CCSI
Carbon Capture Simulation Initiative

CARBON CAPTURE
MULTIDISCIPLINARY
SIMULATION CENTER

Extension and Calibration of Coal Combustion Models

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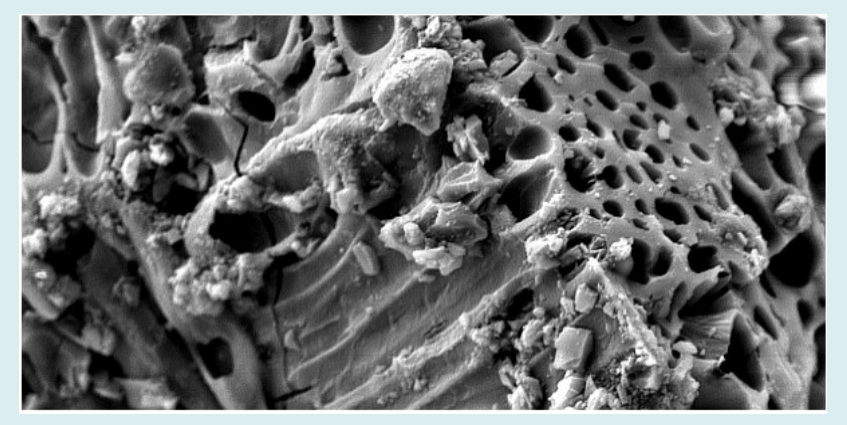
SUMMARY/MOTIVATION:
TWO SYNERGISIC PROGRAMS

- **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

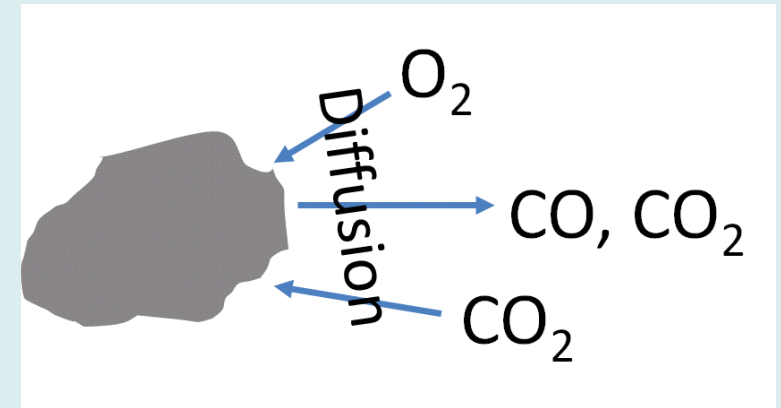
- **CCSI**- Carbon Capture Simulation Initiative
- DoE Office of Fossil Energy
- Primary goal of assisting industry in making carbon capture a feasible reality
- Provides tools for industry friendly (small cluster and desktop) models and simulation based design

BACKGROUND

•Raw coal rapidly devolatilizes in boiler environments to leave behind a coal char. The morphology and reactivity of the coal char depend heavily on devolatilization conditions.



