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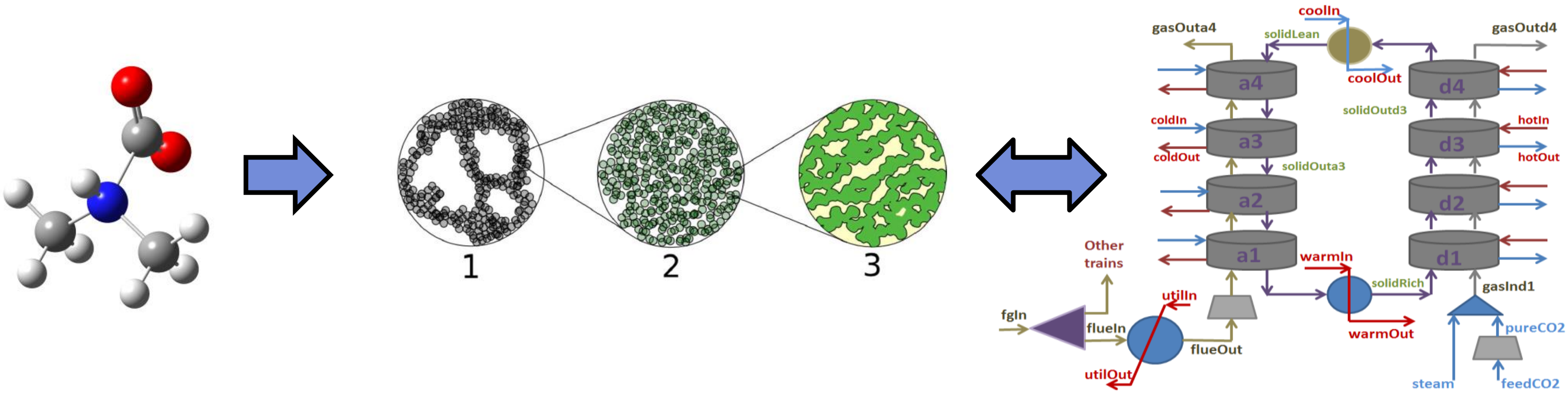
Carbon Capture Simulation Initiative

Multi-scale modeling with generalized dynamic discrepancy

David S. Mebane,[*,§] K. Sham Bhat[¶] and Curtis B. Storlie[¶]
[§]National Energy Technology Laboratory
[*]Department of Mechanical and Aerospace Engineering, West Virginia University
[¶]Statistical Sciences Group, Los Alamos National Laboratory

U.S. DEPARTMENT OF ENERGY

multi-scale modeling



- Multi-scale modeling is an inherently statistical problem.
  - What information will be propagated to the next scale?
  - What is the role of experimental data?
  - How do we account for uncertainty in parameters and models?
  - How do we handicap (or prevent) extrapolation?

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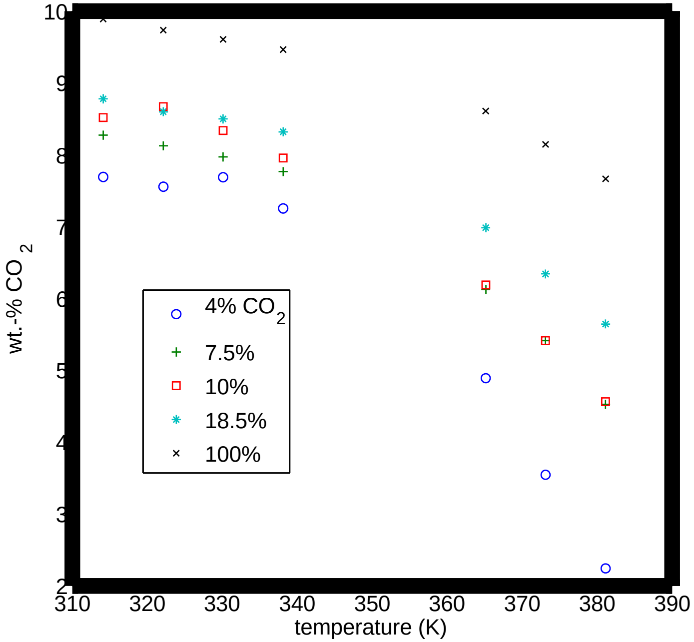
equilibrium problems

