US Department of Energy, Office of Science
Office of Biological and Environmental Research (BER)
Subsurface Biogeochemical Research (SBR)
FY10 Fourth Quarter Performance Measure

Introduction

The fourth FY10 SBR overall Performance Assessment Rating Tool (PART) measure for Pacific Northwest National Laboratory is to '*Provide a report that reconciles differences between model simulations of the field experiment and field data, and posses a path forward for future modeling to account for site complexity'*. This milestone is focused on research being performed at the Hanford Integrated Field Research Challenge (IFRC) site, located in the 300 Area of Hanford Site in southeastern Washington State. The 1600 m^2 Hanford IFRC site contains 36 groundwater monitoring wells placed within the footprint of the historic South Process Pond where uranium fuels-fabrication wastes were discharged. A 2 km^2 U(VI) groundwater plume exists at this location that exceeds regulatory limits. Uranium concentrations in the plume show complex seasonal changes that have not been predictable with any model applied. DOE is evaluating suitable and effective remedial strategies for the site.

The Hanford IFRC is investigating fundamental interactions between hydrologic, geochemical, and microbiologic processes that control uranium behavior in the plume with an emphasis on mass transfer. Mass transfer is one of several critical processes controlling the longevity of the U plume and its remediation, and involves the rate of U exchange between grain interiors and bathing fluids, and between waters in less permeable and more permeable sediment facies. Scientific understanding of the field-scale role of mass transfer is developed through comprehensive field characterization, injection experiments with non-reactive tracers and different uranium concentrations, monitoring experiments during periods of hydrologic transients and water table oscillations, and reactive transport modeling that incorporates physical and chemical heterogeneities. An important aspect of the research is the performance of manipulative experiments to investigate in-situ mass transfer rates and adsorption/desorption kinetics controlling U plume dynamics.

This report considers the first manipulative reactive transport experiment with U(VI) that was performed at the Hanford IFRC in October 2009. This was a preliminary experiment intended to test site infra-structure and to qualitatively measure the in-situ response of groundwater U concentrations to perturbation. It was performed over a time interval that was expected to be a stable period for the Columbia River, and before the experimental planning described in our Third Quarter Performance Measure (Q3) was initiated. This report first summarizes field and modeling results from the experiment. The site, and correspondingly the results are complex. The model includes kinetic adsorption/desorption controlled by surface complexation and mass transfer, and a heterogeneous hydraulic conductivity field parameterized through hydraulic measurements as described in our First, Second, and Third Quarterly Measures (Q1, Q2, and Q3). Potential physical and chemical causes are then discussed for disparities between the model and experiment, and for data features that are beyond the capabilities of the model. The collective results are evaluated to identify the best path forward for both future model improvements, and more definitive field experiments to quantify the in-situ kinetic process.

The Experiment

An exploratory U(VI) injection experiment was performed in October 2009 to test site infrastructure, and to obtain preliminary data on the response of U(VI) to perturbations in groundwater composition for more robust experimental planning. The experiment was performed during a period of projected stable river conditions, and before the project had the capability to pre-model potential experiment behavior as described in Q3. Thus, the field experimental conditions were not optimized.

The experiment was similar to Case 2 as described in Q3, and involved the injection of upgradient groundwater with 5 µg/L U(VI) and 180 mg/L bromide over the entire screened interval of Well 2-9 (e.g., upper high K, middle low K, and deep high K conductivity zones). At that time, the average background U(VI) concentrations in the IFRC site groundwater that was determined at the multi-level well clusters was 44 µg/L in the shallow zone, 40 µg/L in the middle zone, and 26 µg/L in the deep zone (Figure 1). Pumped groundwater concentrations from the fully screened IFRC wells at this same time were closer to the deep water concentration, ranging from 22 µg/L to over 30 µg/L (Figure 1). The withdrawal well was 1 km from the injection well requiring infrastructure testing and optimization for successful performance of the injection experiment. The upgradient groundwater was injected at a rate of 681.4 L per minute for 6.3 h (as compared to 24 h in Case 2), yielding an injected volume of 264,979 L. Wells in the IFRC site were monitored for approximately 366 h while the plume (tracer) was within the domain of the well-field. The desorption plume dissipated by the final, down-gradient well tier (e.g., 3-28, 2-29, and 2-23). However unexpected river stage oscillations began at approximately 1500 min that increased in intensity throughout the remainder of the experiment. These oscillations affected the observed breakthrough behaviors.

As in all past IFRC tracer experiments, the center of mass of the injected plume moved through the central region of the well field (e.g., 2-26/2-27/2-28; 2-14; 2-15; 2-17; 2-18; 2-19) with exit between 3-28 and 2-23 (Figure 1). Tracer and U(VI) breakthrough curves for the different wells were extremely varied, with representative cases shown in Figures 2 and 3. The results for depth-discrete well 2-27d and fully screened well 2-12 were of high quality, and were consistent with each another. Each showed the nearly simultaneous breakthrough of the Br and U(VI) peaks at approximately 50% concentration. U(VI) rebounded in each well to a concentration of 25 µg/L, the background concentration in the deeper aquifer unit. Br tracer displayed more tailing in well 2-27d, with fully screened well 2-12 displaying more variability in U(VI) concentration. Well 2-12 displayed a small, correlated concentration spike in both Br and U at 4500 min.