Pyrolyzed char magnified image

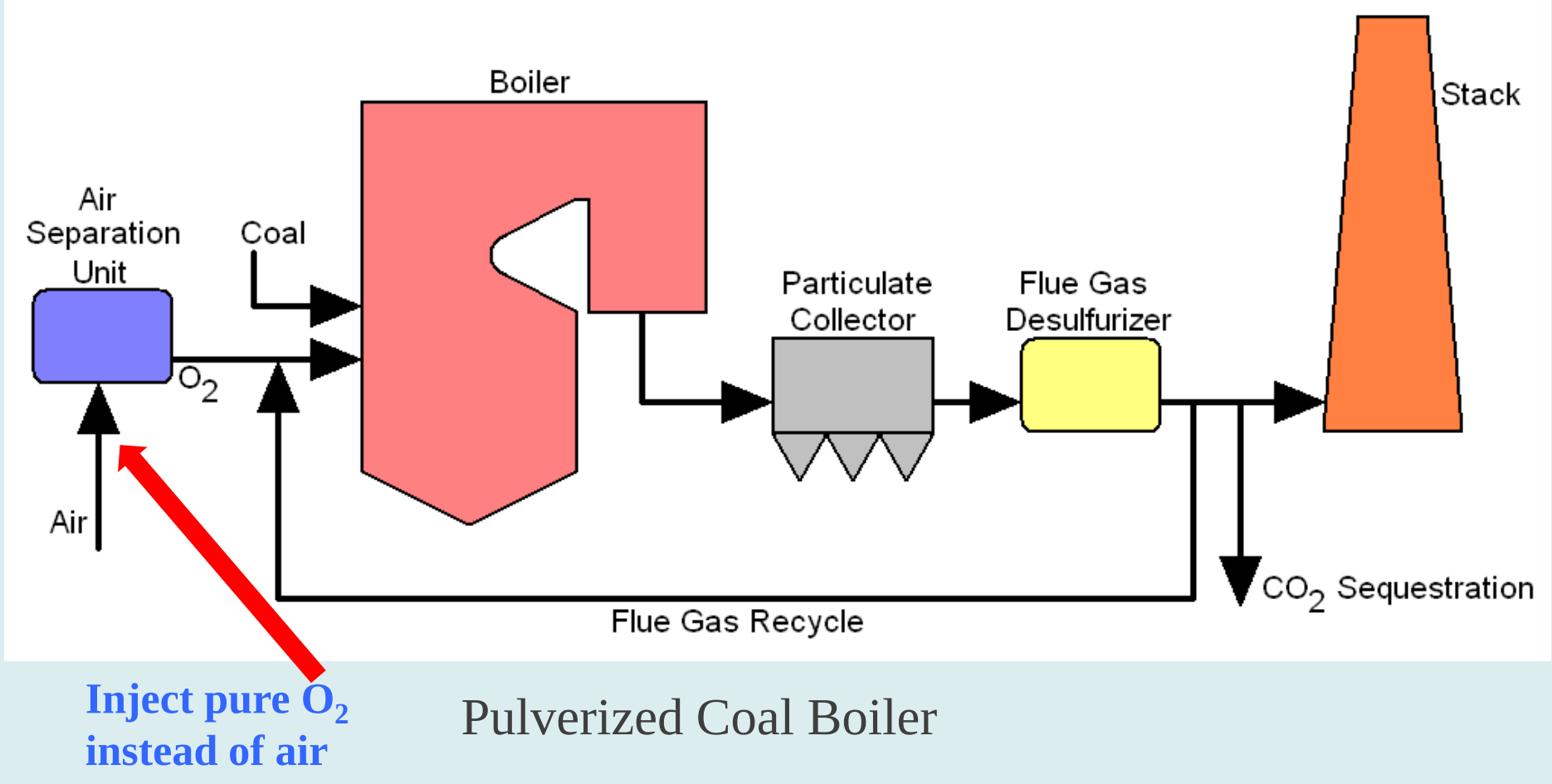


Char Particle Conversion

•Char is converted via exothermic reaction with O_2 and endothermic reaction with H_2O and CO_2, and requires several sub-models to capture mass transfer and kinetics.

•The boiler environment is substantially different from conventional pulverized coal (air-fired), and it requires extended models to capture the extreme environment.

•The char conversion is captured by the CCK\oxy model, the latest iteration in comprehensive char combustion models originating from the CBK model.



