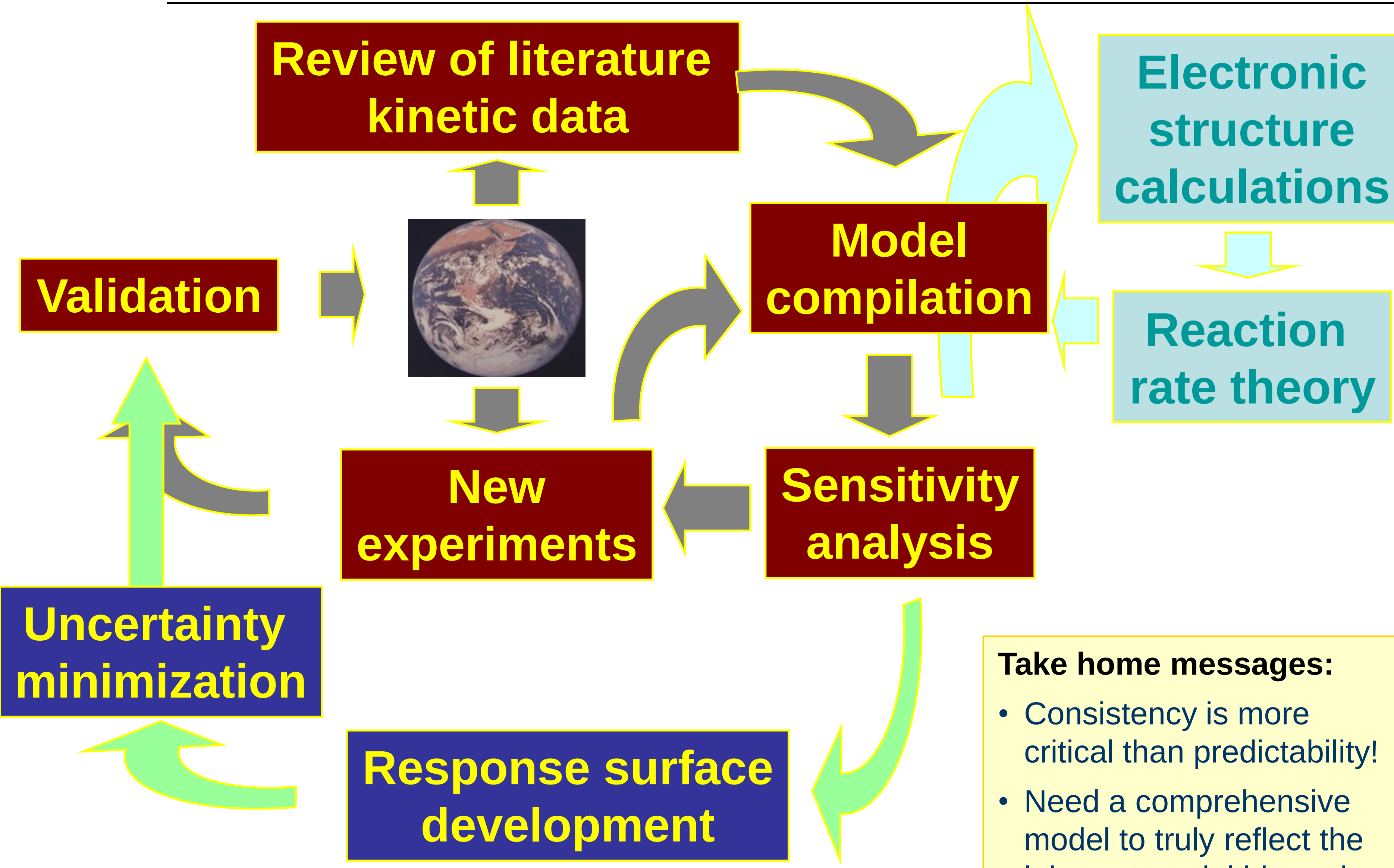
Dryer & Ju/Princeton

Combustion Reaction Model Development (the old approach)

proliferation of models



A New Approach



Take home messages:

- Consistency is more critical than predictability!
- Need a comprehensive model to truly reflect the inherent model hierarchy.

Knowledge (Uncertainty) Quantification by Polynomial Chaos Expansions

$$\mathbf{x} = \mathbf{x}_0 + \boldsymbol{\alpha}\boldsymbol{\xi}$$

$$x_i = \frac{\ln k_i / k_{i,0}}{\ln f_i}$$

Data structure that describes a chemical model + associated uncertainty

$$\eta_r(\mathbf{x}) \cong \eta_{r,0} + \sum_{i=1}^{N} a_{r,i} x_i + \sum_{i=1}^{N} \sum_{j \geq i}^{N} b_{r,ij} x_i x_j$$

