Bayesian Uncertainty Quantification and Calibration of a Clean Coal Design Code

Troy Holland[1,2] Sham Bhat[2] Peter Marcy[2] James Gattiker[2] Joel D. Kress[2]
Thomas H. Fletcher[1]

CO$_2$ Summit: Technologies and Opportunities
Santa Ana Pueblo, NM, April 2016

[1]Chemical Engineering, Brigham Young University, Provo, UT 84602
[2]Los Alamos National Laboratory, Los Alamos, NM 87544

CARBON CAPTURE MULTIDISCIPLINARY SIMULATION CENTER

CCSI
Carbon Capture Simulation Initiative

Synergistic Programs

- **CCSMC**- Carbon Capture Simulation Multi-disciplinary Center
- Created by PSAAP II, an NNSA program
- Oversight and technical support from NNSA labs (LANL, SNL, LLNL)
- Primary goal of promoting super computing in the community

- **CCSI I**

- DoE Office of Fossil Energy

- Primary goal of assisting industry in making carbon capture a feasible reality

- Provides tools for industry friendly (small cluster and desktop) models and simulation based design

Basic data models from CCSMC are improved via tools designed in CCSI.

Oxy-fuel combustion

Figure 1: Pulverized Coal Boiler

- Inject high purity O_2
- Recycle the flue gas
  - maintains a reasonable temperature
  - reduces the volume of the gas to be treated
  - results in a more easily captured CO_2 stream
- Drastically changes the furnace environment
  - CO_2, H_2O, and O_2 all become important
  - Radiation, O_2 diffusion, and combustion regimes all change
 - Endothermic reactions occur concurrently with oxidation

A potential retrofit technology to give industrial coal power plants a relatively cost-effective carbon capture system.

Char Conversion (my work in Basic Data Models)

Raw coal heats and reacts in several steps:

- Particle heating (typical industrial heating rates at ~ 10^5 K/s)
- Devolatilization/Swelling/Crosslinking
- Char conversion
  - Exothermic (O_2)
  - Endothermic (CO_2 and H_2O)
  - Needs to be modeled with detailed transport and kinetics
  - Current work is focused on the thermal annealing of coal char

My work takes basic data submodels, builds basic data macro-models, and propagates the uncertainty.
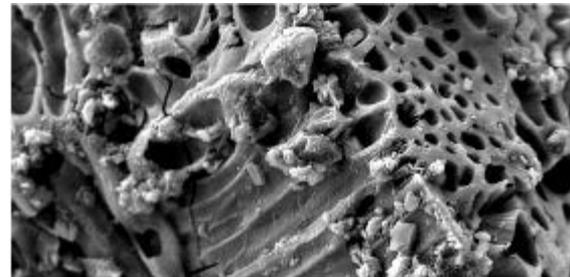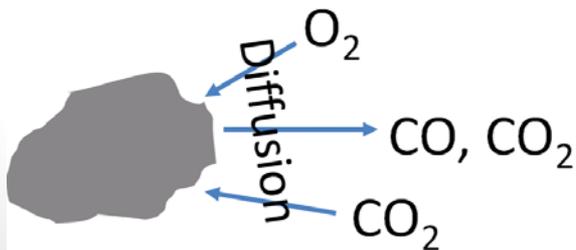

Figure 2- Pyrolyzed char

# Uncertainty Quantification – General Principles
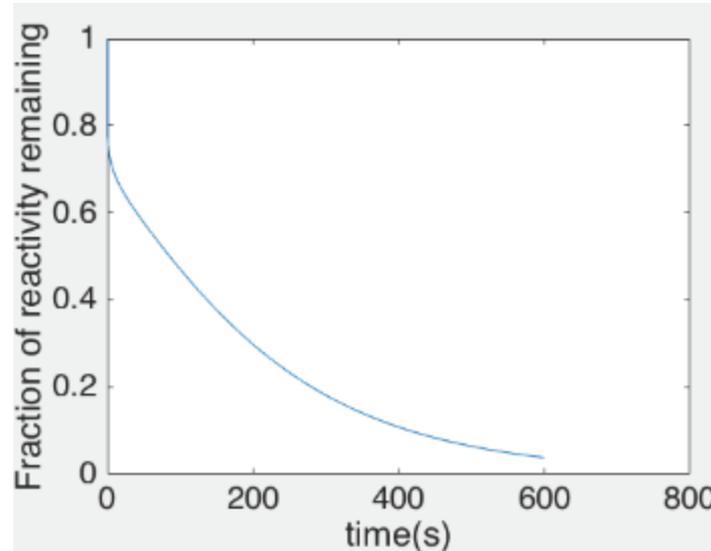
Annealing sub-model curve


Macro Model

Basic Data Model

Single best fit point

Char burnout from comprehensive code
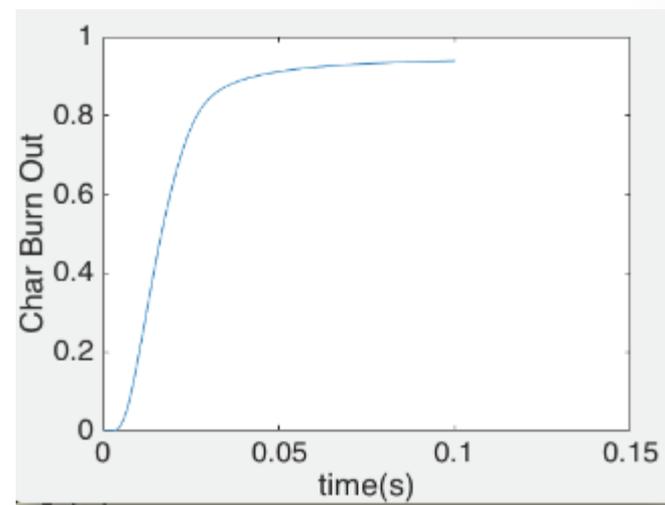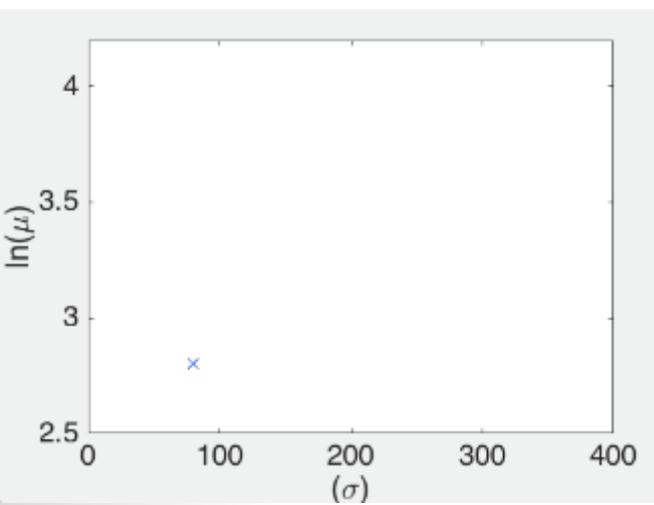
Uncertainty Quantification – General Principles

Annealing sub-model curve

Macro Model

Basic Data Model

**Single best fit point**
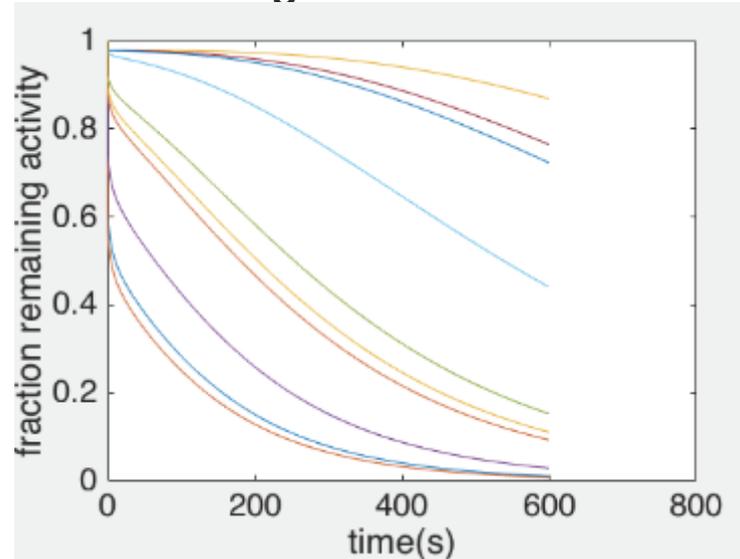
Char burnout from comprehensive code
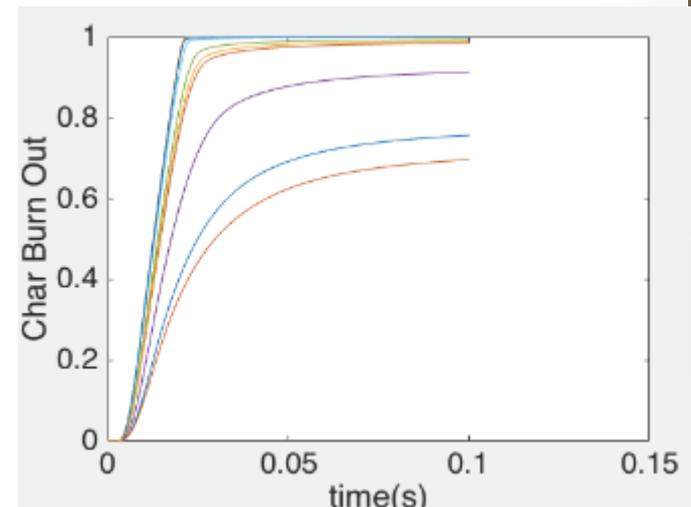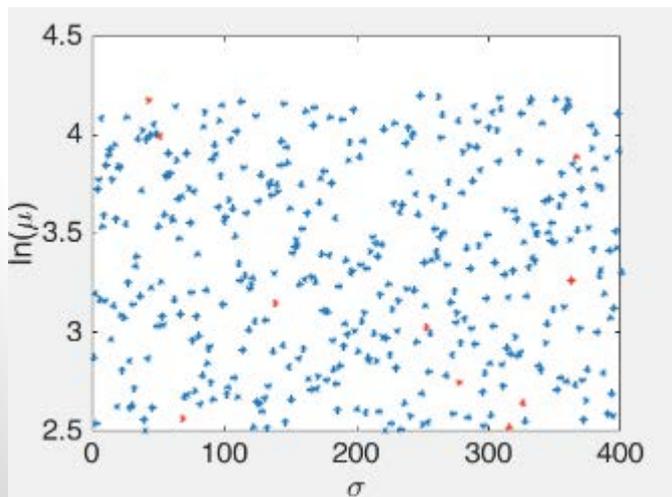
Uncertainty Quantification – General Principles