Pulverized Coal Boiler

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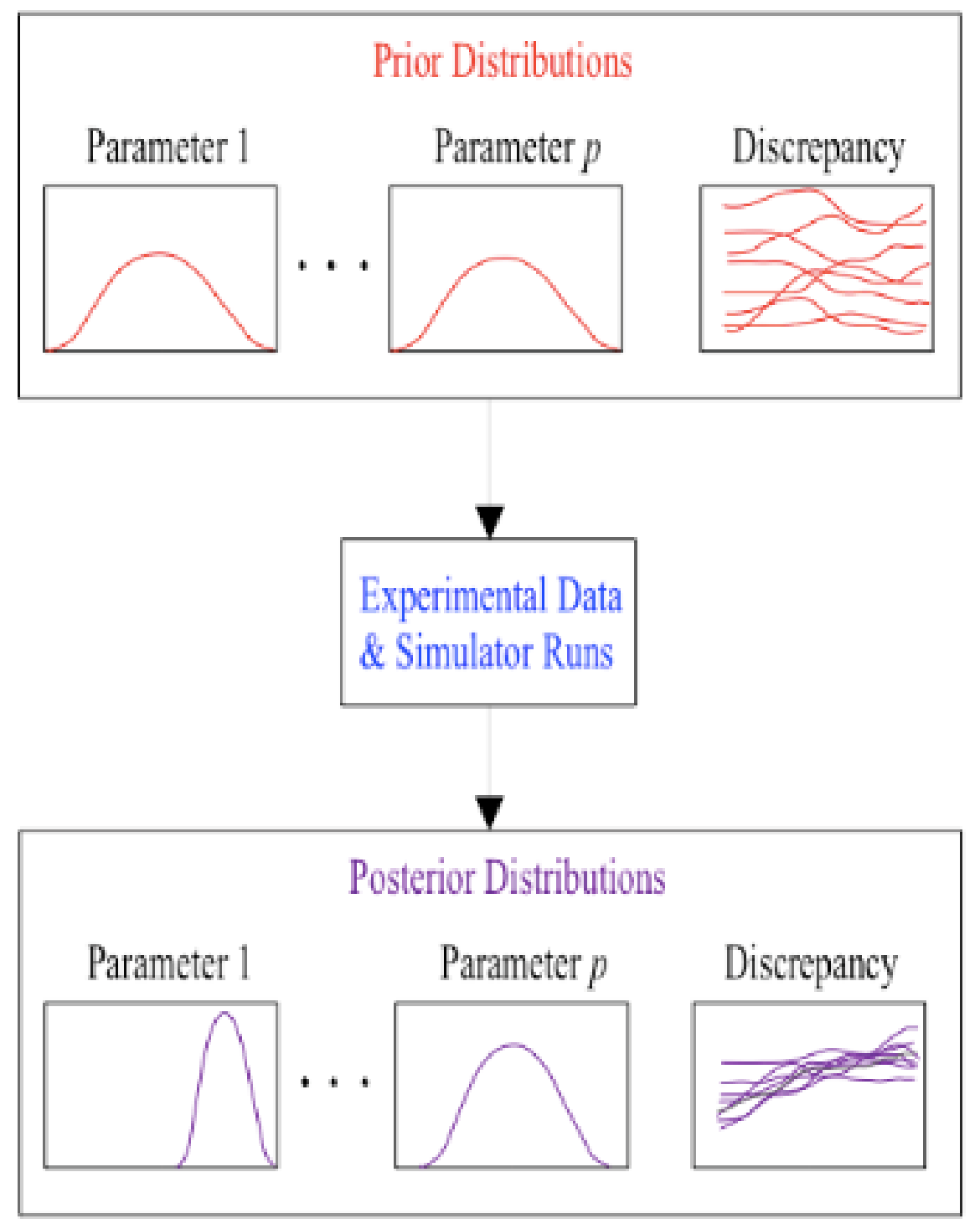
METHODS

- **Sensitivity Analysis**
  - Pick a relevant set of conditions with available data (in this case the high O_2 combustion conditions of Sandia National Labs' entrained flow, flat-flame burner)
  - Catalog the list of variables/parameters input or called within the CCK\oxy code
  - Separate the parameters into "fundamental" parameters and derived parameters
  - Generate a Latin Hyper-cube over all fundamental parameters, varying them throughout their respective parameter space. This allows all parameters to vary simultaneously and crudely capture higher order effects of interacting parameters.
  - Execute the CCK\code using values from the hyper-cube
  - Analyze the results using 3 methods (simple scatter plot, linear approximation, and partial rank correlation coefficients)
- **Calibration and Uncertainty Quantification**
  - Define prior probability density functions for each parameter
  - Generate a Latin Hyper-cube of the entire parameter space
  - Train the statistical emulator
  - Output Gaussian processes for each of η, δ, and ε (the model prediction, discrepancy, and error, respectively)
  - Also output prior probability density functions for each parameter



RESULTS (Sensitivity Analysis)

- The simple scatter plot had far too much noise to detect trends or correlations
- The linear approximation method and partial rank correlation coefficients yielded much more satisfactory results, and consistently highlighted the same seven parameters for all combinations of conditions, coal type, and sensitivity test
- The sensitivity test is essentially a normalized score from 0-1 measuring monotonicity and magnitude of change in output induced by change of input

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

Symbol	Definition
E_A	The mean of the distributed activation energy of the thermal annealing sub-model
N	The order of the oxidation reaction
Ω	The swelling/particle diameter parameter
α	The mode of burning parameter
g_d	The ash grain size
σ	The variance of the distributed activation energy of the thermal annealing sub-model
t_r	The particle residence time

RESULTS (Thermal Annealing Model)

