Modeling Field-Scale Uranium Mass Transfer at Hanford IFRC Site

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Quantitative prediction of solute transport in heterogeneous aquifers will remain a grand challenge for the foreseeable future.

Tracer experiments in intensively instrumented field sites have provided indispensable datasets and insights for testing and developing transport models.

The Hanford IFRC project is the most direct and comprehensive attempt at a field test site for understanding and predicting uranium reactive transport.
Challenges at Hanford IFRC Site

- Physical heterogeneity
- Chemical heterogeneity
- Temporal variability
Objectives

- Modeling conservative tracer experiments to identify physical heterogeneity
- Transfer laboratory research results on multi-rate mass transfer processes to the field scale and investigate effects on transport behavior of uranium
- Identify parameters and processes that control the fate and reactive transport of uranium under field-relevant hydrogeochemical conditions
Modeling Br Tracer Experiment

- Use water level data from existing wells near the IFRC plot to interpolate boundary conditions
- Simulate 3D groundwater flow with interpolated time-varying specified-head boundaries
- Calibrate hydraulic conductivities and porosities
Boundary Condition Interpolation

IFRC Plot-Scale MODEL DOMAIN

South Process Pond

- Columbia River Boundary
- Waste Discharge Site
- Interpolation well Location
- IFC Well Location
Hydrogeologic Cross Section
After 17.5 hours
Calibration Results

Well Cluster 1 (shallow)

Well Cluster 1 (middle)
Calibrated Horizontal $K$ Distribution

$K_h (m/d)$
Uranium Reactive Transport

- Generic surface complexation reactions incorporating effects of variably water chemistry:

\[
>\text{SOH} + \text{UO}_2^{2+} + \text{H}_2\text{O} = >\text{SOUO}_2\text{OH} + 2\text{H}^+ \quad K_1 = -4.42
\]

\[
>\text{SOH} + \text{UO}_2^{2+} + \text{CO}_3^{2-} = >\text{SOUO}_2\text{HCO}_3 \quad K_2 = 16.53
\]

Bond et al. (2007), Liu et al. (2008)
Multi-rate Surface Complexation Model

\[
\frac{\partial C_i}{\partial t} + \frac{(1 - \theta) \rho_s}{\theta} \sum_{j=1}^{N_s} \left( \sum_{k=1}^{M_j} a_{ij} \frac{\partial q_j^k}{\partial t} \right) = \nabla \cdot (\mathbf{v} C_i + D \nabla C_i), \quad i = 1, 2, \ldots, N
\]

\[
\frac{\partial q_j^k}{\partial t} = \alpha_j^k (Q_j^k - q_j^k), \quad j = 1, 2, \ldots, N_s; \; k = 1, 2, \ldots, M_j
\]

Distributed rate constants assumed to follow lognormal probability distribution

\[
p(\alpha) = \frac{1}{\sqrt{2\pi\alpha\sigma}} \exp \left( -\frac{1}{2\sigma^2} (\ln(\alpha) - \mu)^2 \right)
\]
U(VI) Concentrations at Column Outlet

- Measured [Liu at al., 2008]
- Modeled [Liu at al., 2008]
- Modeled [PHT3D]
2D Flow Model Setup

Location of cross section

Horizontal Distance (m)

U source

Hanford Gravel/sand

Ringold Fine-grained Unit

River
Reactive Transport Model Setup

- 10 aqueous master species and 1 mineral (calcite)
- Aqueous complexation, redox-, mineral-, and surface complexation reactions were modeled
- Western BC: groundwater composition
- Eastern BC: river water composition
- U(VI) bearing solution \( (5 \times 10^{-6} \text{ mol/l}) \) introduced as a point source located 147 m from the river.
- Initial U concentration of groundwater and river water is zero.
- Simulated with PHT3D which couples MT3DMS and PHREEQC-2.
Comparison with Field Data

**specific conductance**

**carbonate**

**calcium**
Comparison of Model Results

Multi-Rate SCM

Equilibrium SCM

Diagram showing the comparison over different time periods (150 days, 50 days, 1 year, 5 years, 20 years) for Multirate SCM and Equilibrium SCM.
Uranium Plume Animation

Equilibrium SCM

Time = 0.04167 (1-1-1)  days of 20 years (7320 days)

Multi-Rate SCM

Time = 0.04167 (1-1-1)  days of 20 years (7320 days)
Uranium Mass Balance

AdSORBED U(VI)  Mobile

Multi-Rate SCM

Discharge To River

Elasped time (year)

Year
Parameter Sensitivities for Multi-Rate SCM

Major Questions:

- Does the importance of parameters/processes change between laboratory- and field-scale as a result of the different characteristics of hydraulic and geochemical conditions and variability. If so, why?

- Can chemical model complexity be reduced for field scale conditions?

Approach:

- Comparing parameter sensitivities for models that are based on the conceptual model of Liu et al. [2008] for characteristic lab and field scale conditions
Field Scale - 1D Approximation

Field scale hydraulic parameters:
- Hourly time-variant flow boundary conditions (1 year)
- Calibrated hydraulic conductivities
Parameter Sensitivities: Column Experiment

- **U(VI) concentration**
- **U(VI) flux**
- **Cumulative U(VI) flux**
- **U(VI) total mass in aqueous phase (mobile domain)**

The graphs show the sensitivity of various parameters to the concentration, flux, cumulative flux, and total mass of U(VI) in the aqueous phase. The parameters include:

- $n_{in}$
- $a$
- $b$
- $n_{bat}$
- $a_{L}$
- $a_{R}$
- $b$
- $K_{1}$
- $K_{2}$
- $S_{bulk}$
- $U_{tot}$
- $S_{m}/S_{m}$
- $M = 25$
- $M = 10$
Parameter Sensitivities: Column Experiment

Most important:
- Total uranium
- Sorption site density
- Darcy flux
- Reaction constants

Decrease from 50 to 10 sorption domains drastically affect the simulation results.
Parameter Sensitivities: Field Scenario

Most important:
Same parameters as for column, but more pronounced

Effect of a decrease in sorption domains not as strong as for column
Parameter Sensitivities: Field Scenario

U(VI) concentration

U(VI) flux

Cumulative U(VI) flux

U(VI) total mass in aqueous phase (mobile domain)

Mobile U mass (aqueous phase): Insensitive with respect to several parameters (Contrasting lab-scale behaviour)
Somewhat unexpectedly, parameter sensitivities were mostly similar for lab and field scale models.

Total uranium and total sorption site density are the most important parameters.

For synthetic groundwater (Liu et al., 2008): the 1st sorption constant $K_1 (>SOUO_2OH)$ is more sensitive than the 2nd sorption constant $K_2 (>SOUO_2HCO_3)$.

For average measured groundwater composition (Ma et al., in review): $K_2$ is more sensitive than $K_1$.

At the field scale many parameters are insensitive with respect to the total mass in the mobile aqueous domain (As the plume remains within the model domain over the whole simulation period).
Summary

- Column River water can encroach inland as far as 330 m through Hanford Formation at high river stage.
- U plume in multi-rate SCM is much more dynamic and consistent with field observation of a very dynamic U plume than that in equilibrium SCM.
- U breakthrough curves and mass balance indicate that U adsorption/desorption never attains equilibrium due to highly dynamic flow field and chemistry variations caused by intrusion of river water.
- Parameter sensitivities are similar for both lab and field scales; total U and total sorption site density are the most important parameters.
- At the field scale many parameters are insensitive with respect to the total mass in the mobile aqueous domain.
Thank you!