Fully screened wells 2-15 and 3-29 displayed more complex, but consistent behavior in spite of a significant distance between them (~30 m; Figure 1). In many ways, 2-15 was similar to 2-12, but with obvious data perturbations at 4000-4500 min, 9000 min, and 17000-21000 min. Tracer breakthrough in 2-15 was complete by 12000 min and U tended toward a concentration of 25 µg/L before 17,000 min (with the exception of the 4500 min spike), as it did in 2-12. However, the correlation of behaviors in the data perturbations was different in 2-12 and 2-15. U(VI) and Br were inversely correlated at 4000-4500 min, and positively correlated at 9000 and 12000 min in well 2-15. Well 3-29 displayed almost identical behavior to 2-15 in terms of data perturbations and trends, but exhibited lower breakthrough concentrations of both Br and U.

Three fully screened wells, 2-8, 2-10 and 2-18, displayed highly erratic breakthrough behavior (Figure 3, shown for only 2-10 and 2-18). For up-gradient well 2-10, the erratic behavior began at 2200 min after passage of the Br tracer plume and was associated with U(VI) concentrations only. The erratic behavior for down-gradient well 2-18 began at the same time and involved both Br and U(VI) concentrations. In all three of these wells, U(VI) concentrations oscillated between average values observed in the upper and lower aquifer zones, e.g., 25 µg/L and 40 µg/L.

Model Simulations

The reactive transport model (eSTOMP) was used on the EMSL Chinook computer to simulate the fate of the injected upgradient groundwater. These results were summarized and best visualized in the form of plume animations of U(VI) and tracer Br that were not included with this report because they cannot be shown in Microsoft Word. The animations reveal a plume that was relatively stationary for the first 6 h (360 min) of injection. After that, a complex trajectory was followed driven by river oscillations that involved rapid migration through the erosional channel in the central, deep high K zone of the site. Representative static comparisons are provided between field-measured breakthrough curves and model simulations for two select wells for the first 6000 min of the experiment (Figure 4). We note that the simulations required significant computer time and consequently, they were not carried out for the full duration of the experiment as shown in Figure 2.

We presented modeling results for Well 2-9 in the Third Quarterly Measure that closely matched experimental results. Model simulations for Wells 2-27d and 2-12 in Figure 2 also closely matched experiment. However there were significant disparities for all other wells as shown for two examples in Figure 4. Model simulations generally became worse with increasing distance from the injection well. For 2-10, model simulations for Br were excellent, and those for U were good for the first 2000 min of the experiment. The model however, could not account for the oscillatory behavior in U concentration that began and continued after 2000 min. Model simulations of Br in Well 2-7 captured some aspects of the tracer behavior, e.g., the initial breakthrough, but failed in other aspects. Unexpected tracer loss occurred between 750 and 2000 min, while unexpected tracer supply occurred from 4000 to 6000 min. The model simulations of U behavior in Well 2-7 were generally similar to the experimental data, but unexpected U supply occurred from 750-2000 min while unexpected U loss or retardation occurred from 3000-6000 min. The experimentally observed behaviors could arise from a plume whose center of mass oscillated over well 2-7 rather than moving directly through as predicted by the model. Oscillatory behavior in the breakthrough behavior of any well could no be described by the model.

Reconciling the differences between model predictions and the field data for this preliminary experiment was complex because of four primary reasons discussed below: i.) uncertainties in the monitoring data resulting from vertical well bore flows, ii.) physical heterogeneities in the flow field that have not yet been fully quantified, iii.) chemical effects that are not included in the model, and iv.) less than optimal experimental conditions.

Monitoring Data

The data from the U transport experiment as described and modeled above, combined with extensive down-hole electromagnetic borehole flowmeter (EBF) measurements have revealed that vertical flow occurs within the IFRC fully screened wells. Vertical flows are known to have a detrimental effect on monitoring data (Elci et al., 2003) and have been observed at other locations (Church and Granato, 1966; Elci et al., 2001; Hutchins and Acree, 2000, and Reilly et al., 1989). Ours is the first observation of vertical well flows at the Hanford site. The vertical flow occurs between the shallow and deep high K zones, and may be either upward or downward depending on well location and river stage. A subset of wells displays upward flow when river stage is increasing (+, Figure 5) and another displays downward flow (-, Figure 5) when river stage increases (Newcomer et al., 2010; Vermeul et al., 2011). These complex effects result from the interaction of river-induced hydraulic pressure waves with the heterogeneous hydraulic conductivity field of the site as further modified by the variable topography of an aquitard that resides at the base of the monitoring wells. The direction of well bore flows are related to the positioning of the monitoring wells in relation to an erosional channel in the surface of the aquitard (blue) that runs through the center of the site.

Well bore flows must be considered to interpret the results of the injection experiment. When upward well bore flow occurs, the sampled well waters will be representative of those in the lower high K zone. When downward well bore flow occurs the sampled waters will be representative of those in the upper high K zone. Detailed well-bore monitoring of two representative wells [2-10(+) and 2-21(-)] has revealed that flows may be consistently in one direction for extended periods (e.g., 7 d max), or they may oscillate between up and down with frequencies of less than a day (Figure 6). Borehole flow depends on the absolute magnitude of river stage, and its degree and frequency of fluctuation (Figure 6). In contrast, the predicted concentrations in Figure 4 are flux-based averages computed from the vertical hydraulic conductivity profiles for each well and are not responsive to vertical head gradients.