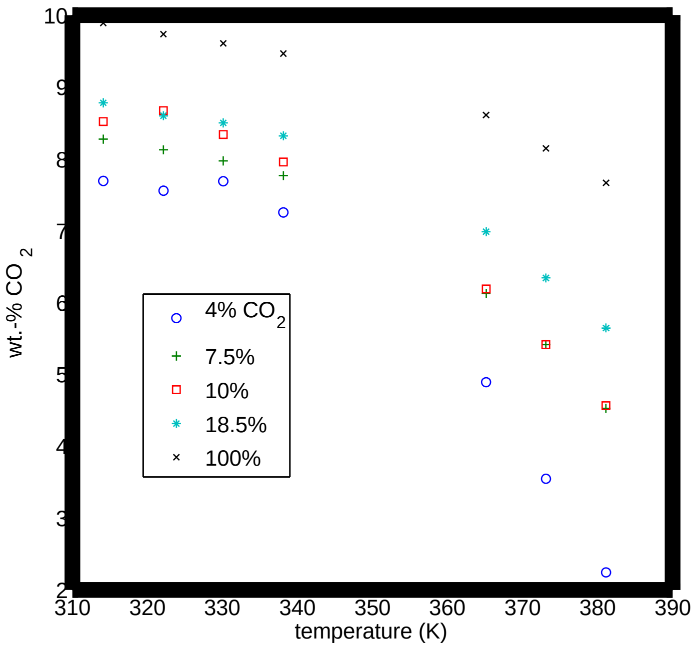
$$f(x, \zeta; \theta) = 0$$
$$Z = \mathfrak{L}(x)$$



equilibrium problems

$$f(x,\zeta;\theta)=0$$

$$\mathcal{Z}=\mathfrak{L}(x)+\delta(\zeta;\xi)+\epsilon(\psi)$$



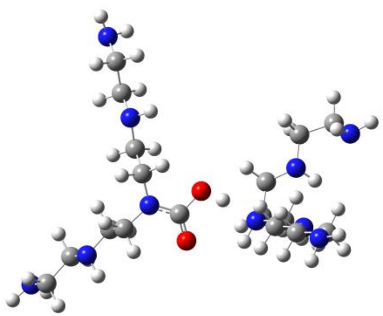
$$\delta\sim MVN(0,\Sigma)\quad \Sigma_{ij}=\sigma^2\exp\left[-\frac{(\zeta_i-\zeta_j)^2}{\phi^2}\right]$$

$$\epsilon\sim MVN(0,\psi I)$$

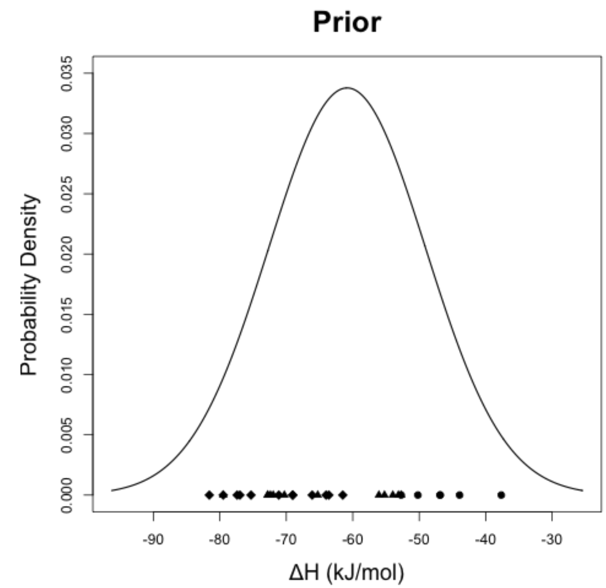
equilibrium problems

$$\kappa = \frac{x^2}{(1-2x)^2 p} = \exp\left(\frac{\Delta S}{R}\right)\exp\left(\frac{-\Delta H}{RT}\right)/P \qquad \theta = \{\Delta H, \Delta S, n_{\mathrm{v}}\}$$

$$z = M n_{\mathrm{v}} x/\rho \qquad \zeta = \{p, T\}$$



rxn	B3LYP	PBE	MP
1	-52.72	-76.36	-62.76
2	-46.86	-70.29	-62.97
3	-50.21	-72.8	-62.76
4	-46.86	-70.71	-64.43
5	-37.66	-69.04	-62.76
6	-43.93	-68.41	-72.38



DS Mebane, KS Bhat, JD Kress, et al., Phys. Chem. Chem. Phys. 15 (2013) 4355.

application in amine-based CO_2 sorbents

$$\kappa = \frac{x^2}{(1-2x)^2 p} = \exp\left(\frac{\Delta S}{R}\right)\exp\left(\frac{-\Delta H}{RT}\right)/P \qquad \theta = \{\Delta H, \Delta S, n_\mathrm{v}\}$$