Any calibration method accomplishes something similar. The remainder of these slides highlight the unique virtues of the CCSI tool set.

Annealing sub-model curve

Macro Model

Basic Data Model
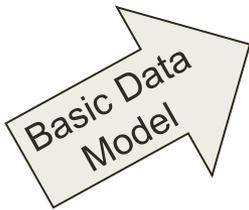
Single best fit point
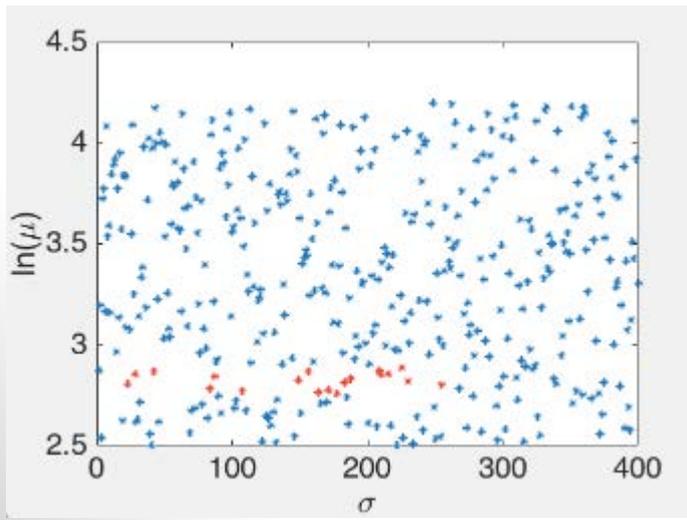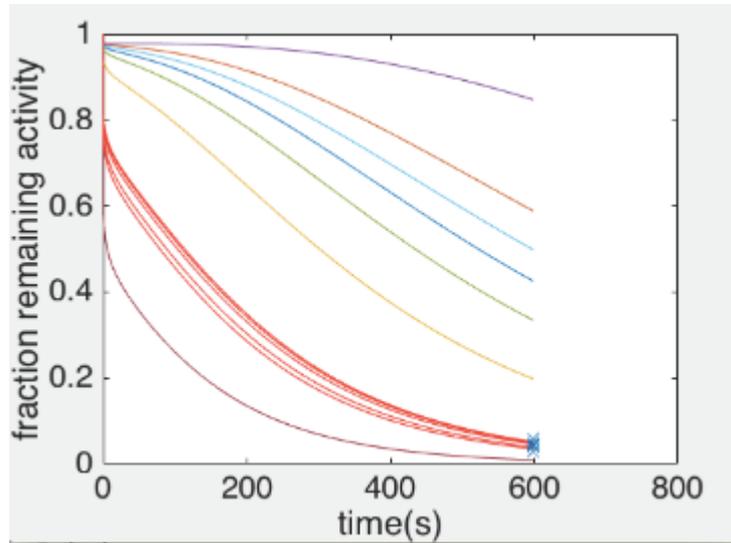
Char burnout from comprehensive code
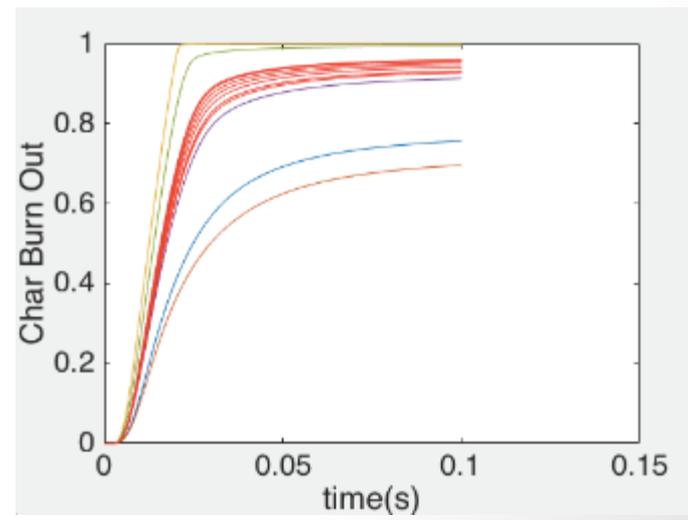
CCSI Calibration/UQ Paradigm

- **General UQ:** Find a plausible set of model parameter values (θ) that best produce the reality of experimental data.

- **Bayesian paradigm:** put a prior distribution on θ and condition on the experimental data to refine this prior distribution.

- Represent the physical system as the model (η) plus discrepancy function (δ) plus the measurement error (ε)
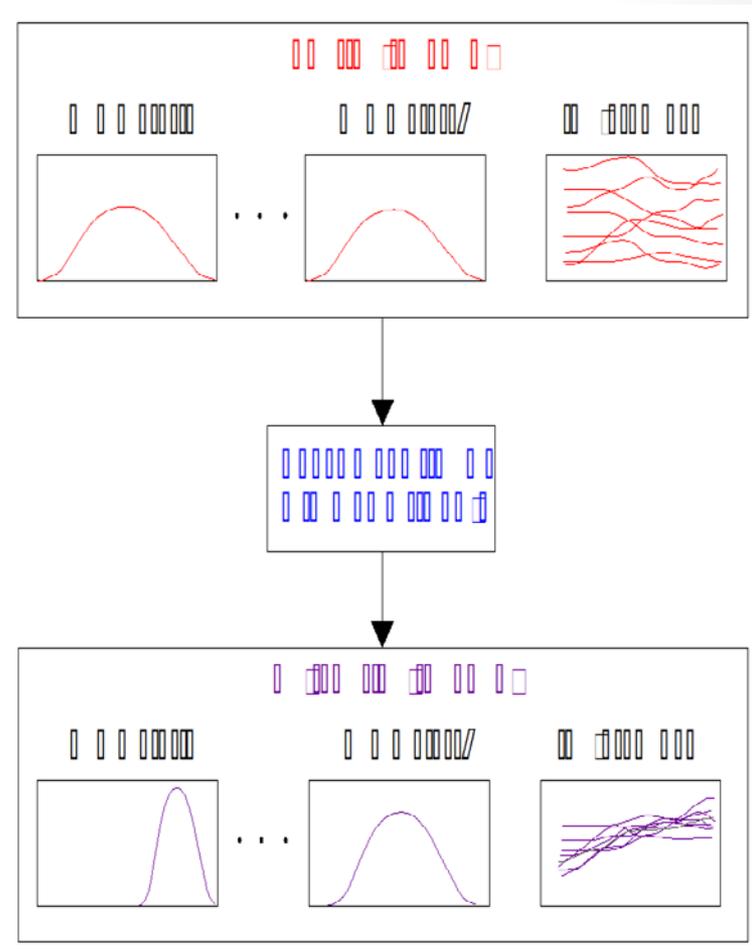
Many traditional UQ methods substantially exaggerate the actual uncertainty, and those that don't exaggerate uncertainty typically fail to account for systematic model bias.