Well bore flows are important because U(VI) concentrations and transport velocities are different in the upper and lower high K zones (Figure 7). The tracer breakthrough shown in Figure 7 is for depth discrete wells and is not influenced by vertical flow. The desorption plume (including its high Br and low U) will move slower in the upper high K zone, but returning background waters will have higher U than in the lower zone. A high transport velocity in the lower high K zone may have allowed tracer to exit this part of the formation, while it was still present in the upper high K zone. A discussion of select examples from Figures 2 and 3 is provided to illustrate this complexity. Well bore flows create data anomalies in the breakthrough behavior monitored by the fully screened wells that cannot be described with the currently applied model that predicts flux-based averages without vertical well-bore gradients.

1.) Well 2-12 appears to be dominated by primarily by upward flow and is monitoring the water composition of the lower high K zone. The small anomaly at 4500 minutes represents a short period of downward flow from the upper high K zone where tracer migration is slower.

2.) Wells 2-15 and 3-29 are also dominated by upward flow, with flow reversals (downward flow) occurring at the anomalies (4500, 9300, and 19500 min). The anomalies represent the intrusion of water from the upper high K zone with higher U, and different concentrations of Br resulting from its slower transport velocities. U(VI) concentrations of 40 µg/L at the end of the experiment are indicative of upper zone water composition.

3.) Wells 2-10 and 2-18 (Figure 3) displayed the effects of widely oscillating well bore flows of relatively short frequency. Well 2-10 monitored the upper aquifer zone for the first 2000 min, followed by the lower zone from 2000-4000 min (Figure 4). Continued oscillations in U concentration thereafter indicated frequent shifts in the direction of well bore flow. Well 2-18 monitored the lower zone from 0-1500 min and the upper zone from 1500-4500 min. Like 2-10, frequent oscillations in both U and Br concentrations thereafter indicated frequent shifts in the direction of well bore flow. In some ways, the Br breakthrough behavior in 2-18 can be viewed as a fragmented superposition of the curves for wells 2-26 and 2-27 in Figure 7.

While these examples demonstrate that the data anomalies can be qualitatively reconciled by considering well bore flows and the differences in water composition between the upper and lower high K zones, there are problematic inconsistencies between wells that are challenging to interpret in a robust or quantitative manner.

Physical Heterogeneities

A major emphasis of IFRC research has been to quantify the hydraulic conductivity field within the experimental domain. This activity was described in our first quarterly measure, and research has continued over the intervening period as it is not complete. It is a complex endeavor as the conductivity field must be quantified in three dimensions at relatively high resolution to describe groundwater movement in a highly dynamic environment. Characterization has involved direct measurements by flow-meter and pumping tests, and inverse and Monte-Carlo modeling of tracer experiments. The Hanford IFRC poses unique challenges for hydrologic modeling because of linkage to the

Columbia River: hydrologic gradients vary by a factor of 10 with corresponding effects on groundwater velocity, groundwater flow directions vary by 90°, and vertical gradients vary hourly, daily, and seasonally. Animations of all of our tracer injections reveal highly complex plume behaviors that change directions and velocities multiple times during an experiment. Given the hydrologic and geologic complexities, any single tracer experiment will only sample a subset of the conditions and environments that must be understood and or quantified to describe transport over the full range of conditions to be encountered.

The hydraulic conductivity model used for the calculations herein was calibrated to our first tracer experiment in Nov 2008, and then used to simulate our March 2008 tracer experiment (Figure 8), both without and with consideration of well-bore flow effects (Ma et al., 2010). This experiment was performed under relatively stable river conditions. The well bore effects were calculated with the MODFLOW Multi-Node Well Package (MNW; Zheng, 2006; Ma et al., 2011). The calculated well-bore effects are strongly dependent on both the large-scale and local hydraulic conductivity structure. It is evident from Figure 8 that our hydraulic conductivity model only yields semi-quantitative predictions of tracer breakthrough at best. For some wells the simulations were not good, e.g., 2-14 and 3-29. Consideration of well bore-flows yielded some improvement, especially 2-10 and 2-8 (Ma et al., 2011), but in no case could the sometimes jagged or abrupt-changing breakthrough curves be described. From Figure 8 we conclude that the IFRC site physical/hydrologic model is not yet sufficiently robust to allow accurate predictions of non-reactive or reactive transport. The IFRC team also believes that disparities between predicted and observed tracer behavior results from inadequately defined hydrologic boundary conditions for the site, in terms of precisely measured water table elevations to accurately establish the magnitude and direction of the time-variable hydraulic gradient.

Geochemical Effects

The reactive transport model applied to the calculations in Figure 4 assumed homogeneous geochemical conditions throughout the entire IFRC domain in terms of adsorbed U and adsorption site concentration. Additionally, it was assumed that the groundwater composition was 35 μg/L in all three aquifer zones, when analyses showed otherwise (Figure 1and Figure 7). We also know from our broader characterization activities that adsorbed U concentrations and the mass of < 2mm sediment (the geochemically reactive fraction) vary significantly across the site (Figure 9; Murray et al., 2011), and that these variations cause significant differences in groundwater U concentrations between wells in the upper aquifer zone (McKinley et al., 2011). Model sensitivity calculations over the data ranges shown in Figure 9 have revealed that the noted variations in adsorbed U and the < 2 mm fraction (e.g., reactive site concentration) in the upper aquifer zone can produce large, local concentration differences from those predicted with the homogeneous model.