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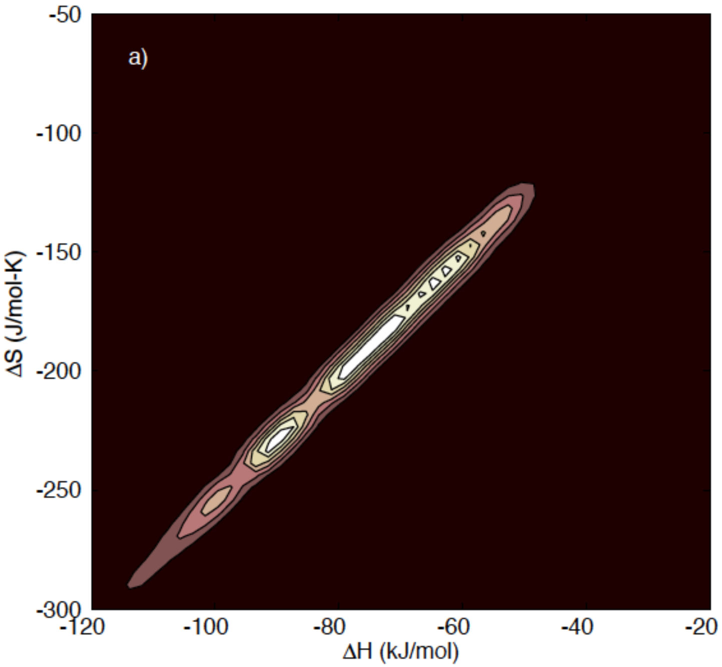
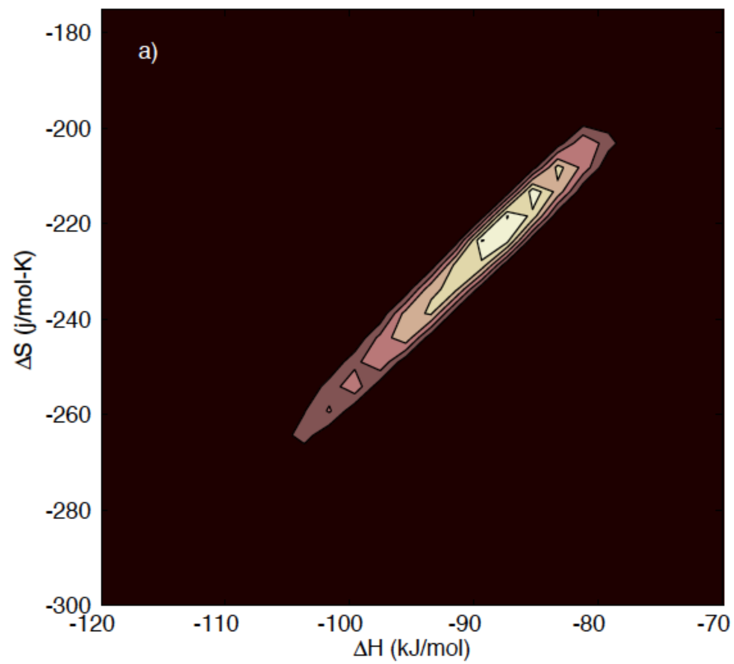