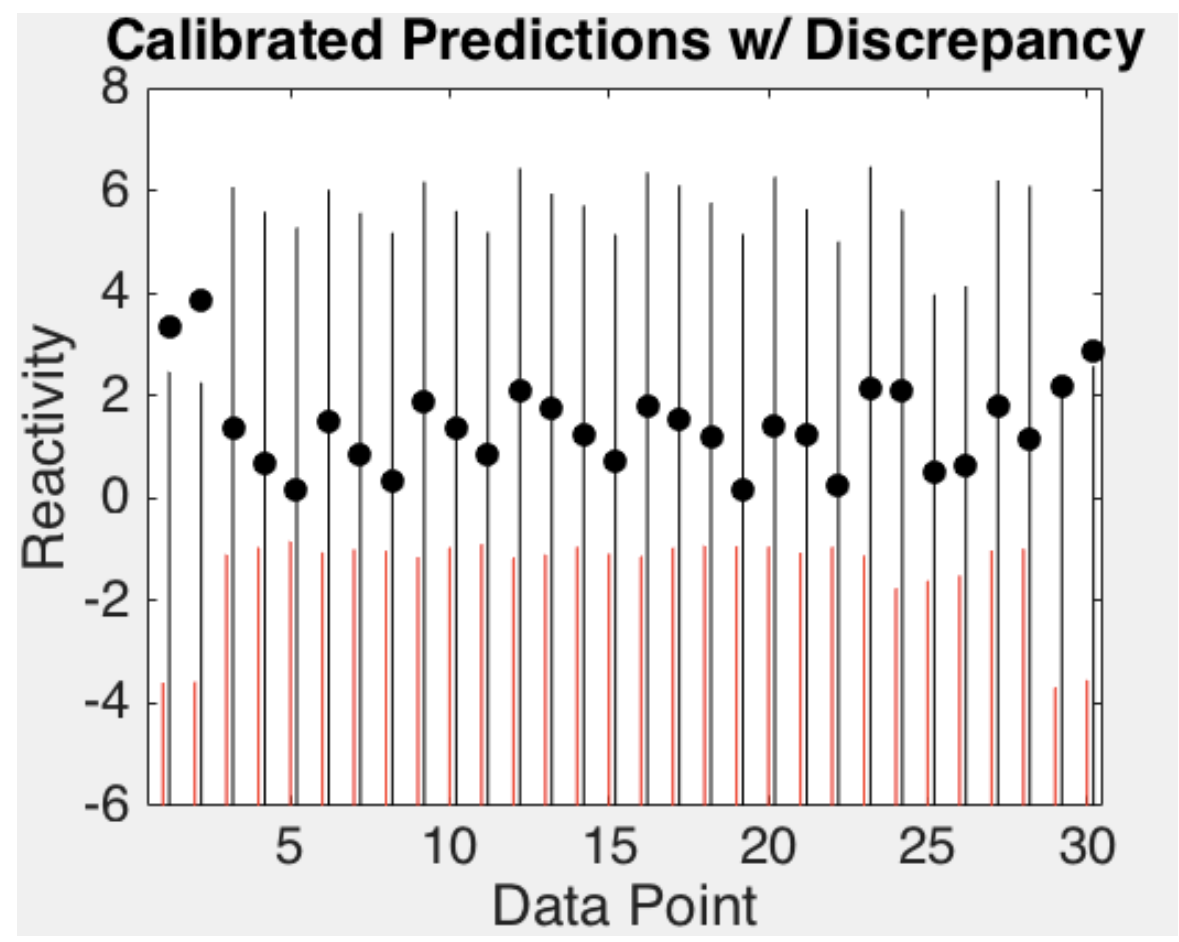
- **Start with the original thermal annealing model and data presented in the CBK model.**