Represents some physics model, e.g. a laminar flame

$$\eta_r(\mathbf{x}, \boldsymbol{\xi}) = \eta_r\left(\mathbf{x}^{(0)}\right) + \sum_{i=1}^{m} \hat{\alpha}_{r,i} \xi_i + \sum_{i=1}^{m} \sum_{j=i}^{m} \hat{\beta}_{r,ij} \xi_i \xi_j$$

Predictions of a chemical model (e.g. mass burning rate) + associated uncertainty

Sheen, *et al*. (2009 & 2011)

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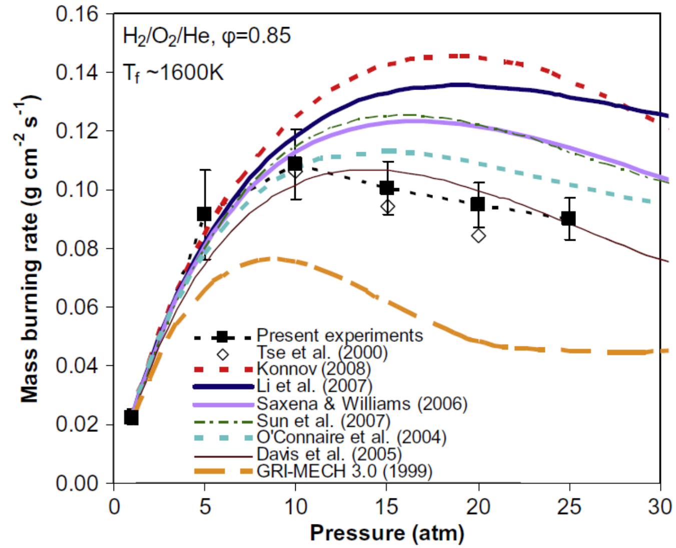

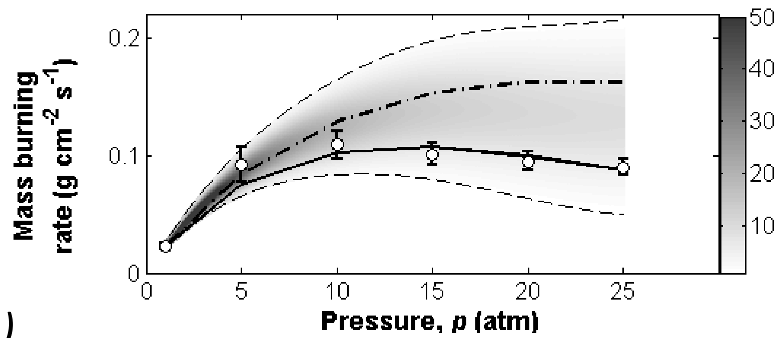

Sheen, *et al*. (2009 & 2011)

Uncertainty Minimization by Polynomial Chaos Expansions

$$\mathbf{x} = \mathbf{x}_0 + \boldsymbol{\alpha}\boldsymbol{\xi}$$

Chemical model
+ associated uncertainty

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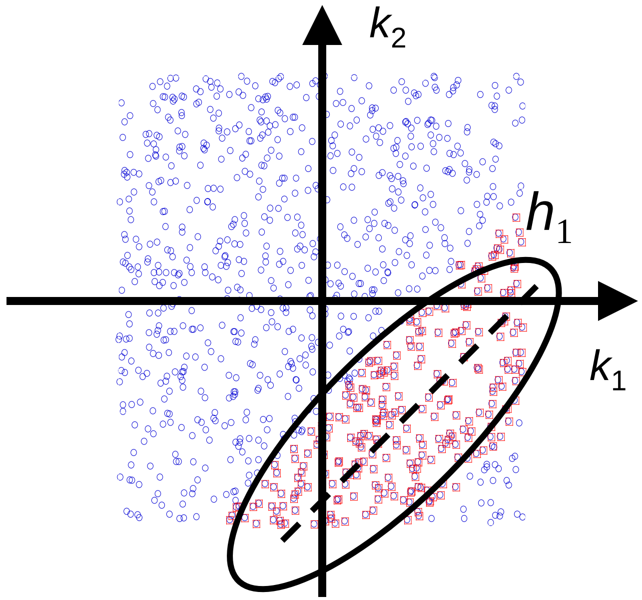
Physics model

$$\eta_r(\mathbf{x}, \boldsymbol{\xi}) = \eta_r\left(\mathbf{x}^{(0)}\right) + \sum_{i=1}^{m} \hat{\alpha}_{r,i} \xi_i + \sum_{i=1}^{m} \sum_{j=i}^{m} \hat{\beta}_{r,ij} \xi_i \xi_j$$