The simplifying geochemical assumptions of homogeneity were made to facilitate tractable simulations in terms computation time. The basic multi-rate surface complexation model is a complex one to integrate into reactive transport simulations because it involves 50 sites with different kinetic rate constants. Our overall modeling strategy was, and still is, to add geochemical complexity and heterogeneity after we demonstrate an ability to accurately simulate non-reactive tracer behavior. Thus an important requirement for any future modeling is that we quantitatively describe the fate of our injected waters in terms of flow path, mixing, and physical mass transfer.

A final geochemical consideration is whether the reaction parameters used for the multi-rate surface complexation process were representative of in-situ reactivity. As discussed in our Second Quarterly Measure, a single set of these parameters was fit to effluent data from one-dimensional transport experiments performed with three intact sediment cores from the saturated zone. Due consideration was given in fitting to the parameters whose values have the most important influence on predicted U behavior. These parameters were identified through sensitivity analysis of the multi-rate model (Greskowiak et al., 2010). The variance in these fitted parameters was unexpectedly small, suggesting they were representative of all three core behaviors. Unknown, however, is their true relevance to in-situ conditions where reactive flow is multi-dimensional, and where the porosity, grain size, and sediment structure may be different. While the answer to this question of in-situ relevance must await a more robust field experiment, our ability to simulate U breakthrough behavior in select wells proximate to the injection well (e.g. 2-27, 2-12, and early 2-10) suggests that our laboratory measured parameters may be realistic.

Experimental Design

The October 2009 U desorption experiment was not optimized to evaluate our laboratory-derived kinetic model. Requirements for such testing were developed and described in our Third Quarterly Measure. The optimal experiment will inject at least 20 times more water than used here over a period of 10-20 days. A much larger and more sustained deflection in groundwater U concentrations will occur throughout the well field that will allow participation of more adsorption sites holding contaminant U(VI). Multiple non-reactive tracer additions will be made during the course of the injections to define water migration and mixing patterns in the most robust manner possible. The final experimental design may also employ hydrologic control through a dipolar injection-withdrawal system to maximize mass balance. Important modifications to the monitoring system, however, are needed to yield breakthrough data that is not complicated by vertical borehole flow.

Path Forward

We conclude from the analyses in this report that the major causes for discrepancies between the Oct 2009 U experiment data and model predictions are: 1.) vertical well-bore flows that greatly complicate the monitoring data for both U and Br breakthrough, 2.) inadequacy in the hydraulic conductivity model and hydrologic boundary conditions that prevent accurate descriptions of non-reactive tracer behavior and the complex plume migration patterns that occur, and 3.) an experimental design that did not sufficiently perturb the geochemical regime in the well-field. Inadequacies were also identified in the geochemical model, but these were considered secondary, at this time, to the three issues identified above.

An important long-term objective of Hanford IFRC research is the development of a pragmatic reactive transport simulator that can describe seasonal U(VI) concentration dynamics in the groundwater of the IFRC site. It is thus critical that a well-designed U transport experiment be performed and subject to the most robust reactive transport modeling possible as a first step in the documentation of this capability. The following steps are proposed for FY2011 to achieve success in this endeavor.

- Mitigate the vertical well-bore flow issue in the well-field to enable unambiguous monitoring results. This problem has been under intense study for the last 9 months with the testing of a sequence of mitigation approaches of increasing severity. As a result of this testing that is now complete, we now plan to grout the lower 2/3 of the fully screened wells in November 2010 to yield a monitoring system within the upper high K zone (UHKZ) of the U-plume. The rationale for this decision and detailed plans will be described in a report to be presented to BER at the beginning of FY 2011.

- Perform robust tracer testing in the UHKZ after the well mitigation task is complete during two distinct hydrologic periods. The two hydrologic periods will present different gradients, groundwater flow directions and velocities, oscillation frequencies, and plume trajectories through the well-field to yield a rich transport data set. The new tracer results will be utilized to further calibrate the hydraulic conductivity field by inverse modeling.

- Add four new wells at the apices of the modeling domain (e.g., 60 m beyond the IFRC domain) with continuous water level monitoring and no disturbance to alleviate ambiguities with the hydrologic boundary conditions. The hydrologic gradient across the IFRC site is very small but dynamic, and may vary by a factor of 10. Wells of opportunity (from the Hanford site monitoring array) at sub-optimal locations are currently used for these measurements that require sustained and repeated water-level measurements to the accuracy of mm. Occasional movement of the monitoring equipment for well access for other measurements creates uncertainties and ambiguities in the water level record that reduces the accuracy of plume modeling. Improved boundary measurements will lead to more accurate predictions of complex plume trajectories that result from river stage oscillations.

- Optimize plans for an improved U(VI) desorption experiment in the UHKZ that will access necessary time-scales and maximize tracer and U mass balance. The general attributes of a successful desorption experiment that appropriately considers the complex implications of multi-rate desorption behavior were discussed and identified in the Third Quarterly Measure. These calculations were based on injection into the fully screened interval. These attributes must now be optimized (e.g., injection volumes, durations, timing, and hydrologic control) for the UHKZ in follow up calculations that consider: i.) the range in hydrologic and geochemical conditions that may be encountered in this zone during the experiment, and ii.) multiple realizations of the potential hydraulic conductivity field supported by recent IFRC geostatistical evaluations.