bivariate ΔH-ΔS posterior (left) with and (right) without *ab initio* priors



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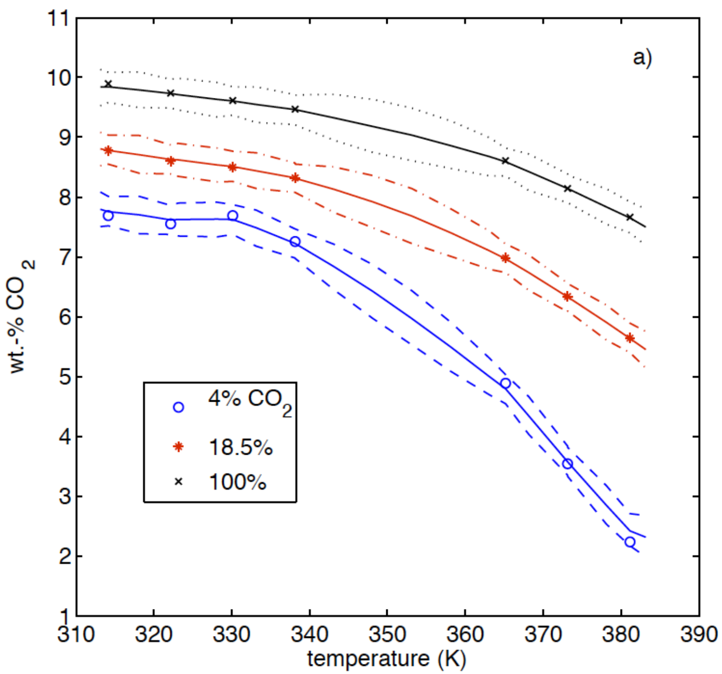
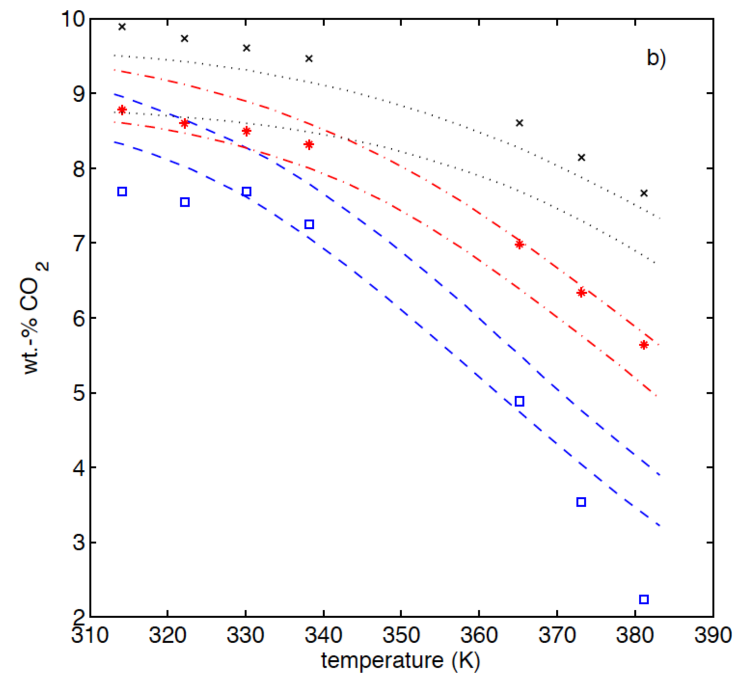


(left) model + discrepancy predictions (right) model-only predictions, with 95% confidence bounds

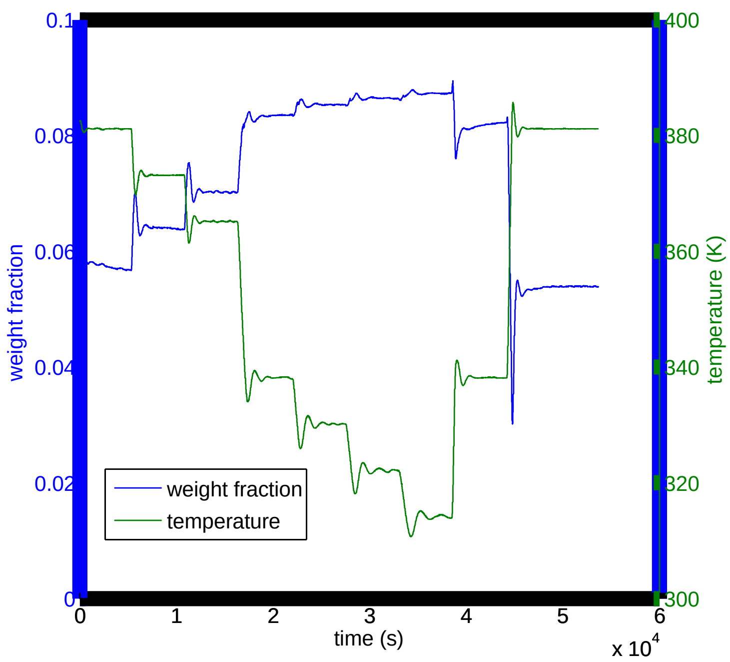


DS Mebane, KS Bhat, JD Kress, et al., Phys. Chem. Chem. Phys. 15 (2013) 4355.