Past CCSI UQ Applications – Solvent and Sorbent Models

Sorbent apparatus schematic

- Sample Equations:
  - Thermodynamics (**assumed known**)
  - Mass transport (calibrated)
  - Kinetics (calibrated)

$$D = D_0 \left(\frac{T}{T_{ref}}\right)^{\beta} \left(\frac{\mu}{\mu_{ref}}\right)^{\alpha}$$

$$k = k_0 \exp\left(\frac{-E_A}{R}\left(\frac{1}{T} - \frac{1}{T_{ref}}\right)\right)$$
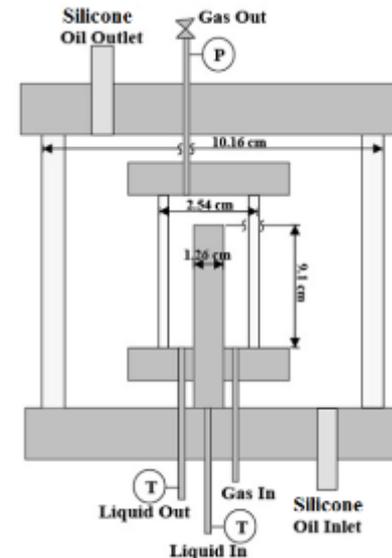
I mention these models very briefly to highlight the flexibility of the tool set.

Prior Distributions – Domain Expert Belief about the System
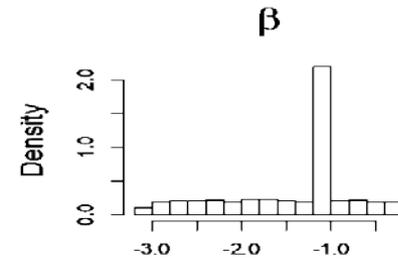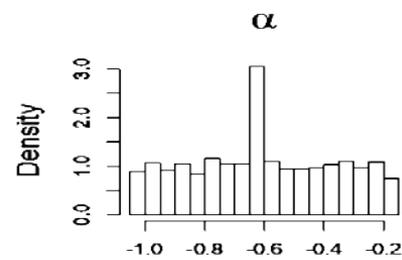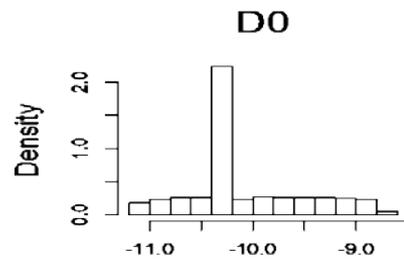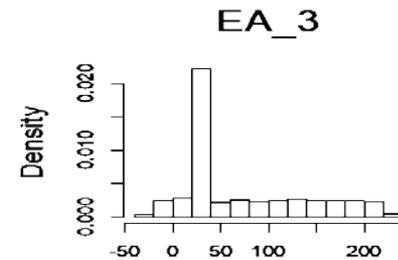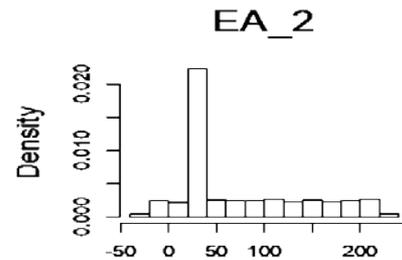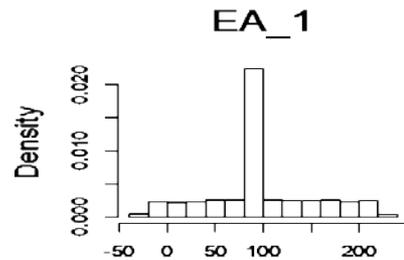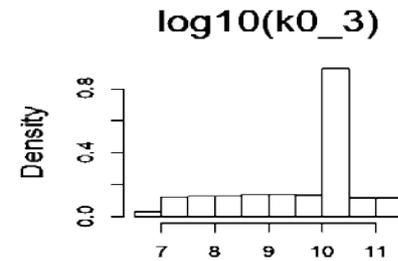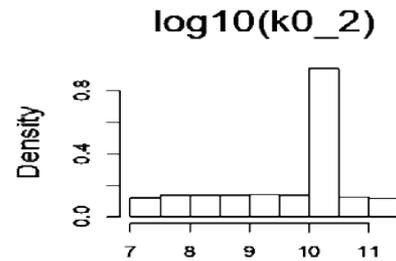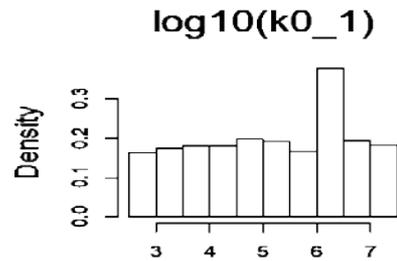
The domain expert had past experience to give him some idea about where the true parameters might be.

Past Basic Data Models – Solvent Posteriors
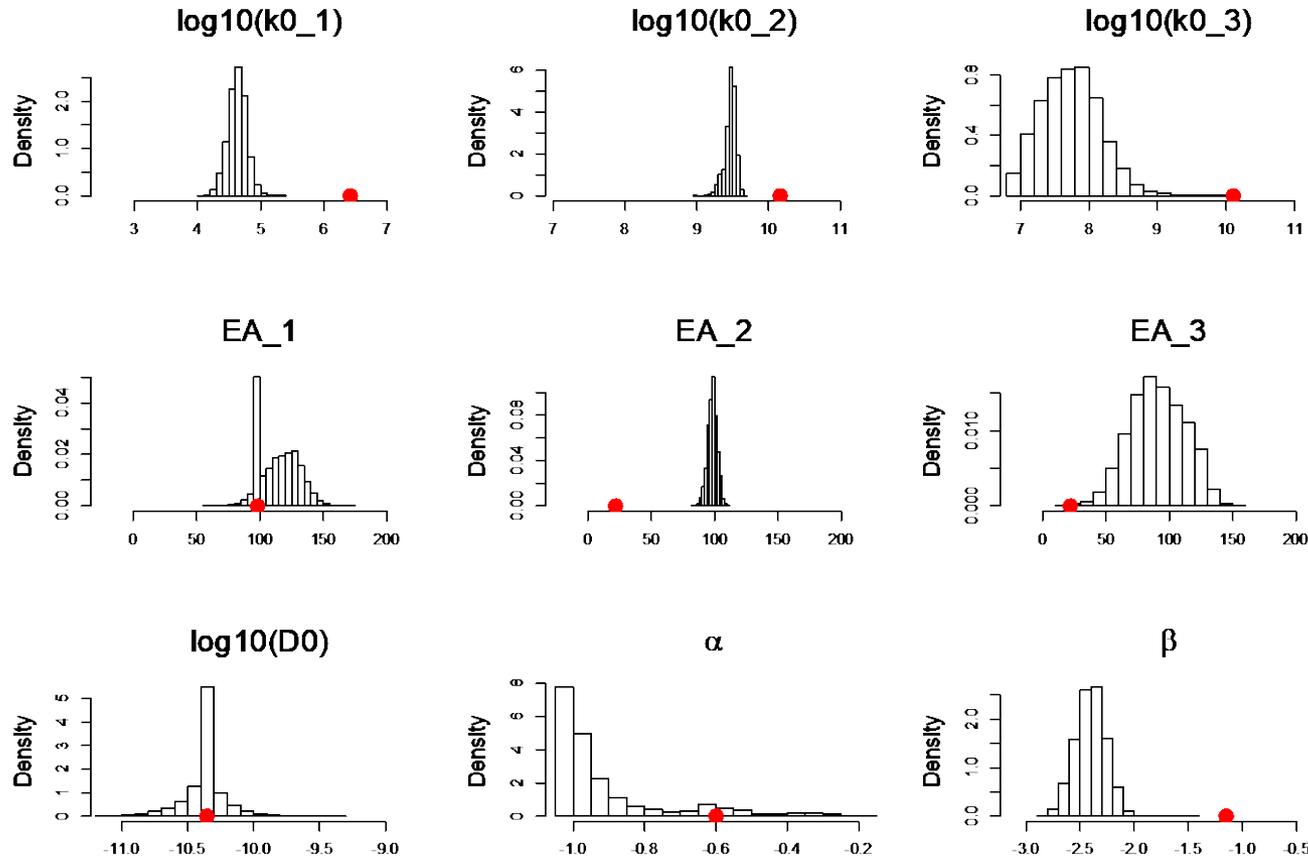
The domain expert's initial belief was generally incorrect, but the data as a whole led to well defined peaks of parameter density.

My Work – A Radically Different Model
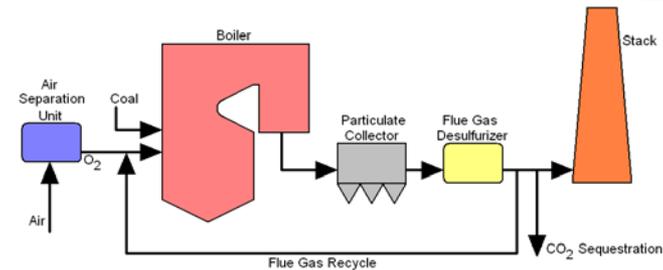
- CCK\oxy is single particle model with detailed physics for all stages of combustion and gasification from raw coal to complete burnout
- Direct and indirect industrial application
  - CCSCM uses exascale computing to optimize industry designs
  - Industry directly applies the comprehensive code to train surrogates
- Each sub-model contains uncertain parameters and model discrepancy
- The most sensitive parameters are targeted and addressed

The next several slides are a practical example applying the CCSI tool suit to a model and relevant data. The output is a calibrated model with informed discrepancy from reality and quantified uncertainty.