The overall strategy for reactive transport model development at the IFRC has be an iterative one where we first seek to describe dynamic local groundwater flow driven by river stage variations, and then build in the added complexities of kinetic U(VI) adsorption/desorption geochemistry. It is clear that we cannot expect to predict the behavior of U, regardless of geochemical model, if we cannot predict non-reactive tracer behavior. In fact, the quantitative evaluation of the U geochemical model and its parameters can only follow as a consequence of the accurate modeling of tracer behavior. The path forward described above is intended to rectify this problem, and to allow the development of state-of-science data sets for coupled model evaluation.

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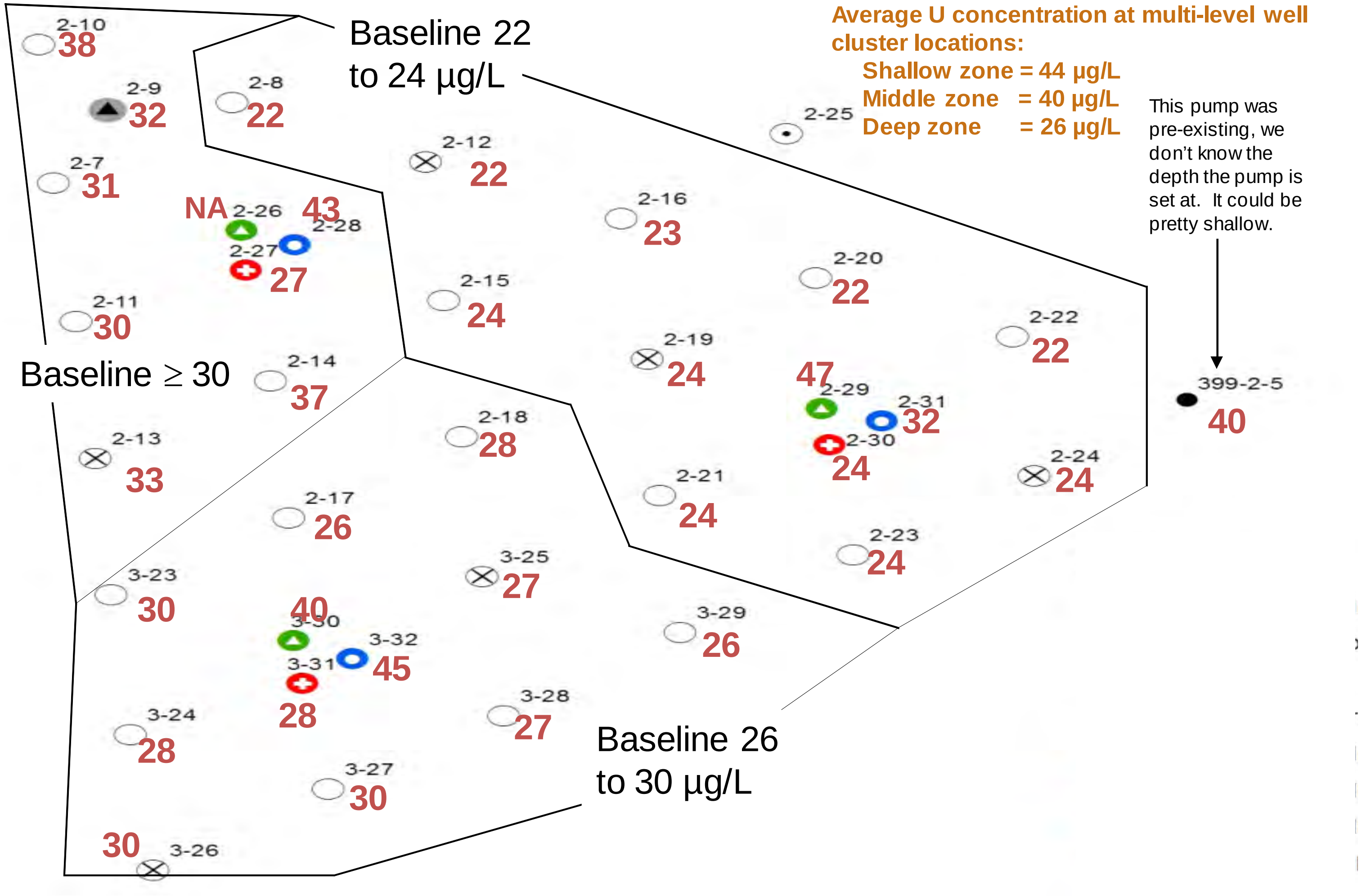


Average U concentration at multi-level well cluster locations:
Shallow zone = 44 µg/L
Middle zone = 40 µg/L
Deep zone = 26 µg/L

This pump was pre-existing, we don't know the depth the pump is set at. It could be pretty shallow.

Figure 1. Uranium (VI) concentrations measured in IFRC groundwater monitoring wells immediately preceding the October 2009 desorption injection experiment. All concentrations given in µg/L (parts per billion).

