PDF Methods for Uncertainty Quantification

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Sources of Uncertainty in Reactive Transport

Homogeneous & heterogeneous chemical reactions between n species $A_1, A_2, \ldots, A_n$:

$$\alpha_1 A_1 + \alpha_2 A_2 + \ldots + \alpha_m A_m \rightleftharpoons \alpha_{m+1} A_{m+1} + \ldots + \alpha_n A_n$$

Model: Concentrations $c_i(\mathbf{x}, t) \equiv [A_i]$ satisfy a system of ADR eqs.

$$\frac{\partial c_i}{\partial t} + \nabla \cdot (\mathbf{u} c_i) = \nabla \cdot (\mathbf{D}_i \nabla c_i) + F_i(c_1, c_2, ..., c_n), \quad i = 1, \ldots, n$$

Sources of uncertainty:

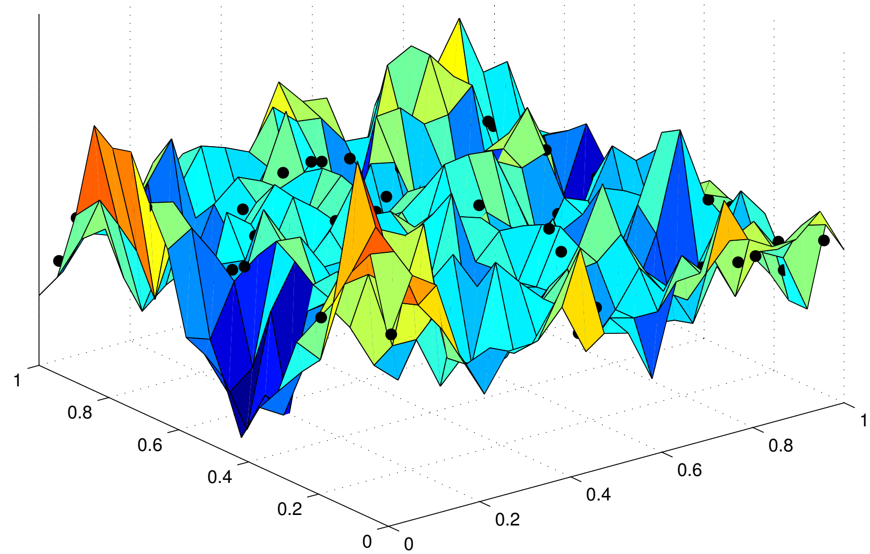
- Flow velocity $\mathbf{u}(\mathbf{x}, t)$
- Reaction rate constants $\kappa_i(\mathbf{x})$

Heterogeneous Chemical Reactions

Transport equation for $\alpha \mathcal{A} \rightleftharpoons \mathcal{A}_{(s)}$:

$$\frac{\partial c}{\partial t} = -\mathbf{u} \cdot \nabla c + \alpha f_\alpha(c), \qquad f_\alpha = -k(c^\alpha - C_{\mathrm{eq}}^\alpha)$$

- Parameter $k(\mathbf{x})$
- Random field $k(\mathbf{x}, \omega)$
- Ergodicity



- Governing equation becomes stochastic
- Solutions are given in terms of PDFs

PDF Methods

- Motivation
  - To avoid the linearization of $f_\alpha(c)$
  - To obtain complete statistics
  - To develop an efficient & accurate tool for UQ
- Raw distribution:

$$\Pi(c, C; \mathbf{x}, t) \equiv \delta[c(\mathbf{x}, t) - C]$$

- Probability density function (PDF):

$$p_c(C; \mathbf{x}, t) = \langle \Pi(c, C; \mathbf{x}, t) \rangle$$

Indelman and Shvidler (1985); Pope (1981)

PDF Equation & Closures

- Stochastic PDE for the raw distribution in $\mathcal{R}^4$: $\tilde{\mathbf{x}} = (x_1, x_2, x_3, C)^T$

$$\frac{\partial \Pi}{\partial t} = -\tilde{\nabla} \cdot (\tilde{\mathbf{u}} \Pi) \qquad \tilde{\mathbf{u}} = (u_1, u_2, u_3, f_\alpha)^T$$

- Deterministic PDE for PDF

$$\frac{\partial p}{\partial t} = -\tilde{\nabla} \cdot (\langle \tilde{\mathbf{u}} \rangle p) - \tilde{\nabla} \cdot \langle \tilde{\mathbf{u}}' \Pi' \rangle$$