CCSI UQ Tools –1 Sensitivity Analysis

- Sensitivity analysis over ~25 (confirmed with CCSI decomposition of variance tool)

Table 1 – Total sensitivity measures for all O_2 conditions and each individual condition

Mean Sensitivity Measures		Sensitivity for O_2 Mole Fraction=0.12		Sensitivity for O_2 Mole Fraction=0.24		Sensitivity for O_2 Mole Fraction=0.36	
Variable	Importance	Variable	Importance	Variable	Importance	Variable	Importance
E_A	0.74	E_A	0.76	E_A	0.72	E_A	0.75
N	0.51	N	0.55	N	0.51	N	0.48
Ω	0.27	Ω	0.40	Ω	0.22	α	0.22
α	0.20	g_d	0.20	α	0.22	σ	0.20
g_d	0.20	t_r	0.18	g_d	0.21	g_d	0.19
σ	0.18	α	0.18	σ	0.17	Ω	0.17
t_r	0.14	σ	0.17	t_r	0.12	t_r	0.11

An important first step to refining complex models: Determine which submodels are worth the time it takes to improve them.

Sample data

- The body of literature data shows that annealing depends on many things, but most especially on
  - Heating rate
  - Soak time
  - Peak particle temperature
  - Coal precursor

This sample shows that annealing conditions (or pyrolysis conditions) DO in fact have an enormous impact.

**Sample raw data used in the calibration (from a South African bituminous coal, Senneca et al. 1999)**
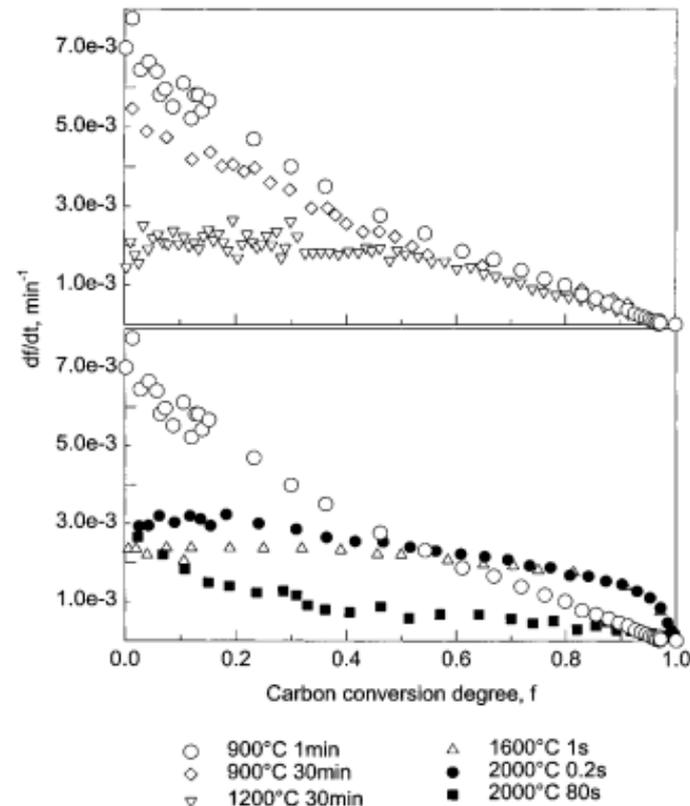
Figure 3. Typical gasification-rate profiles of heat-treated samples.

Calibration Step 1: Define the Model

$$\frac{df_i}{dt} = -k * exp(\frac{-E_{A\_anneal,i}}{R * T}) * f_i$$

- k – the Arrhenius preexponential factor
- E_A – the activation energy of bin i
- f_i – the fraction of active sites assigned to bin i

Sample "binned" log-normal distribution
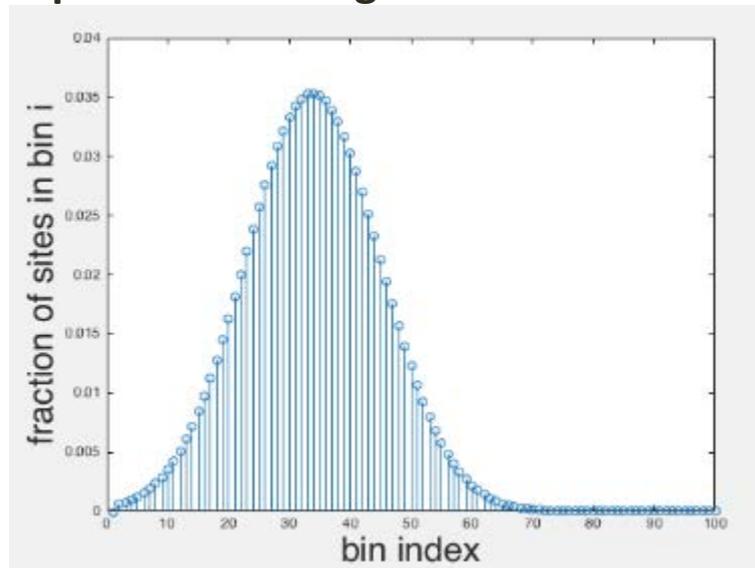
Calibration Step 2: Choose Parameters and Priors

- Choose the parameters and their priors
  - Informed by sensitivity analysis
  - In this case, find k and the right activation energy distribution
  - Parameters: σ, μ, and k
  - Priors limited by the activation energy of amorphous carbon reordering to crystalline graphite (~800 kJ/mol) and observed rates of activity decrease

$$\frac{df_i}{dt} = -k * exp(\frac{-E_{A\_anneal,i}}{R * T}) * f_i$$

Priors contain any past information/experience that lead a domain expert to believe parameter values lie in a given range and probability distribution

Calibration Step 2:
Choose Parameters and Priors

- Literature attempts (past experience) found a shallow bowl of parameter space

- No justification to weight the priors, but some justification to bound them

Optimized data fit from mid-90's literature
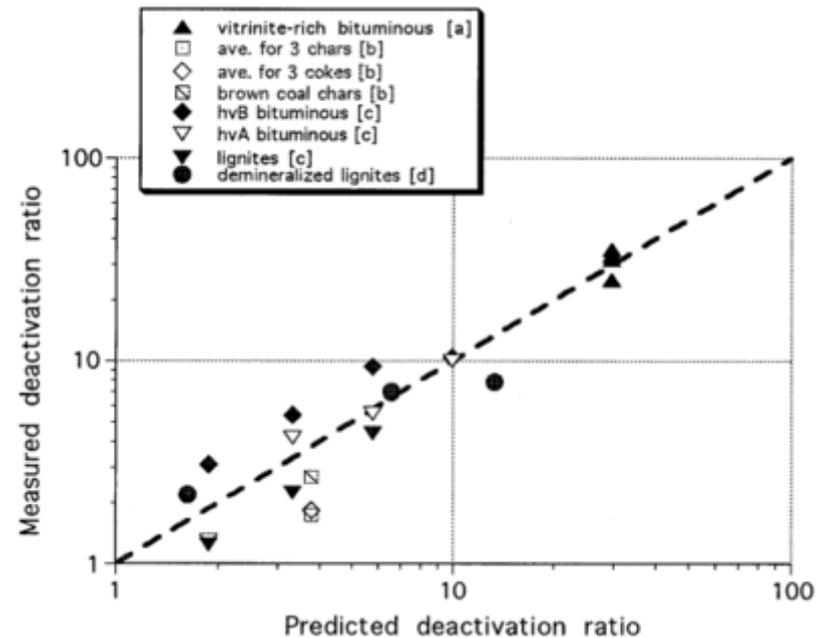
Figure 4: Original CBK annealing model