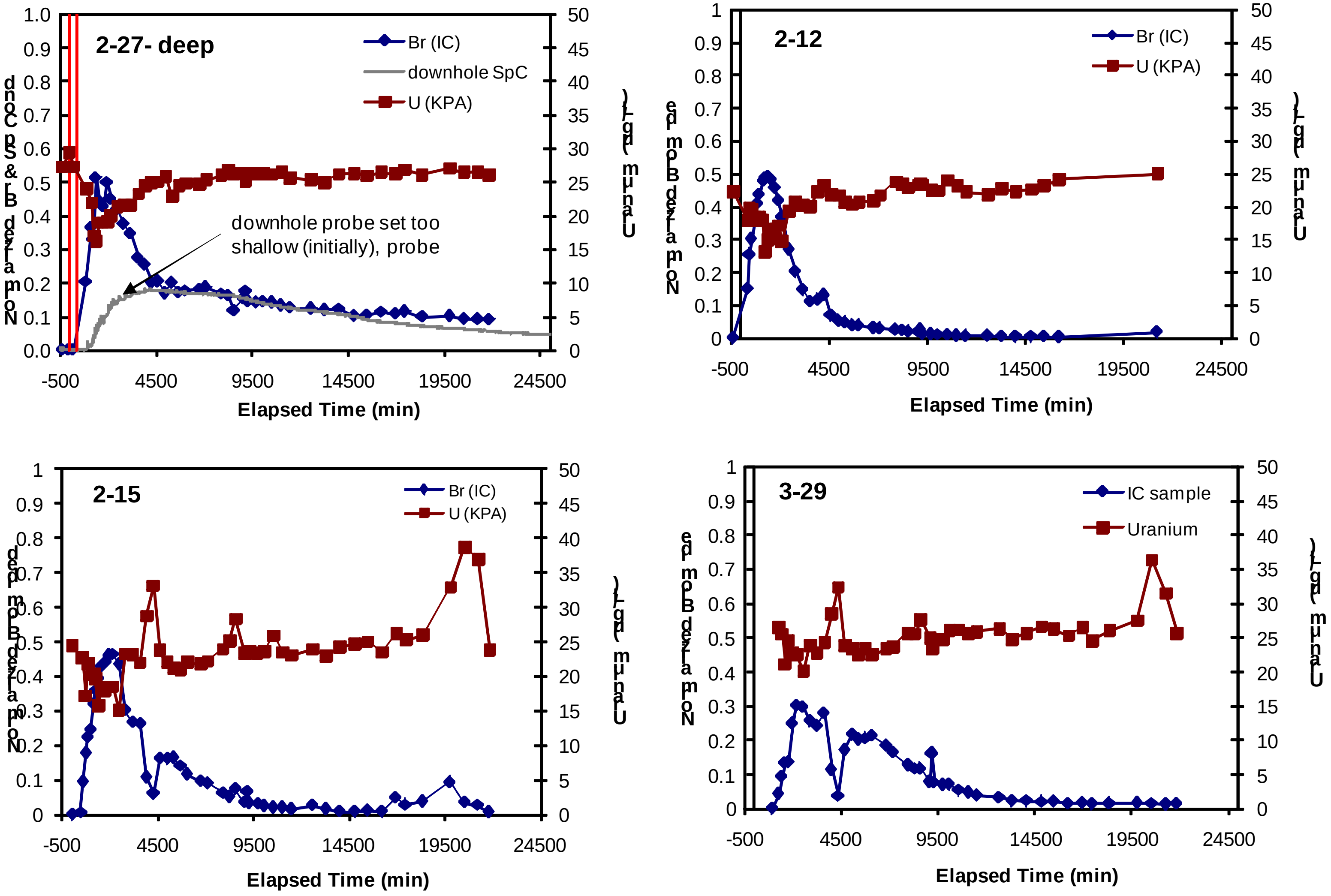


Figure 2. Breakthrough behavior of Br and U(VI) at wells 2-27 deep, 2-12, 2-15, and 3-29 over the entire course of the October 2009 U desorption experiment. Note well locations in Figure 1.