dynamic problems

$$\frac{\partial x}{\partial t} = f(x, \zeta(t); \theta)$$

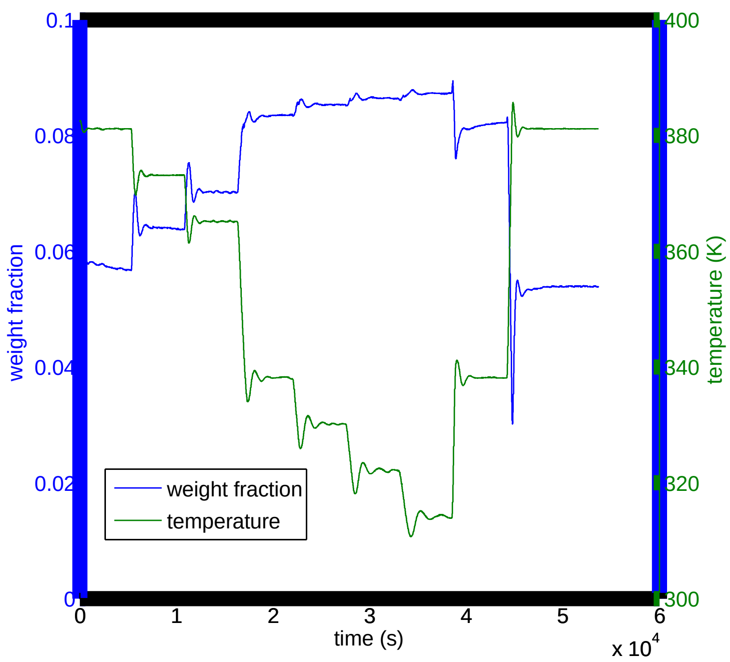
$$Z = \mathfrak{L}(x)$$



dynamic problems

$$\frac{\partial x}{\partial t} = f(x, \zeta(t); \theta)$$

$$\mathcal{Z} = \mathfrak{L}(x) + \delta(t, \zeta(t); \xi) + \epsilon(\psi)$$



$$\delta \sim MVN(0, \Sigma)$$

$$\Sigma_{ij} = \sigma^2 \exp\left[-\frac{(\zeta(t_i) - \zeta(t_j))^2}{\phi_\zeta^2} - \frac{(t_i - t_j)^2}{\phi_t^2} \right]$$

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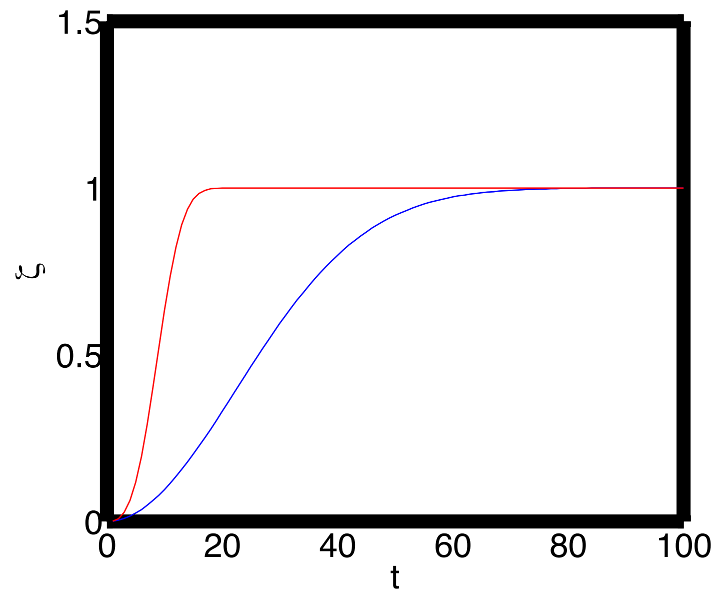

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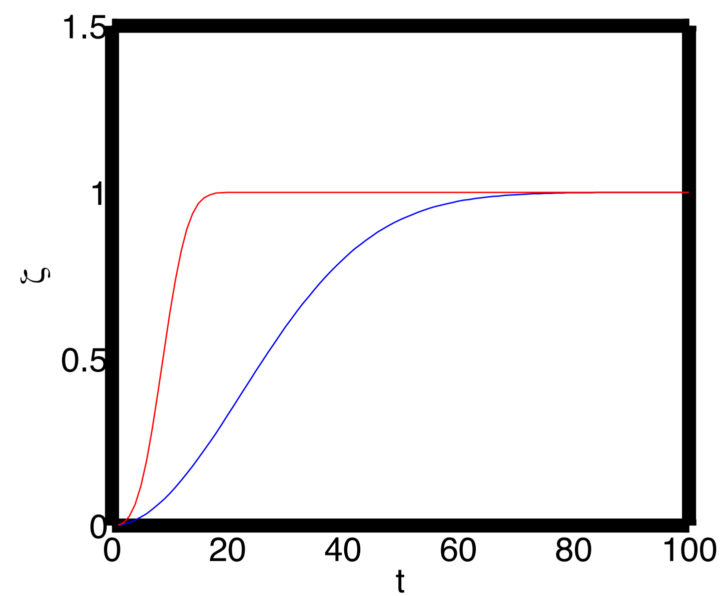
dynamic problems