Calibration Step 2:
Choose Parameters and Priors
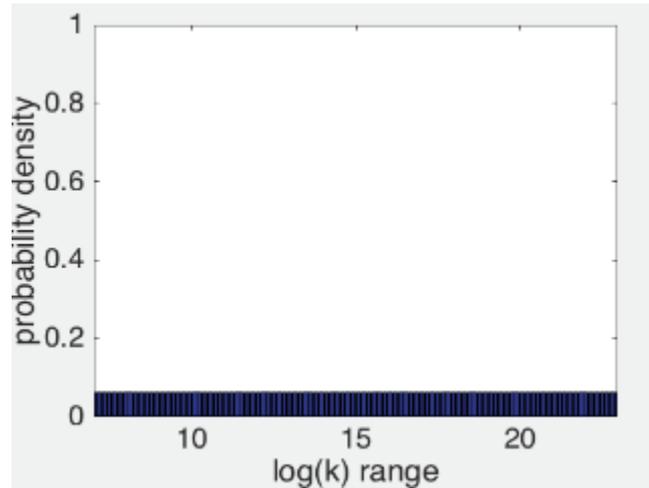
Uniform probability density priors for μ, σ, and k
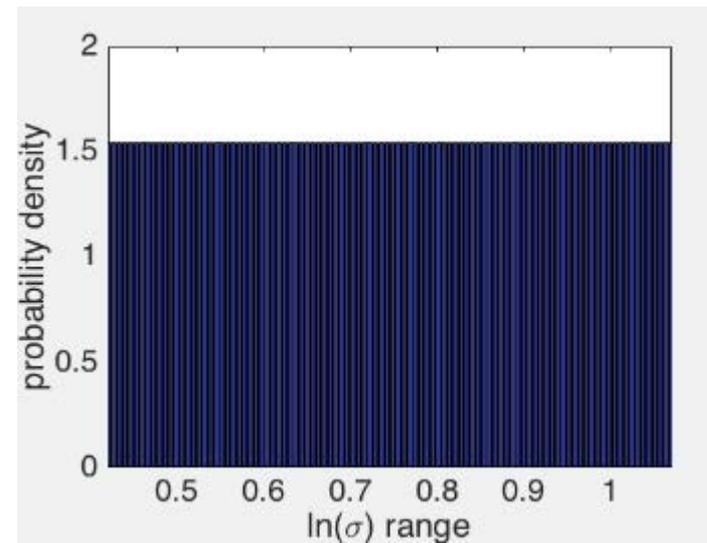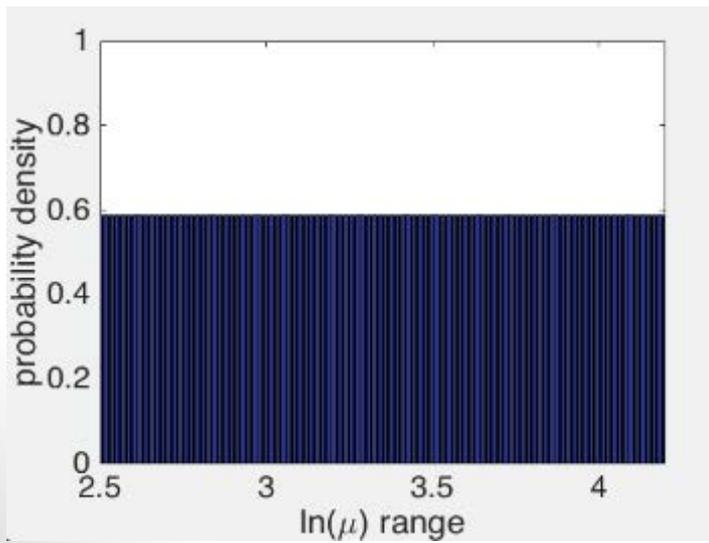
Calibration Step 3
Train the Emulator

- The emulator is a surrogate model with uncertainty
- It is "trained" using the annealing model outputs and is able to predict outputs for the model at any set of input conditions, even if the model was not actually run at those conditions
- Every prediction comes with defined uncertainty

Calibration Step 4:
Execute the GPMSA code

- Matrices of model inputs and outputs train the emulator
- The emulator executes tens of thousands of model runs to produce posterior distributions
- The posteriors show uncertainty around the parameter space

The GPMSA code ultimately shows both model predictions (and attendant uncertainty) and model + discrepancy predictions. This allows the engineer to quantify how precisely the model predicts data, and how accurately the model mimics reality.

Calibration Step 4:
Original Annealing Model with Original Data

$$y_i = \eta(\mathbf{x}_i, \boldsymbol{\theta}) + \delta(\mathbf{x}_i) + \varepsilon_i$$

Red lines: η only

Black Dots: data points

Calibrated Predictions
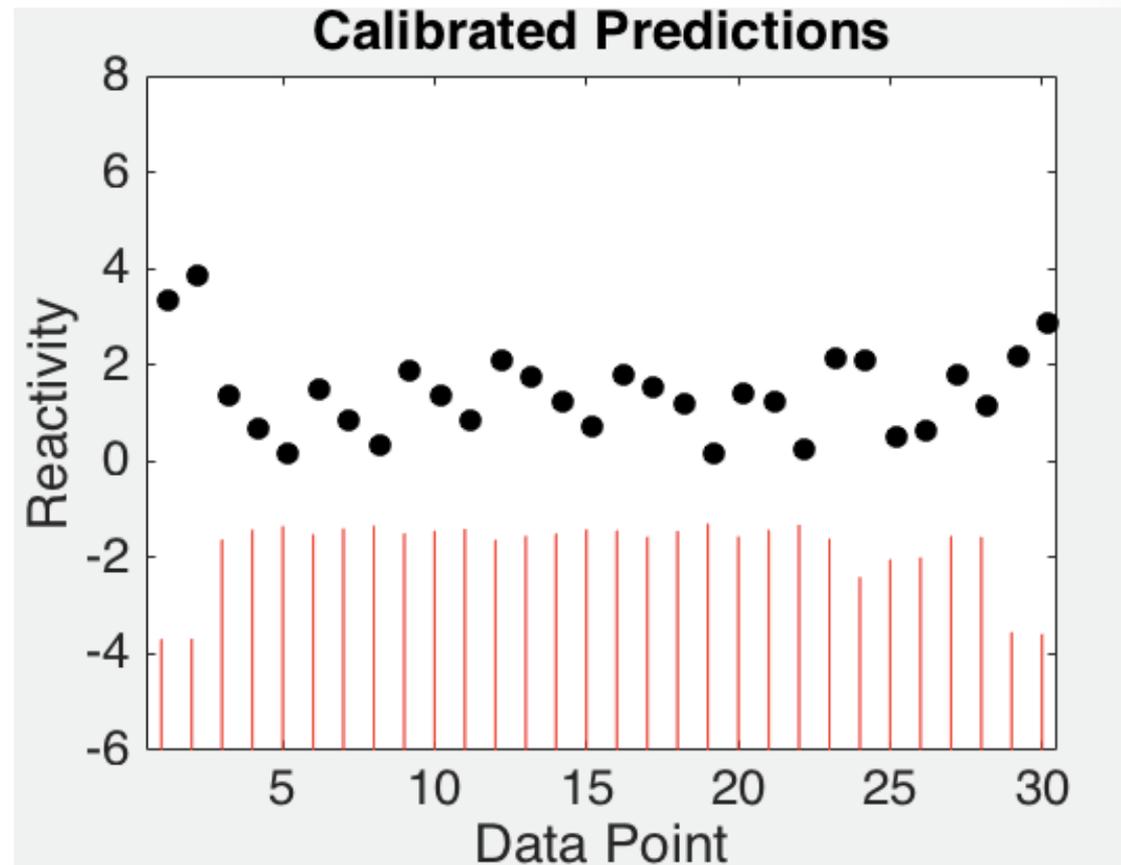
**The initial model does not capture the data at all.**

Calibration Step 4:
Original Annealing Model with Original Data

$$y_i = \eta(\mathbf{x}_i, \boldsymbol{\theta}) + \delta(\mathbf{x}_i) + \varepsilon_i$$

Red lines: η only

Black Dots: data points

Black Lines: η+δ+ε

Calibrated Predictions w/ Discrepancy
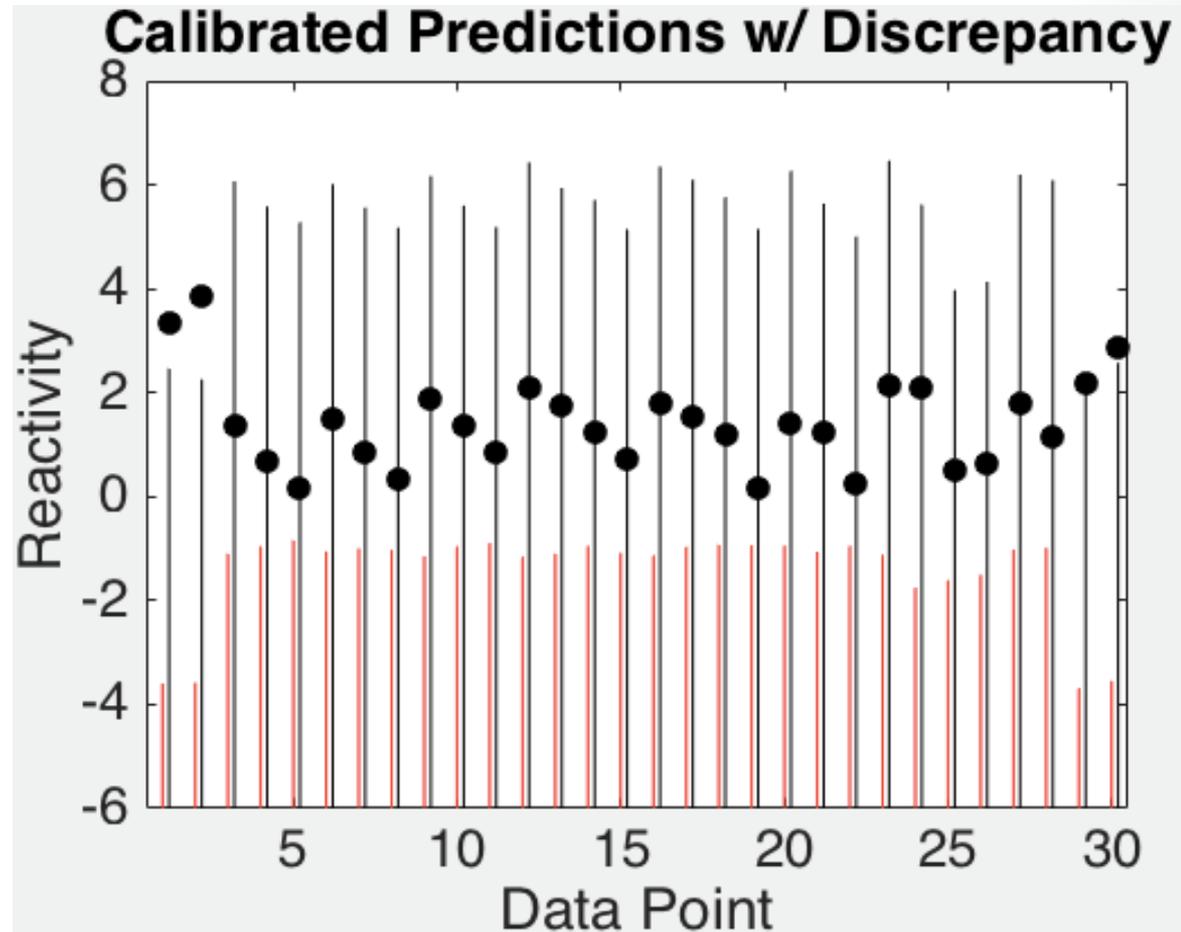
With the addition of a large discrepancy, the model mostly (but not entirely) captures the data.