Predictions
+ associated uncertainty

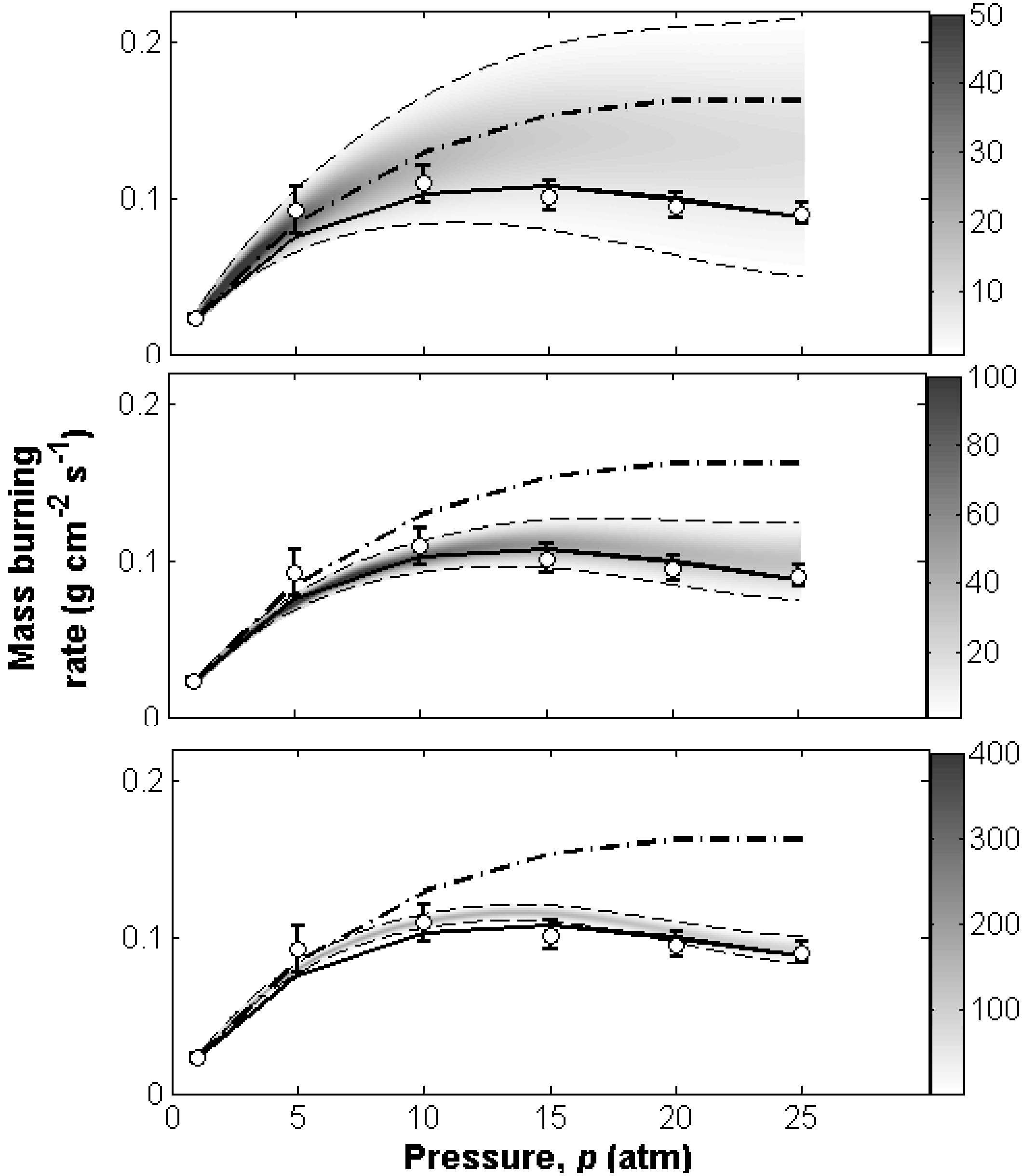


$$\Phi\left(\mathbf{x}_0^*\right) = \min_{\mathbf{x}_0} \left\{ \sum_{r=1}^{M} \frac{\left[\eta_{r,0}^{\text{obs}} - \eta_r\left(\mathbf{x}_0\right)\right]^2}{\left(\sigma_r^{\text{obs}}\right)^2} + \sum_{n=1}^{N} \frac{\left(x_{0,n}\right)^2}{\left(\sigma_n\right)^2} \right\}$$

$$\Sigma = \left[\sum_{r=1}^{n} \frac{1}{\left(\sigma_r^{\text{obs}}\right)^2} \left(\mathbf{b}\mathbf{x}_0^* \mathbf{x}_0^{*T} \mathbf{b} + \mathbf{a}\mathbf{x}_0^{*T} \mathbf{b} + \mathbf{b}^T \mathbf{x}_0^* \mathbf{a}^T + \mathbf{a}\mathbf{a}^T \right) + 4\mathbf{I} \right]^{-1}$$

$$\boldsymbol{\alpha}^* = \Sigma^{1/2}$$

Uncertainty Minimization by Polynomial Chaos Expansions – Measured Progress!



Considering no combustion experiments

Model constrained by knowledge prior to 2010

Model constrained by the latest knowledge

The Future

- Detailed reaction models with well defined uncertainties as the integral part of engine design tools

- Unified, consistent base model

- Active design of experiments

- Two-way knowledge propagation.