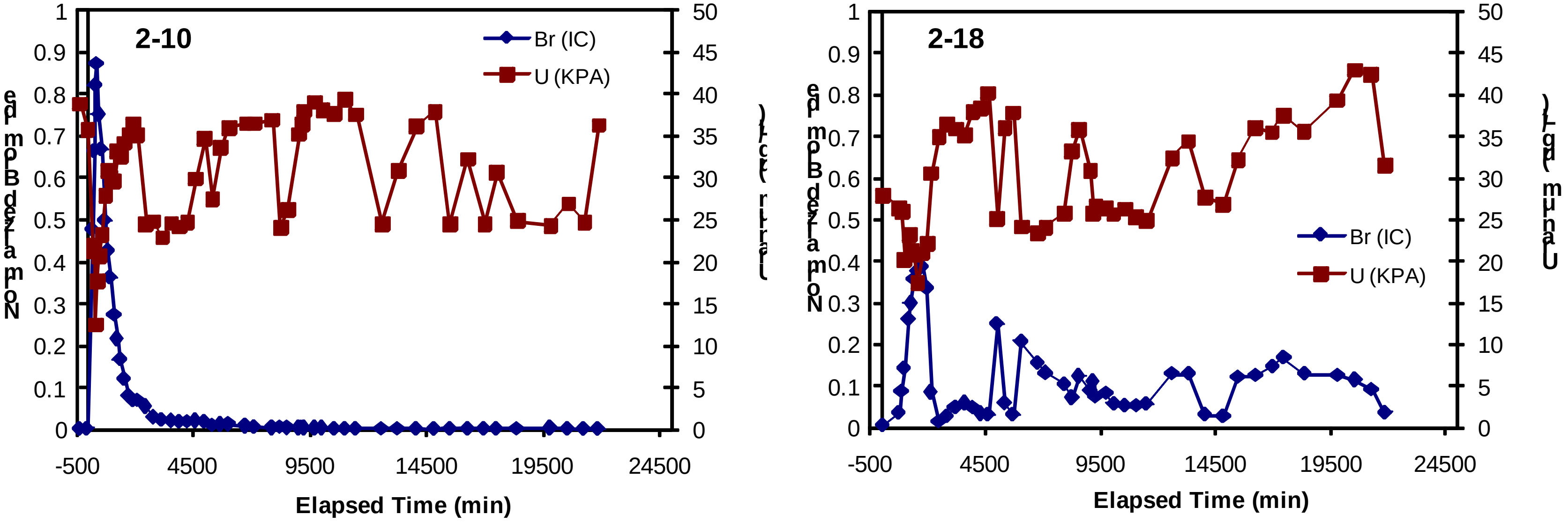


Figure 3. Breakthrough behavior of Br and U(VI) at wells 2-10 and 2-18 over the entire course of the October 2009 U desorption experiment. Note well locations in Figure 1.

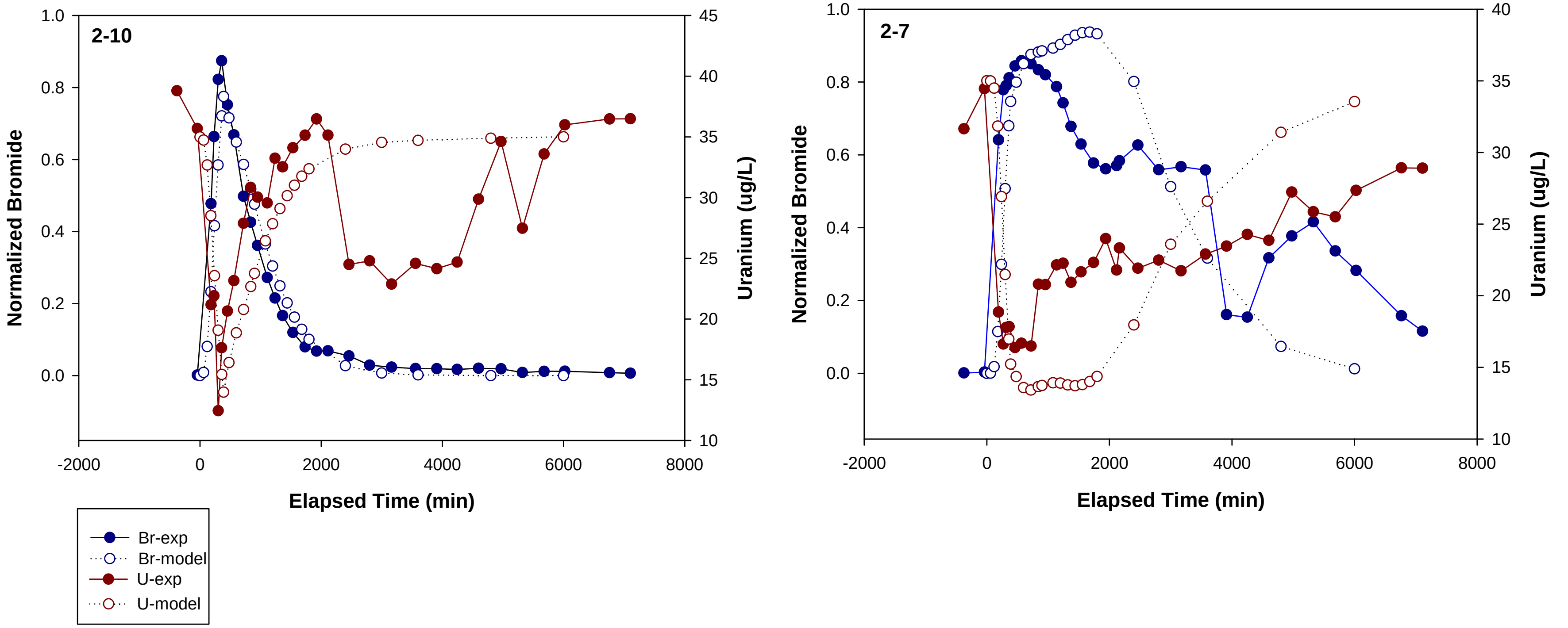


Figure 4. STOMP model simulations of Br and U(VI) breakthrough at wells 2-10 and 2-7.