Calibration Step 5 (iterative):

- Consider possibilities to reduce discrepancy and error
  - More data
  - Better quality data
  - Better physics in the model
  - If the model requires the discrepancy function to match data points, the model lacks important physics that should be identified and added.
    - Here we know that heating rate, peak temperature and coal type play an important roll that is neglected by the annealing model.

Calibration Step 5:
Consider possibilities to reduce discrepancy

- Reduce ranges from maximum potential values to ranges that only include the data
- Transform variables to more heavily sample the most important regions of parameter space

$$y_i = \eta(\mathbf{x}_i, \boldsymbol{\theta}) + \delta(\mathbf{x}_i) + \varepsilon_i$$

Red lines: η only

Black Dots: data points

Calibrated Predictions
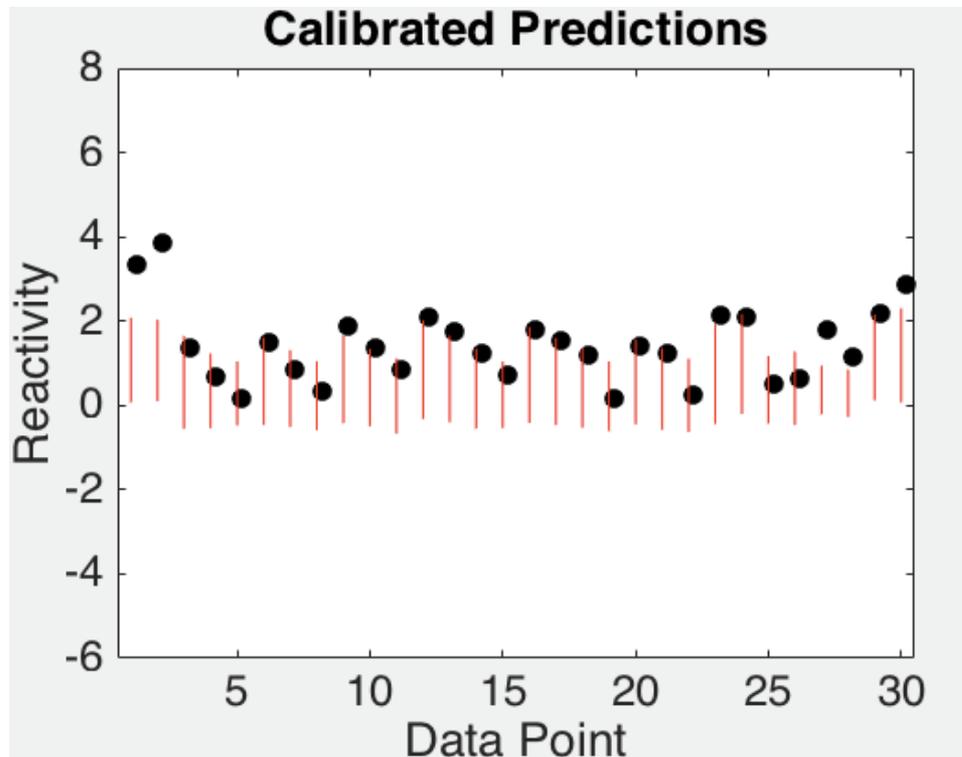
A smart exploration of the parameter space can be quite important.

Calibration Step 5:
# Improve the Experimental Design

- Reduce ranges from maximum potential values to ranges that include the data
- Transform variables to more heavily sample the most important regions of parameter space

$$y_i = \eta(\mathbf{x}_i, \boldsymbol{\theta}) + \delta(\mathbf{x}_i) + \varepsilon_i$$

Red lines: η only

Black Dots: data points

Black Lines: η+δ+ε

Calibrated Predictions w/ Discrepancy
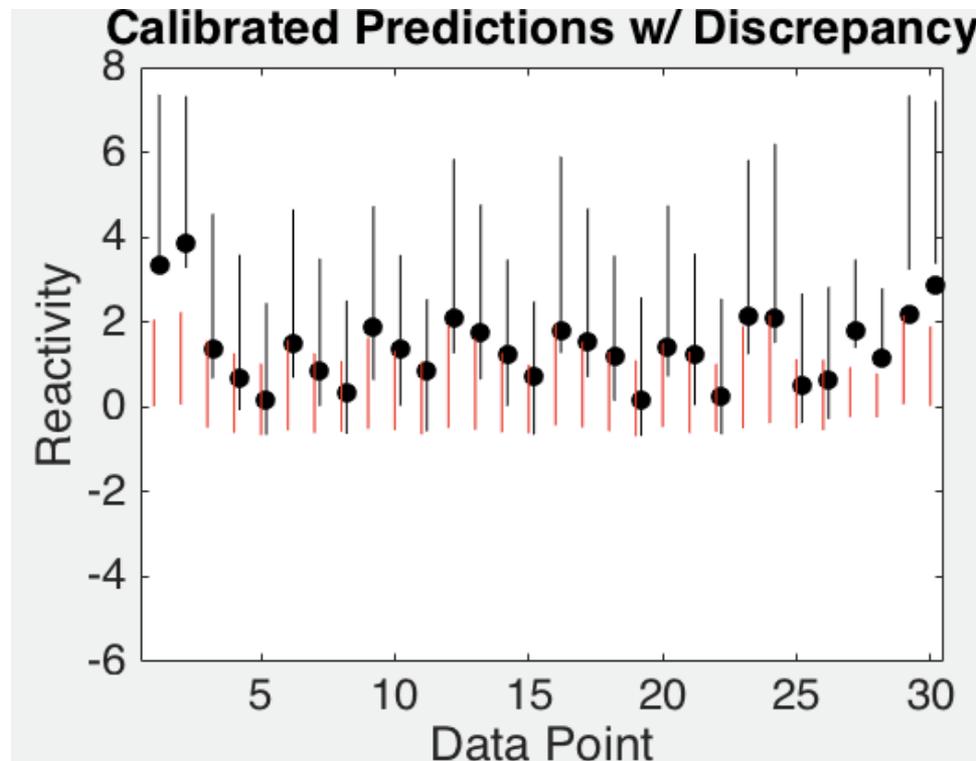
Despite the great improvement, the discrepancy and model still do not intersect all points. More is needed.