$$\frac{\partial x}{\partial t} = f(x, \zeta(t); \theta)$$

$$\mathcal{Z} = \mathfrak{L}(x) + \delta(t, \zeta(t), \boldsymbol{\zeta}; \xi) + \epsilon(\psi)$$



$$\delta \sim MVN(0, \Sigma)$$

$$\Sigma_{i_m j_n} = \sigma^2 \exp\left[-\frac{(\zeta_i(t_m) - \zeta_j(t_n))^2}{\phi_\zeta^2} - \frac{(t_m - t_n)^2}{\phi_t^2} - \frac{\|\boldsymbol{\zeta}_i - \boldsymbol{\zeta}_j\|^2}{\boldsymbol{\phi}^2} \right]$$

dynamic problems

$$\frac{\partial x}{\partial t} = f(x, \zeta(t); \theta)$$

$$G(x, \zeta) = 0$$

$$\mathcal{Z} = \mathfrak{L}(x) + \delta(t, \zeta(t), \boldsymbol{\zeta}; \xi) + \epsilon(\psi)$$

$$\delta \sim MVN(0, \Sigma)$$

$$\Sigma_{i_m j_n} = \sigma^2 \exp\left[-\frac{(\zeta_i(t_m) - \zeta_j(t_n))^2}{\phi_\zeta^2} - \frac{(t_m - t_n)^2}{\phi_t^2} - \frac{\|\boldsymbol{\zeta}_i - \boldsymbol{\zeta}_j\|^2}{\boldsymbol{\phi}^2} \right]$$

dynamic discrepancy

$$\frac{\partial x}{\partial t} = f(x, \zeta(t); \theta) + \delta(x, \zeta(t); \beta) \qquad \delta \sim MVN(0, \Gamma)$$

$$\mathcal{Z} = \mathfrak{L}(x) + \epsilon(\psi)$$

$$\Gamma(\vartheta, \vartheta') = \sigma_0^2 + \sum_{k=1}^{K} \sigma_k^2 \Gamma_1(\vartheta_k, \vartheta'_k) + \sum_{k=1}^{K-1} \sum_{l=k}^{K} \sigma_{kl}^2 \Gamma_2([\vartheta_k, \vartheta_l], [\vartheta'_k, \vartheta'_l]) + \cdots$$

$$\vartheta = \{x(t), \zeta(t)\}$$

$$\Gamma_1(\vartheta_k, \vartheta'_k) = \mathcal{B}_1(\vartheta_k)\mathcal{B}_1(\vartheta'_k) + \mathcal{B}_2(\vartheta_k)\mathcal{B}_2(\vartheta'_k) - \frac{1}{4!}\mathcal{B}_4(|\vartheta_k - \vartheta'_k|)$$

$$\Gamma_2([\vartheta_k, \vartheta_l], [\vartheta'_k, \vartheta'_l]) = \Gamma_1(\vartheta_k, \vartheta'_k)\Gamma_1(\vartheta_l, \vartheta'_l)$$

Bayesian smoothing spline analysis of variance (BSS-ANOVA)
BJ Reich, CB Storlie and HD Bondell, Technometrics 51 (2009) 110.

dynamic discrepancy

$$\frac{\partial x}{\partial t} = f(x, \zeta(t); \theta) + \delta(x, \zeta(t); \beta)$$

$$\mathcal{Z} = \mathfrak{L}(x) + \epsilon(\psi)$$

$$\delta(\vartheta; \beta) = \beta_0 + \sum_{k=1}^{K} \sum_{m=1}^{M} \beta_{mk} \varphi_{m1}(\vartheta_k) + \sum_{k=1}^{K-1} \sum_{l=k+1}^{K} \sum_{m=1}^{M} \beta_{mkl} \varphi_{m2}(\vartheta_k, \vartheta_l) + \cdots$$

$$\vartheta = \{x(t), \zeta(t)\}$$

$$\beta_{mk} \sim N(0, \lambda_{m1}\sigma_k^2)$$

$$\beta_{mkl} \sim N(0, \lambda_{m2}\sigma_{kl}^2)$$

- Input domain for the GP drastically reduced.
- Computational complexity of the GP evaluation reduced.
- Integration of the SDE takes place in the course of the MCMC routine.

dynamic discrepancy

$$\frac{dx}{dt} = k_x \left[(1 - 2x - a)a - x^2/\kappa_x \right]$$

$$a = \frac{\kappa_a p (1 - 2x)}{1 + \kappa_a p}$$

$$z = M n_{\mathrm{v}} (x + a)/\rho + \epsilon(\psi)$$

Proof-of-concept with "reality" (left) creating data for calibration of a simpler model with dynamic discrepancy (below).