$$y_i = \eta(x_i,\theta) + \delta(x_i) + \varepsilon_i$$

•Red lines: η only

•Black Dots: data points

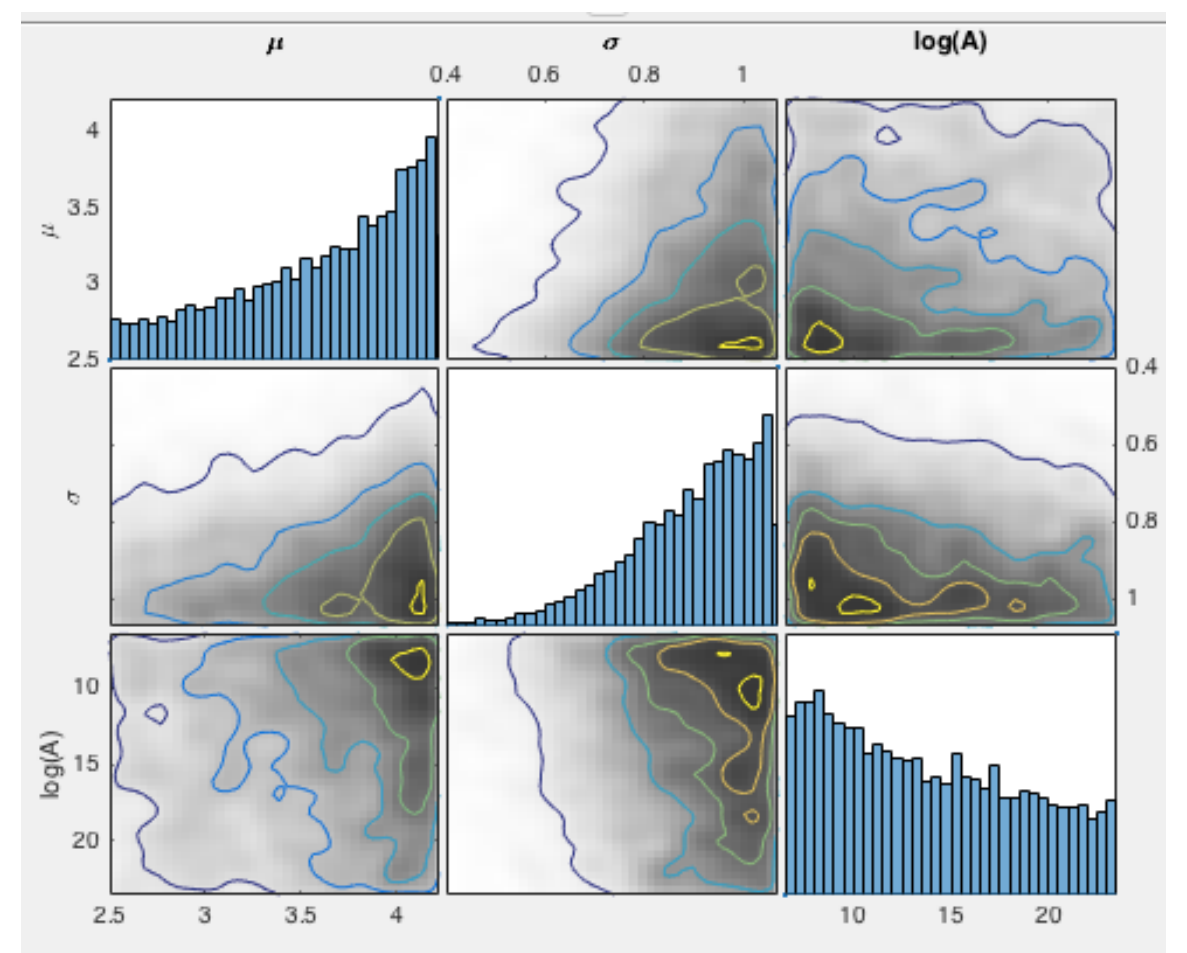
•Black Lines: η+δ+ε (model+discrepancy from reality+error)



•The diagonal shows univariate posterior probability density functions

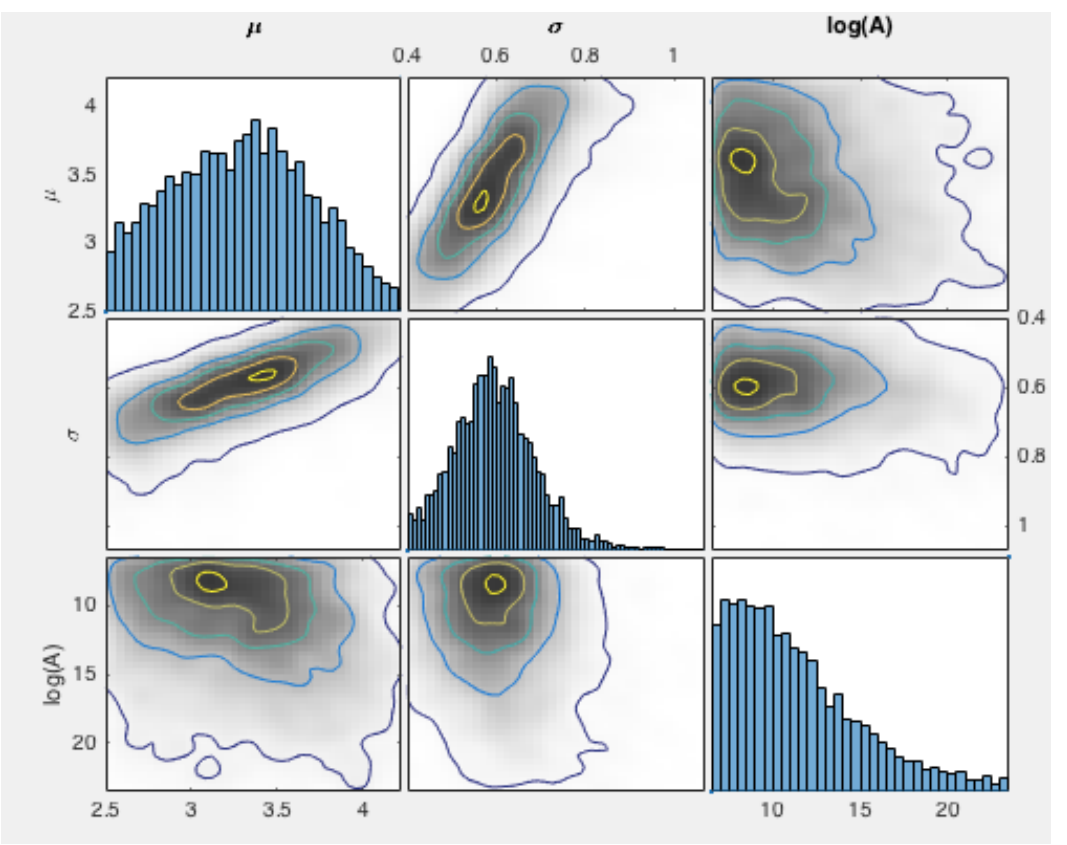
•The off diagonals show the bivariate posteriors

