
Hanford 300 Area

Coupled Process Modeling of the IFRC Site: Approaches, Insights, and Future Plans

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Bromide and Heat Tracer Transport Modeling

Experimental results:

In March 2009, a solution chilled to a temperature of 9.5°C and containing bromide at a concentration of ~100 mg/L was injected through the well into the aquifer for 567 minutes with a flow rate of 16.3 m^3/hr.

- The tracer response in the northern multi-level well cluster shows that tracer arrival significantly lagged in the middle zone (blue), and the earliest arrival occurs in the deeper zone (red), closely followed by that in the shallow zone (green), consistent with EBF hydraulic test results (Figure 1, below left).

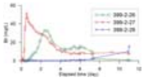


Figure 1. Br breakthrough curves at a well cluster.

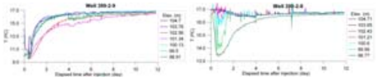


Figure 2. Temperature breakthrough curves at different depths at two wells.

- Temperature data at different depths of any observation well exhibit a significant variability, reflecting the important role of aquifer heterogeneity in the vertical direction (Figure 2, above right).

Transport modeling:

- MODFLOW and MT3DMS were used to simulate the tracer experiments; PEST was used to help calibrate the model.
- The model domain was discretized by 60 columns, 61 rows and 27 layers, covering a domain of 120 m length ×122 m width × 22.5 m depth (Figure 3, to the right).
- The western, eastern, northern and southern sides of the grid were defined as time-varying specified-head boundaries interpolated with water levels from the wells shown in Figure 3. The bottom of the grid was represented as a no-flow boundary, and the uppermost layer also treated as no-flow.
- The measured data were used as the initial input for the model and the hydraulic conductivity, porosity and dispersivity were the main calibration parameters.

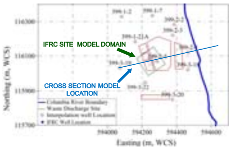


Figure 3. Location of 3D IFRC site model domain and cross section model.

Insights gained from modeling:

- The groundwater flow field was highly dynamic with significant variations in both directions and magnitudes of flow vectors (Figure 4).
- Groundwater head data alone were insufficient to constrain the model calibration because of the small gradients persisting in the highly permeable Hanford Formation at the Hanford IFRC site.
- The tracer plume movement was highly sensitive to the transient nature of the flow field induced by the Columbia River. Great care had to be taken to ensure the boundary conditions were properly constructed for successful analysis and understanding of the tracer tests. Figure 5 shows the comparison between observed and simulated Br concentrations at four observation wells.
- Significant vertical flow gradients occurred in the aquifer induced by the aquifer heterogeneity and highly dynamic fluctuations of the Columbia River. This in turn caused significant intra-borehole vertical flow with alternating upward and downward movements in the long screened observation wells, impacting the representativeness of the water samples from these wells (Figure 6).
- Temperature data served as a cost-effective proxy for conservative solute tracers that can underpin an improved calibration of the 3D flow and transport model to further resolve the hydrogeological heterogeneity of the Hanford IFRC site.

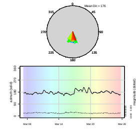


Figure 4. Gradient directions and magnitudes, over time, determined from a 3-well calculation.

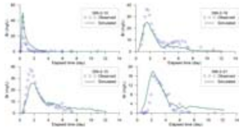
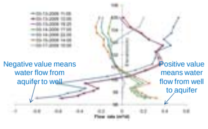


Figure 5. Comparison between observed and simulated Br concentrations at four observation wells.


Negative value means water flow from aquifer to well

Positive value means water flow from well to aquifer

Figure 6. Flows in the well 399-2-7 calculated with MODFLOW Multi-node-well Package.

Background

Quantitative description and prediction of contaminant transport in physically and chemically heterogeneous aquifers remain one of the greatest challenges in subsurface sciences. As an integral part of the Hanford IFRC project, we have developed a comprehensive modeling research program to assist the planning for and interpret the results from the field research program. In an iterative and complementary way, field experiments and modeling activities work together to enable us to gain new insights on the fate and transport of uranium contaminants at the field scale and to improve our predictive capabilities required for uranium remediation and risk assessment.

The modeling at the Hanford IFRC site has proceeded on a multi-tiered approach to cope with the field experiments of increasing complexity and an increasing number of coupled processes. The work involves close collaboration among UA, PNNL and CSIRO scientists.

2-D Field Scale Reactive Transport Modeling

A 2-D field-scale reactive transport model was constructed with PHT3D based on MT3DMS/PHREEQC-2 (www.pht3d.org). The model accommodates the combined effects of physical and chemical heterogeneities by incorporating laboratory-characterized U(VI) surface complexation reactions (SCR) together with dual-domain, multi-rate mass transfer processes. It incorporates the key characteristics of the field-measured hydrogeochemical conditions (The location of model is shown in Figure 3). The field-scale model was used

- to assess the importance of multi-rate mass transfer processes on U(VI) reactive transport under hypothetical scenarios; and
- to evaluate the effect of variable geochemical conditions caused by dynamic river water-groundwater interactions on U(VI) migration.