$$\kappa_x = \kappa_z(T; \theta_r)$$

$$k_x = k_x(T; \theta_r)$$

$$\kappa_a = \kappa_a(T; \theta_r)$$

$$\frac{dx}{dt} = k \left[(1 - 2x)^2 p - x^2/\kappa \right] + \delta(x, p, T; \beta)$$

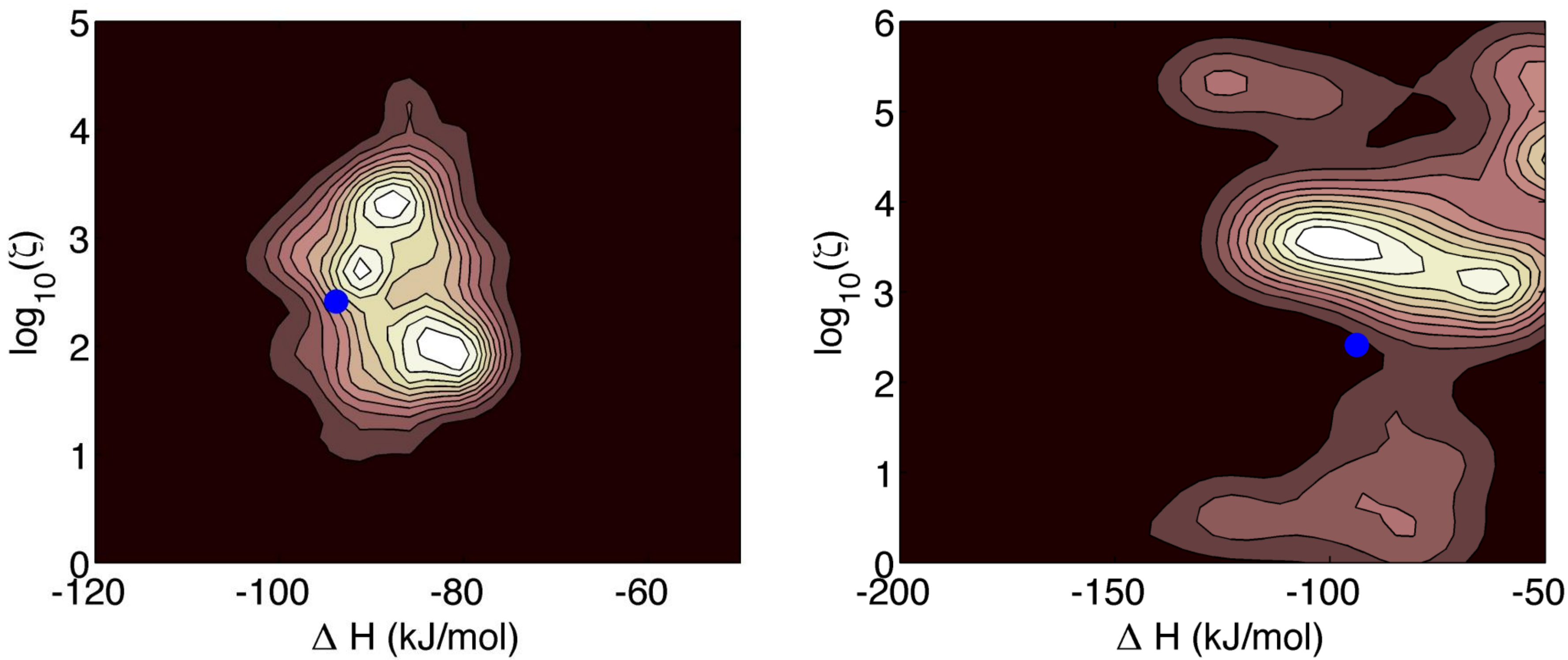
$$z = M n_{\mathrm{v}} x/\rho + \epsilon(\psi)$$