Calibration Step 5:
Improve the Experimental Design

- Reduce ranges from maximum potential values to ranges that include the data
- Transform variables to more heavily sample the most important regions of parameter space
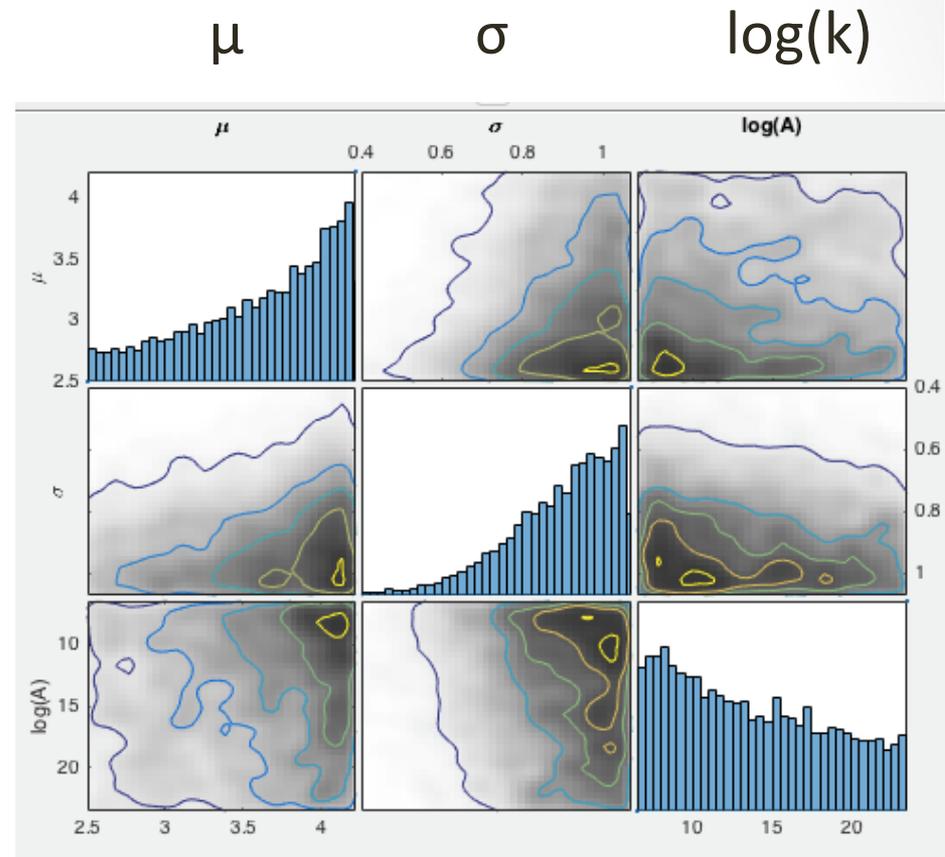
When the majority of the probability density is piled up on a boundary, the model is very likely deficient.

Calibration Step 5:
## Original Annealing Model with Expanded Data

- Expand the data set (legacy code is common, new data might well be available)

$$y_i = \eta(\mathbf{x}_i, \boldsymbol{\theta}) + \delta(\mathbf{x}_i) + \varepsilon_i$$

Red lines: η only

Black Dots: data points

Calibrated Predictions

(plot: x-axis "Data Point" from 50 to 150, y-axis "Reactivity" from -6 to 8)
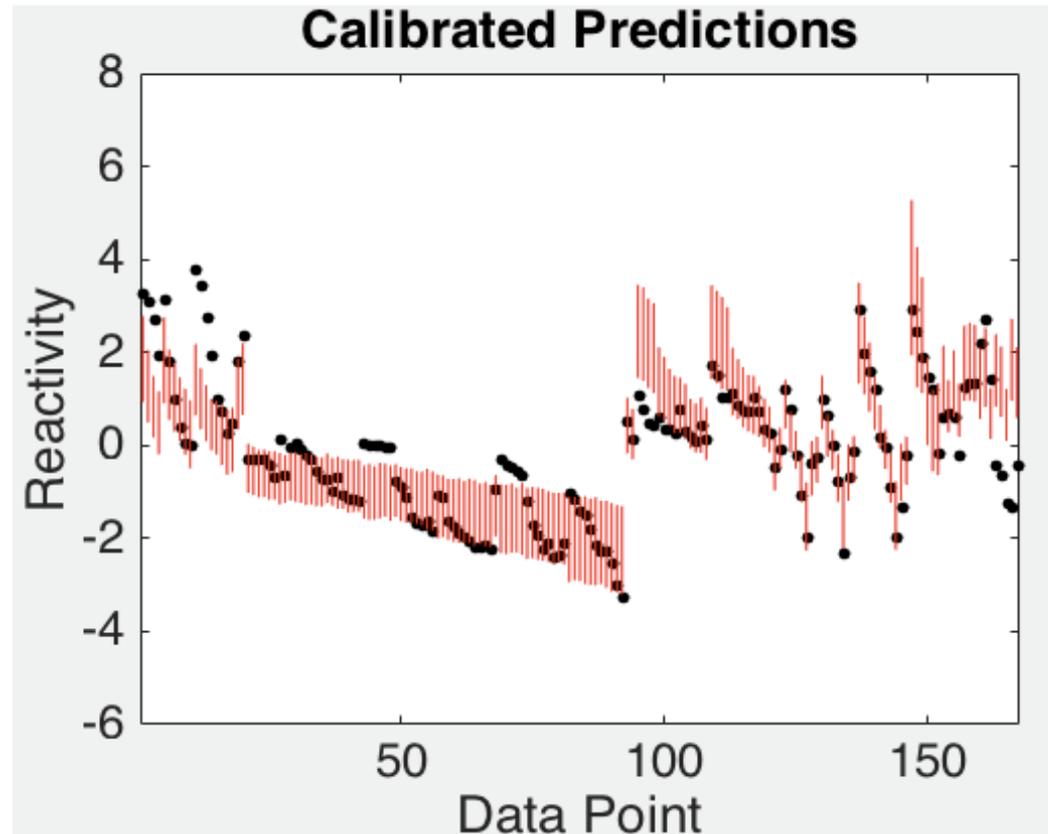
More data improves the fraction of points that the model can capture, but still fails to capture about 1/3 of the data.

Calibration Step 5:
Original Annealing Model with Expanded Data

- Expand the data set (legacy code is common, new data might well be available)

$$y_i = \eta(\mathbf{x}_i, \boldsymbol{\theta}) + \delta(\mathbf{x}_i) + \varepsilon_i$$

Red lines: η only

Black Dots: data points

Black Lines: η+δ+ε

Calibrated Predictions w/ Discrepancy
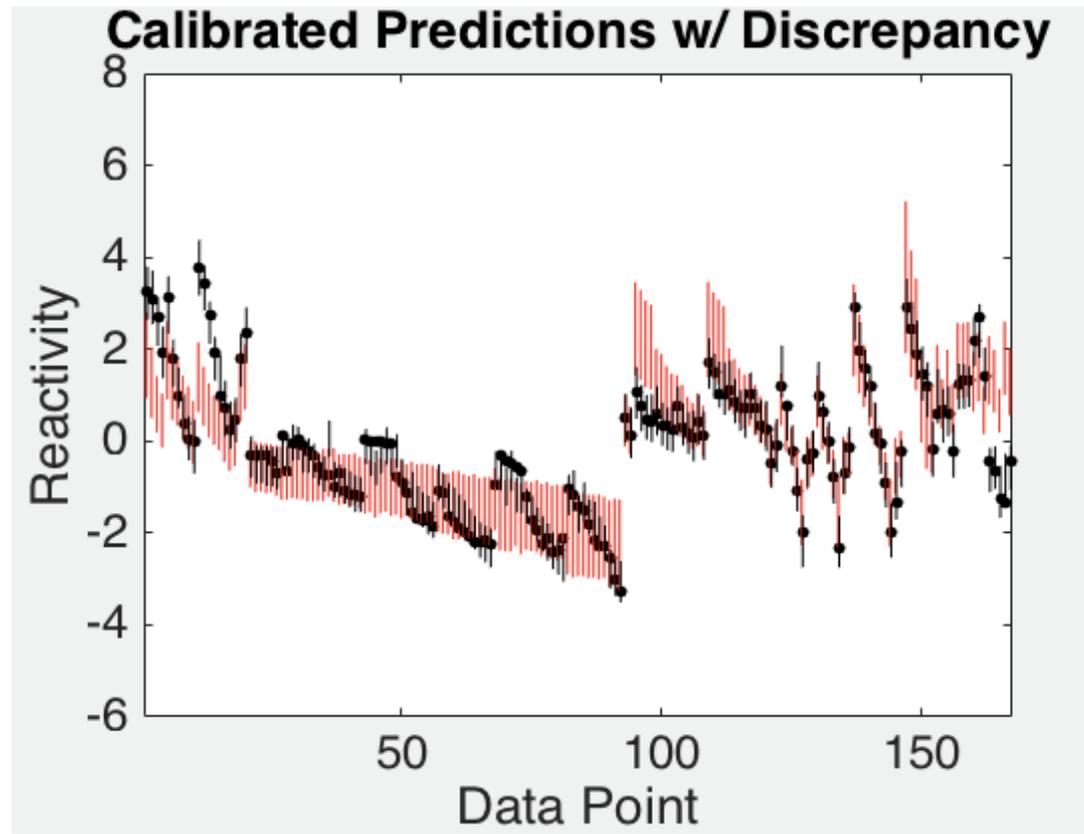
Discrepancies can now capture all the data, and are greatly reduced, but are still far from 0.