Assessment of importance of multi-rate mass transfer processes:

- In general, river water intrusion enhances U(VI) adsorption.
- U(VI) breakthrough curves and mass balance indicate that U(VI) adsorption/desorption never attains equilibrium due to variable flow field and chemistry caused by intrusion of river water. The multi-rate SCR model appears to be a crucial consideration for future reactive transport simulations of uranium at the IFRC site and elsewhere under similar hydrogeochemical conditions.
- Compared to ESCM, MRSCM results demonstrate that the coupling of multi-rate mass transfer processes and surface complexation reactions lead to (Figure 7):
  - ✓ a high concentration zone of the plume that was more dynamic and responsive to the flow field;
  - ✓ more rapid growth of the U(VI) plume;
  - ✓ eventual loss of U(VI) mass from groundwater to the river.

The results of the MRSCM appeared more consistent with previous field observations of plume dynamics at the Hanford 300A site.

Multi-Rate SCM

Equilibrium-SCM

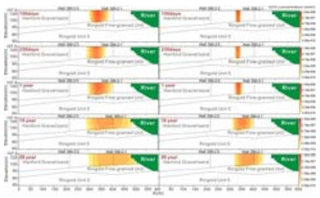


Figure 7. Comparison of uranium plumes simulated with Multi-Rate SCM model (left) and Equilibrium-SCM model (right) at different times

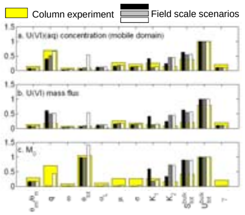


Figure 8. Laboratory scale and field scale composite sensitivities.

Sensitivity analysis:

A rigorous sensitivity analysis was carried out to identify and compare the individual processes that control the fate of U(VI) at both the laboratory and field scales, and to identify the mechanisms that affect the importance of these processes at different scales. Our work to date has shown (Figure 8):

- Parameter sensitivities were overall similar for lab- and field-scale models.
- Total uranium and total sorption site density are the most important parameters.
- Some deviations in sensitivities between lab- and field-scale models largely result from the different extent of disequilibrium conditions depending on the advection regimes.
- Field scale sensitivities are higher in the plume fringe where the degree of sorption disequilibrium is higher.

3-D Uranyl Reactive Transport Modeling at the IFRC Site

Laboratory column experiments were performed using three intact core sediments from the IFRC site to calibrate a model for reactive transport of U(VI) (Figure 9).

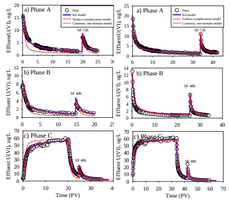


Figure 9. Effluent U(VI) concentrations during U(VI) desorption from the long-term contaminated sediments (phases A and B) and short-term U(VI) adsorption and desorption (phase C). Left plot for ICE1 and right for ICE2.

A uranyl reactive transport model (STOMP) for the IFRC site that couples uranyl geochemistry and mass transfer kinetics has been developed based on laboratory core sediment results (Figure 9). The model has been used to provide insights into the observations during the field uranium desorption experiment (Figure 10). The model will incorporate the heterogeneity in flow, initial U(VI) concentration, grain-size distribution, and adsorption site density at the IFRC site to:

- understand the behavior of uranyl geochemical and mass transfer processes under natural and perturbated physical and chemical conditions;
- use such an understanding to plan future field experiments to evaluate physical and chemical processes controlling uranyl reactive transport;
- develop a site-wide U(VI) reactive transport model for data interpretation, predictive projection, and extrapolation of the IFRC site results to other Hanford and DOE sites.

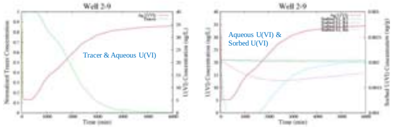


Figure 10. An example of simulated concentrations of tracer, aqueous and sorbed U(VI) during U(VI) desorption experiment at the IFRC site.

Further Research

- To explore how the complex interactions among flow, transport and geochemical processes impact and ultimately determine the uranium fate and transport at the Hanford IFRC site.
- To investigate how the variation in river water chemistry with time and the calcite mineral dissolution/precipitation (equilibrium or kinetically) in the groundwater/river water mixing zone influences U(VI) adsorption/desorption behavior at the Hanford IFRC site frequently subject to Columbia River intrusion.
- To evaluate and compare two alternative approaches that both simulate coupled diffusional mass transfer and (non-linear) surface complexation processes: (1) the chemical non-equilibrium approach, which approximates the bulk process as distributed rate surface complexation kinetics, and (2) the physical non-equilibrium approach, in which the bulk kinetic processes result from a combination of diffusional mass-transfer and instantaneous surface complexation reactions in the immobile domains.

Conclusions

- The groundwater flow field at the Hanford IFRC site is highly dynamic and the groundwater head data alone were insufficient to constrain the model calibration; the tracer plume movement was highly sensitive to the transient nature of the flow field. Great care must be taken to ensure the boundary conditions are properly constructed for successful analysis and understanding of the tracer tests.
- There existed significant intra-borehole vertical flows induced by the aquifer heterogeneity and highly dynamic fluctuations of the Columbia River, impacting the representativeness of the water samples.
- U(VI) migration in multi-rate SCR model is more dynamic and synchronous with the groundwater flow field which is consistent with field observations. The multi-rate SCR model appears to be a crucial consideration for future reactive transport simulations of uranium contaminants at the Hanford IFRC site.
- Parameter sensitivities are overall similar for both lab and field scales and total U und total sorption site density are the most important parameters.

References

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