$$\kappa = \kappa(T; \theta)$$

$$k = k(T; \theta)$$

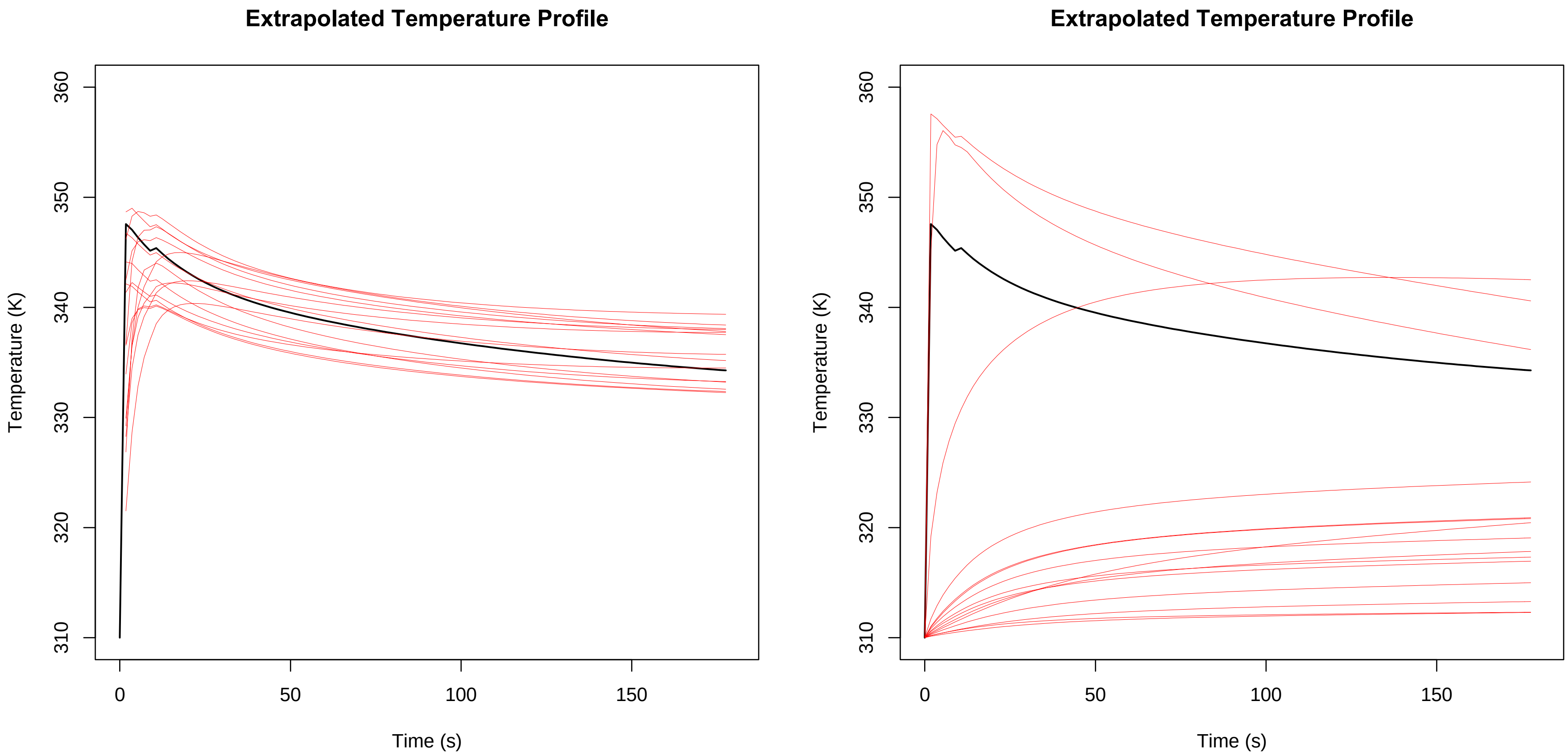
$$\theta = f(\theta_r)$$

dynamic discrepancy



Bivariate posteriors for two of the parameters in t. Left is a case with informative priors, and right with uninformative priors. The blue dot is “reality.”

dynamic discrepancy



Extrapolative predictions in the case of upscaling to a steady-state process model. Left is a case with informative priors, and right with uninformative priors. The black line is “reality.”

dynamic discrepancy

1. Given inputs ζ_k, data Z_k is obtained.
2. The model is calibrated to the data Z_k, yielding a posterior $\Omega_k(\theta, \delta)$.
3. Solution of the resulting model with the steady-state model $G(x, \zeta)$ leads to a distribution of input profiles $\pi_k(\zeta)$.
4. A sample $\zeta_{k+1} \sim \pi_k(\zeta)$ is produced.

Conjecture: **If the dependence of the model is equivalent to the reality and the variation in the discrepancy is sufficient, then the above iteration is convergent in the sense that as $k \to \infty$, extrapolative predictions ζ_k will have mean of reality and variance equal to the variance of the observation error.**

conclusions

- A novel, Bayesian perspective on multi-scale modeling is introduced.
- A new framework for Bayesian calibration in dynamic contexts has been demonstrated.
- The dynamic discrepancy leads to the possibility of an iterative process that can prevent extrapolation.

thanks

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Contact:
David S. Mebane david.mebane@mail.wvu.edu

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