- Closure approximations

$$\frac{\partial p}{\partial t} = -\frac{\partial \tilde{u}_i p}{\partial \tilde{x}_i} + \frac{\partial}{\partial \tilde{x}_j} \left[\tilde{D}_{ij} \frac{\partial p}{\partial \tilde{x}_i} \right]$$

DIA (Kraichnan, 1987); LES (Koch and Brady, 1987); Weak approximation (Neuman, 1993); Closure by perturbation (Cushman, 1997)

Reactive Transport in "Homogeneous" Media

Physical homogeneity: $\mathbf{u}$ is deterministic (certain)

Chemical heterogeneity:

$$p_k(k) = \frac{1}{k\sqrt{2\pi\sigma^2}} \exp\left[-\frac{\left(\ln k + \sigma_\kappa^2/2\right)^2}{2\sigma^2}\right]$$

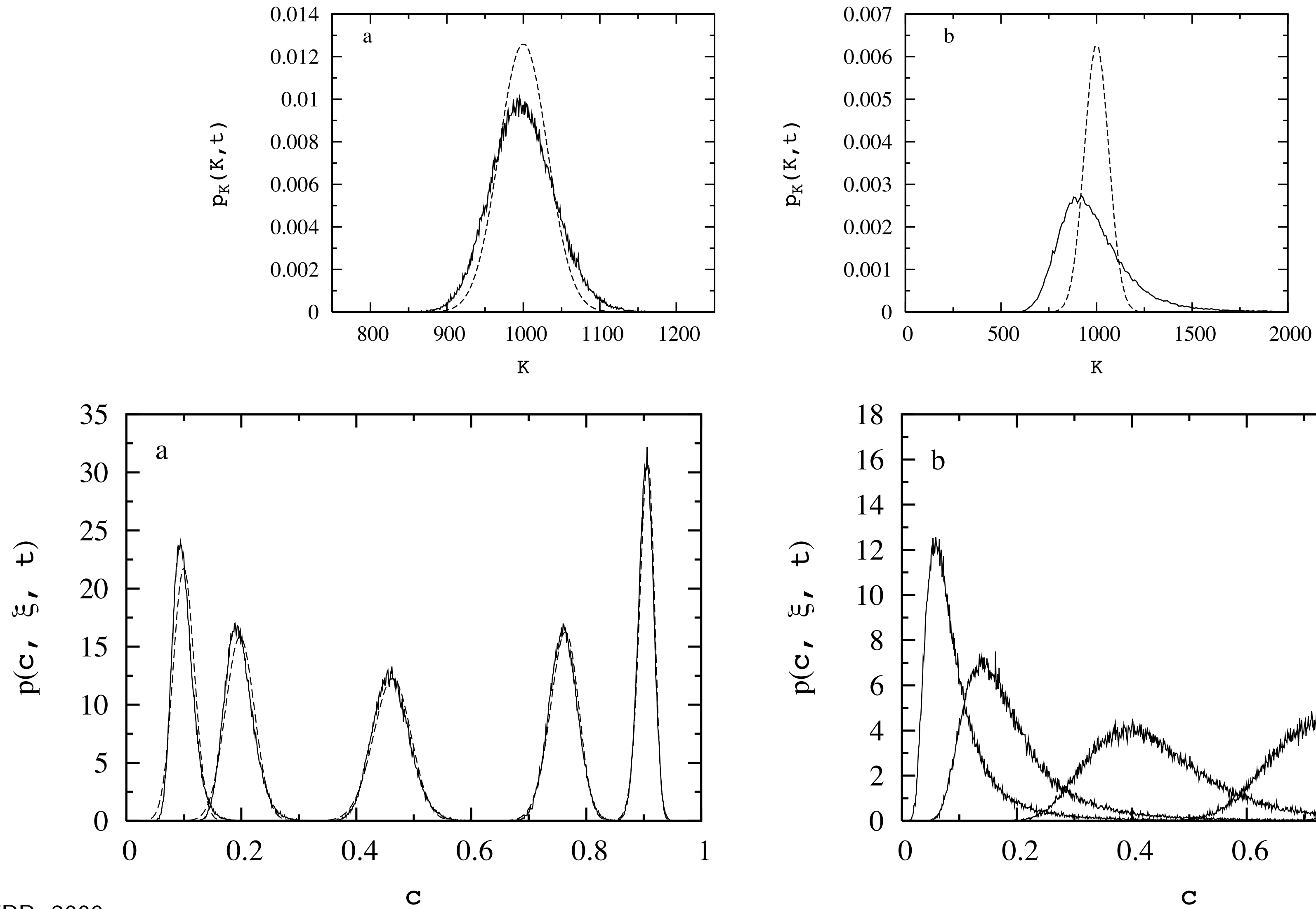
Reynolds decomposition

$$k(x) = \overline{k}\,[1 + \kappa(x)]\,, \qquad \overline{\kappa} = 0, \qquad \overline{\kappa(x)\kappa(x')} = \sigma_\kappa^2 \rho_\kappa(x - x')$$

PDF solution

$$p_c(c; x, t) \sim p_{\mathcal{K}}(\mathcal{K}; t), \qquad \mathcal{K}(t) = \int_0^t k(t')\mathrm{d}t'$$

PDF for Concentration



WRR, 2009

Reactive Transport in Uncertain Velocity Field

Uncertain velocity $\mathbf{u}(\mathbf{x};\omega)$ and reaction rate constant $\kappa(\mathbf{x};\omega)$

- Transport equation for $\alpha\mathcal{A} \rightleftharpoons \mathcal{A}_{(s)}$:

$$\frac{\partial c}{\partial t} = -\mathbf{u} \cdot \nabla c + \alpha f_\alpha(c), \qquad f_\alpha = -k(c^\alpha - C_{\text{eq}}^\alpha)$$

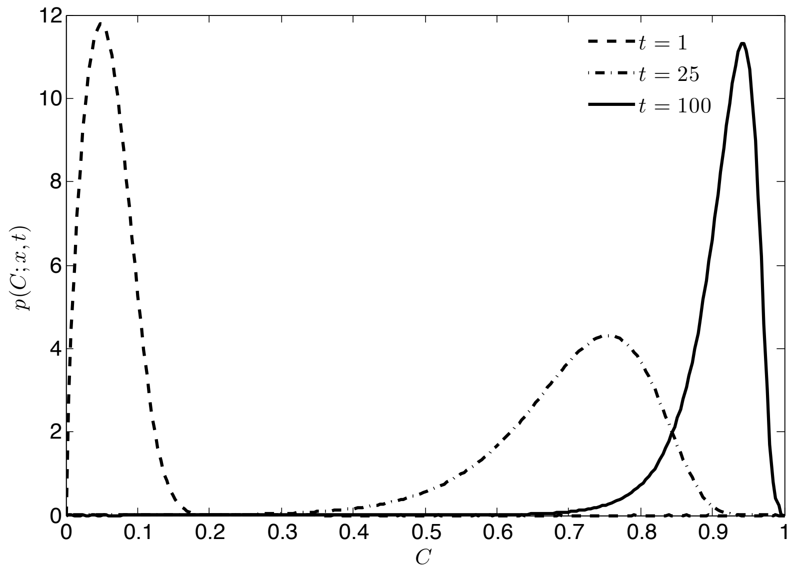
- Stochastic PDE for the raw distribution in $\mathcal{R}^4$: $\tilde{\mathbf{x}} = (x_1, x_2, x_3, C)^T$

$$\frac{\partial \Pi}{\partial t} = -\tilde{\nabla} \cdot (\tilde{\mathbf{u}}\Pi) \qquad \tilde{\mathbf{u}} = (u_1, u_2, u_3, f_\alpha)^T$$

- Deterministic PDE for PDF

$$\frac{\partial p}{\partial t} = -\frac{\partial \tilde{u}_i p}{\partial \tilde{x}_i} + \frac{\partial}{\partial \tilde{x}_j}\left[\tilde{D}_{ij}\frac{\partial p}{\partial \tilde{x}_i}\right]$$

Concentration PDF



	Relative error, $\mathcal{E}$
$t = 250$	0.0008
$t = 500$	0.0011
$t = 750$	0.0023
$t = 1000$	0.0077

Conclusions

- While standard techniques for uncertainty quantification typically yield only concentration's mean and variance, the proposed approach leads to its full probabilistic description.

- The shape of the PDF changes with time, varying between the known initial and steady-state distributions. This makes reliance on assumed PDFs problematic.

This research was supported by DOE ASCR, Appl. Math.