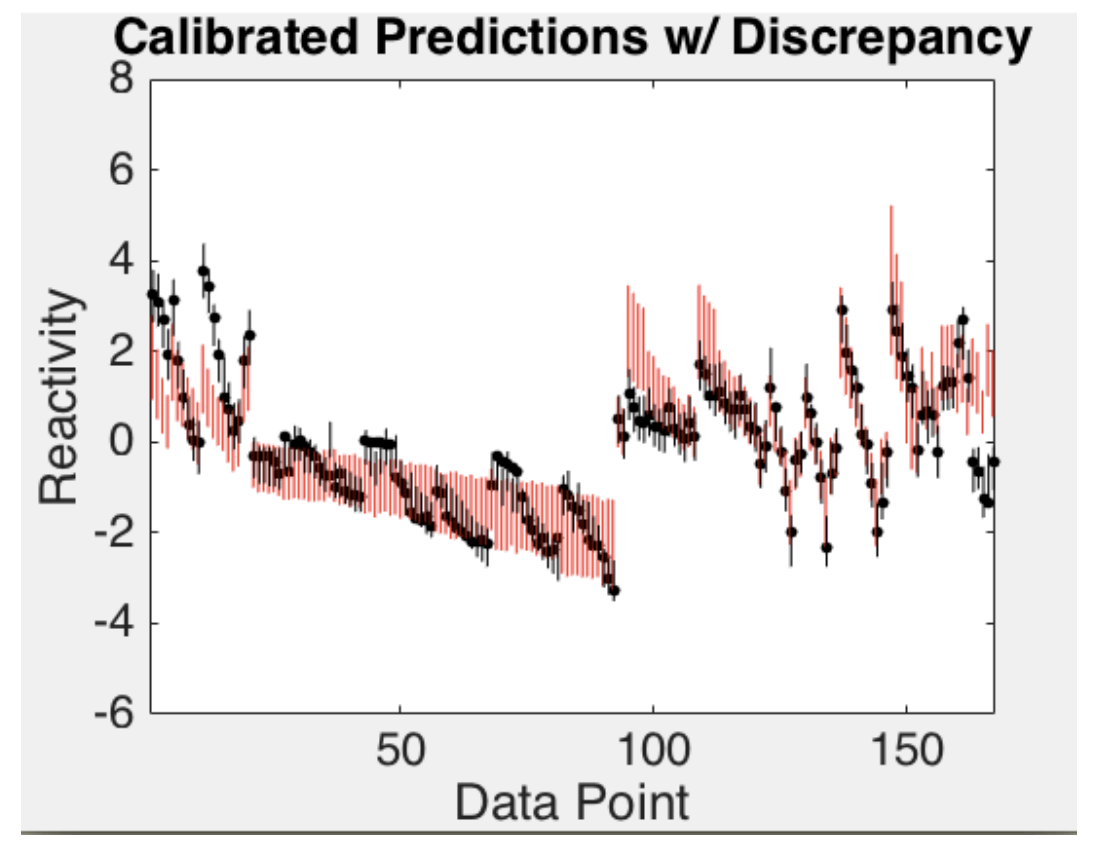
•When probability density accumulates on the edge of the sample space, no good fit is found, implying that the model is unable to capture reality



RESULTS (Thermal Annealing Model)

- **Improve Results**
  - **•Focusing on the most interesting ranges of model operation**
  - **•Transform the input variables to weight the sampling in the sensitive regions of parameter space (improves emulator prediction)**
  - **•Add more data if available**
  - **•Substandtial but inadequate improvement. More physics are needed.**