Calibration Step 5:
Original Annealing Model with Expanded Data

- Expand the data set (legacy code is common, new data might well be available)
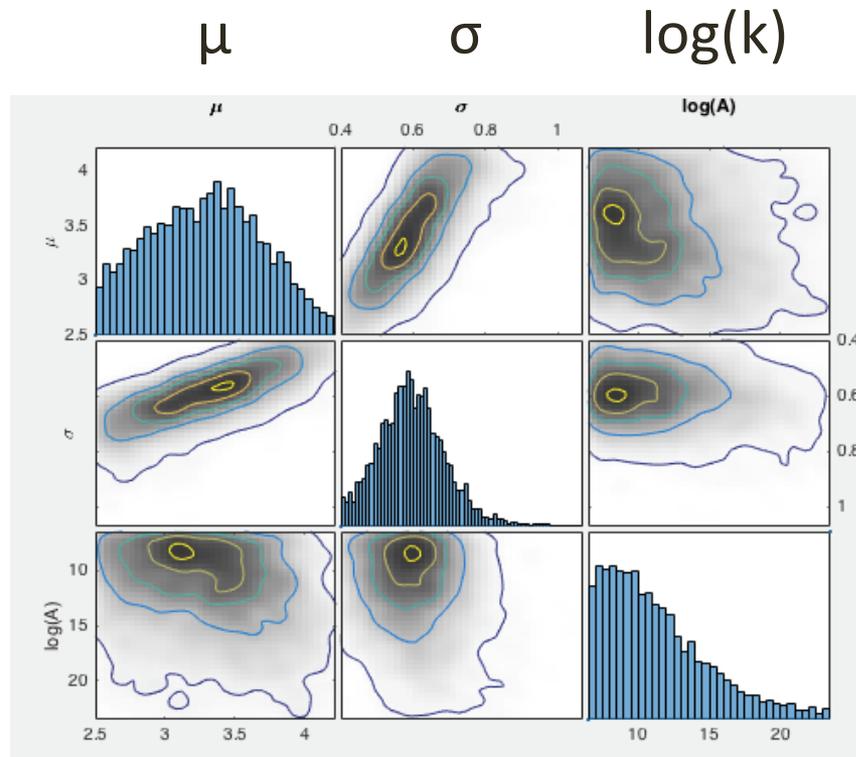

More and better data sharpen the peaks and narrow the parameter space, but no amount of data can overcome a model that has inadequate physics.

Calibration Step 5:
New Annealing Model with Expanded Data

- Manipulate the model form
- Add additional physics
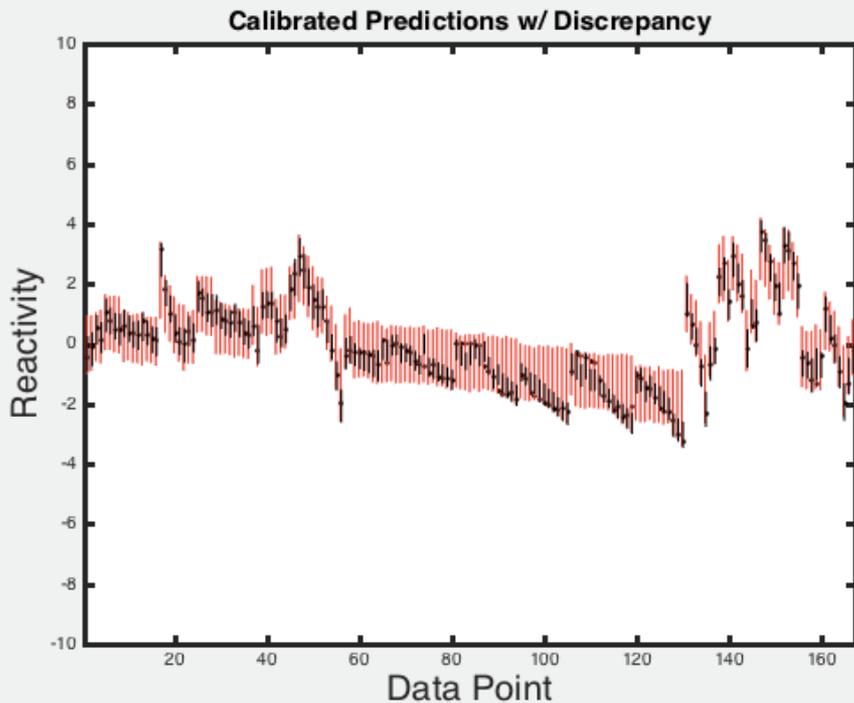
$$\mu = a * \text{Coal Quantificantion} + b \qquad a \qquad b \qquad \log(k)$$
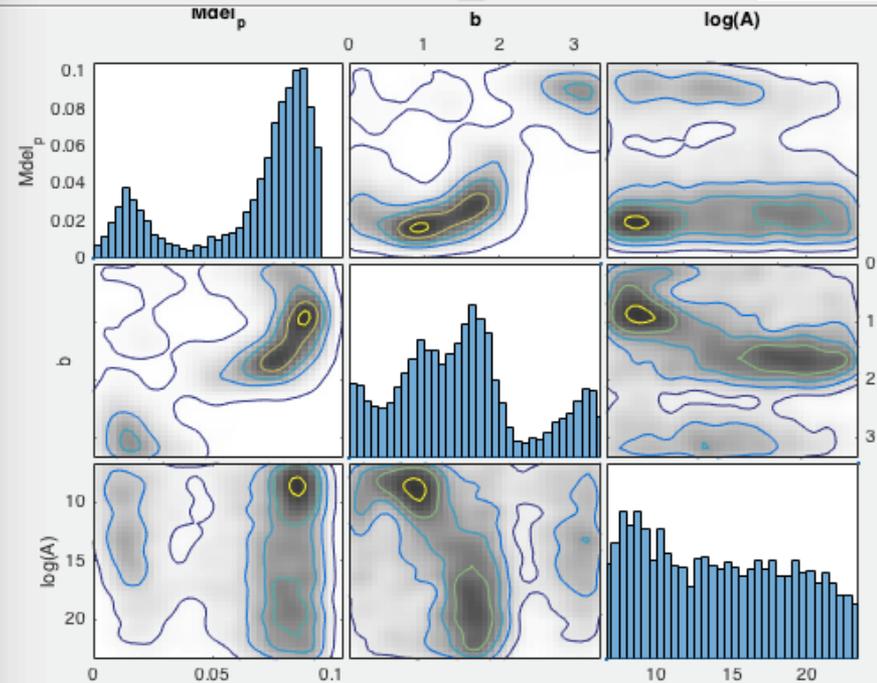
Additional physics (especially more advanced methods to account for heating rate and coal type) greatly improve the model.

Update CCK\oxy
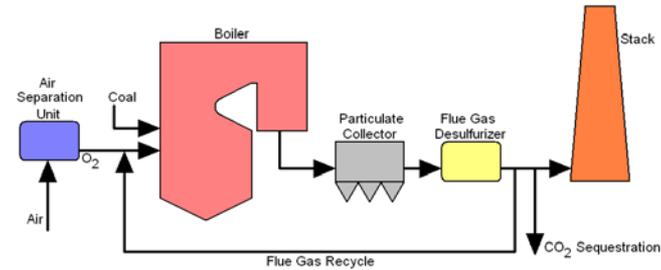
- Add in the annealing code
- Minor updates to other sensitive parameters (swelling, mode of burning, etc.)
- Calibrate kinetic parameters for both gasification and oxidation
  - Hope to make the code coal-general
  - At the very least we will have incremental improvement and quantify the uncertainty

Input:
Coal proximate and ultimate analysis and environmental conditions

Output:
Complete particle temperature and burnout profile, including devolatilization

Applications

- CCK\oxy will predict, in detail, the evolution of coal particle conversion and temperature in time

- A collection of CCK\oxy runs will serve as easily generated data in combustions conditions to train less flexible global models for desktop simulations

Potential form of the global model

$$r_r = -\frac{C1}{C2}\left(\frac{\Phi_{O2}*k_{O2,j}*k_{c,O2,j}*S_{O2,j}*c_{O2,g}}{(k_{O2,j}*S_{O2,j}+k_{c,O2,j})+r_r} + \frac{\Phi_{CO2}*k_{CO2,j}*k_{c,CO2,j}*S_{CO2,j}*c_{CO2,g}}{(k_{CO2,j}*S_{CO2,j}+k_{c,CO2,j})+r_r} + \frac{\Phi_{H2O}*k_{H2O,j}*k_{c,H2O,j}*S_{H2O,j}*c_{H2O,g}}{(k_{H2O,j}*S_{H2O,j}+k_{c,H2O,j})+r_r}\right)$$

Conclusions

- The original annealing model is unable to explain all the data.
- Additional data gives more information about model parameters, but not enough. Additional physics were needed.
- In this case, the activation energy curve should become a function of coal type, heating rate, and (potentially) peak temperature
- The primary advantages of the uncertainty quantification used here are:
  1. The outputs include discrepancy to show where and how physics need to be improved
  2. The outputs are in the form of probability distributions, which is conducive to uncertainty propagation
  3. The method reduces uncertainty to as low as it can be given the data and the model physics (traditional methods often artificially inflate sensitivity)

Disclaimer

Figure references

- http://www.anzbiochar.org/projects.html
- Senneca, Salatino, and Masi; *Energy and Fuels*, 1999
- Hurt, R., Sun, J., and Lunden, M.; A Kinetica Model of Carbon Burnout in Pulverized Coal Combustion; **Combustion and Flame** (1998)