- **Further Improve Results**
  - **•by incorporating the additional parameters to change the distributed activation energy based on coal type, heating rate, and peak particle temperature.**

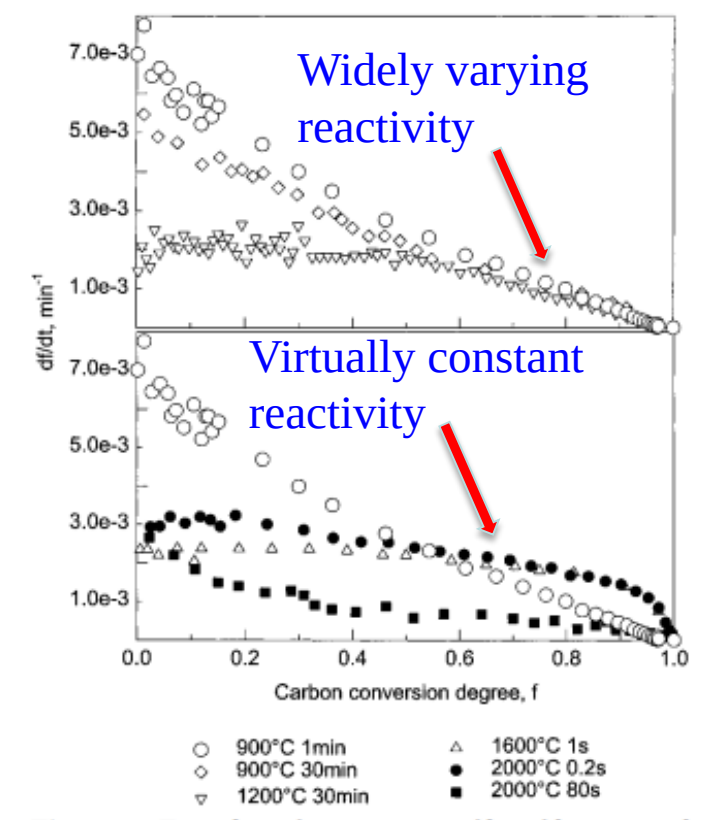
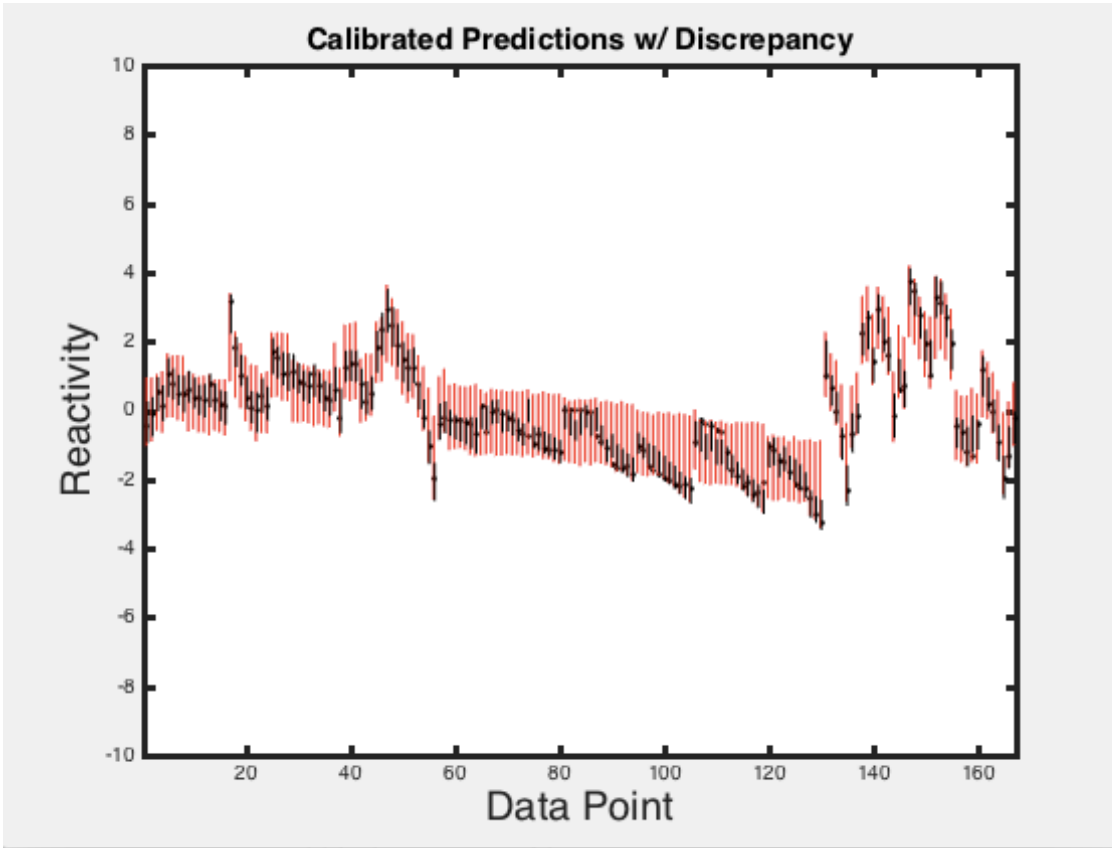
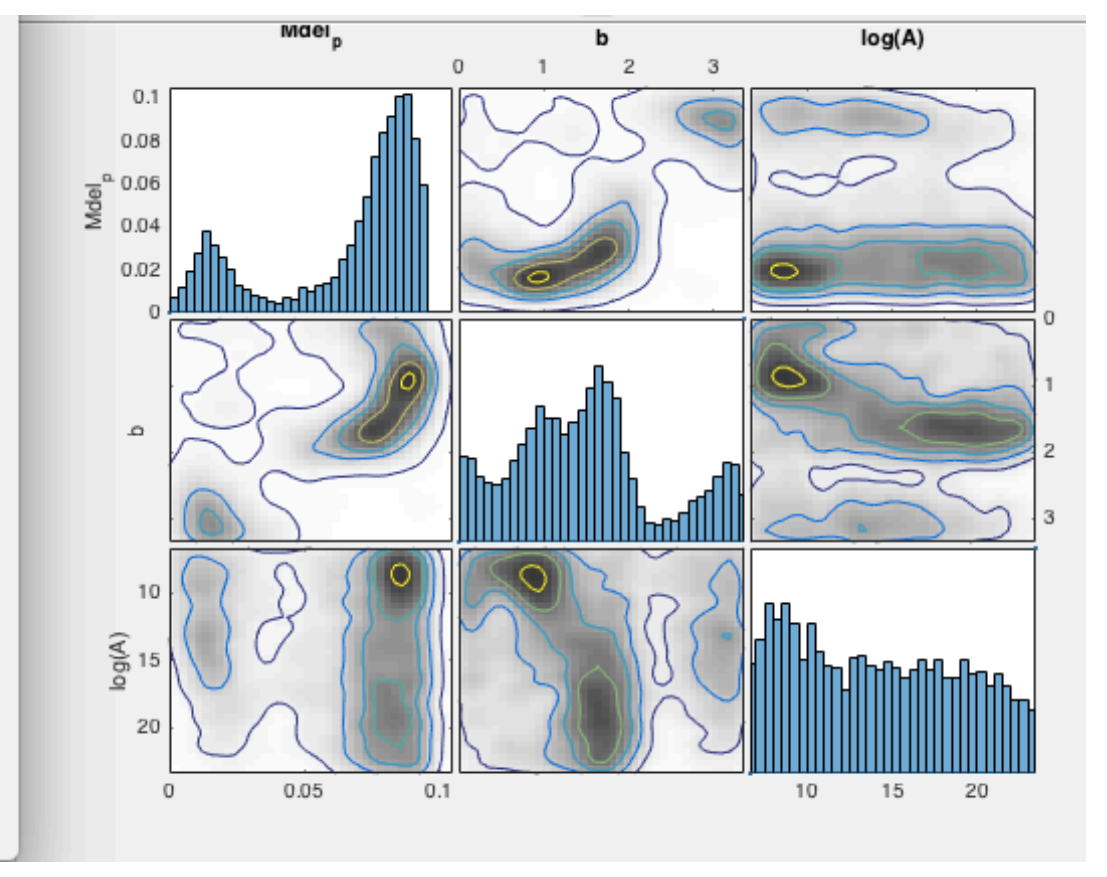


Figure 3. Typical gasification-rate profiles of heat-treated samples.

Sample raw data used in the calibration (from a South African bituminous coal, Senneca et al. 1999)

μ=a*Coal Quantificantion+b a b log(k)





CONCLUSIONS

- The parameters of the thermal annealing sub-model are the most sensitive. It is debatable whether or not this should be the case, but either way the annealing sub-model must be implemented appropriately, either to reduce the impact it has on the model, or to best capture a very sensitive effect.
- The initial annealing sub-model is grossly inadequate to capture the data, and while additional data and careful parameter exploration help, additional physics are/were needed.