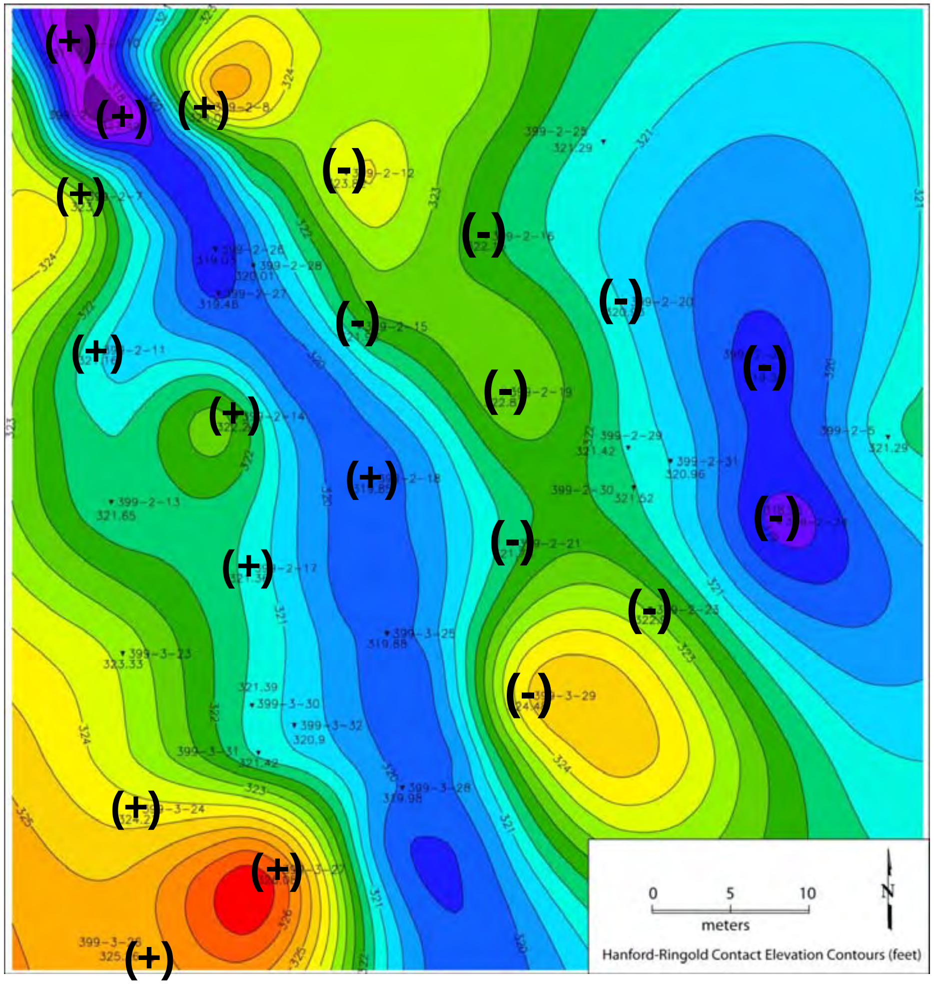


Figure 5. Topography of the Hanford-Ringold contact within the IFRC site. Blue and purple are low elevation, yellow-orange-red are high elevation. An erosional channel through the central region of the site that is filled with high K material (blue) is a preferred flow path. The correlation of well-bore flow with river stage is related to the topography of the contact and its subsequent impact on groundwater flow and linkage to the river.

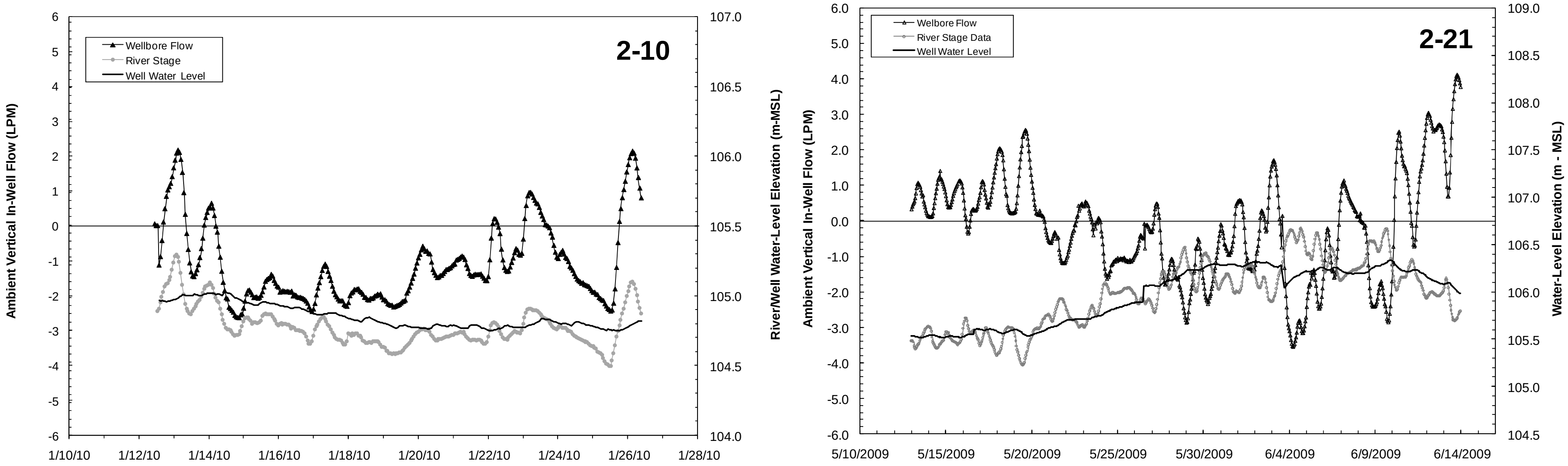
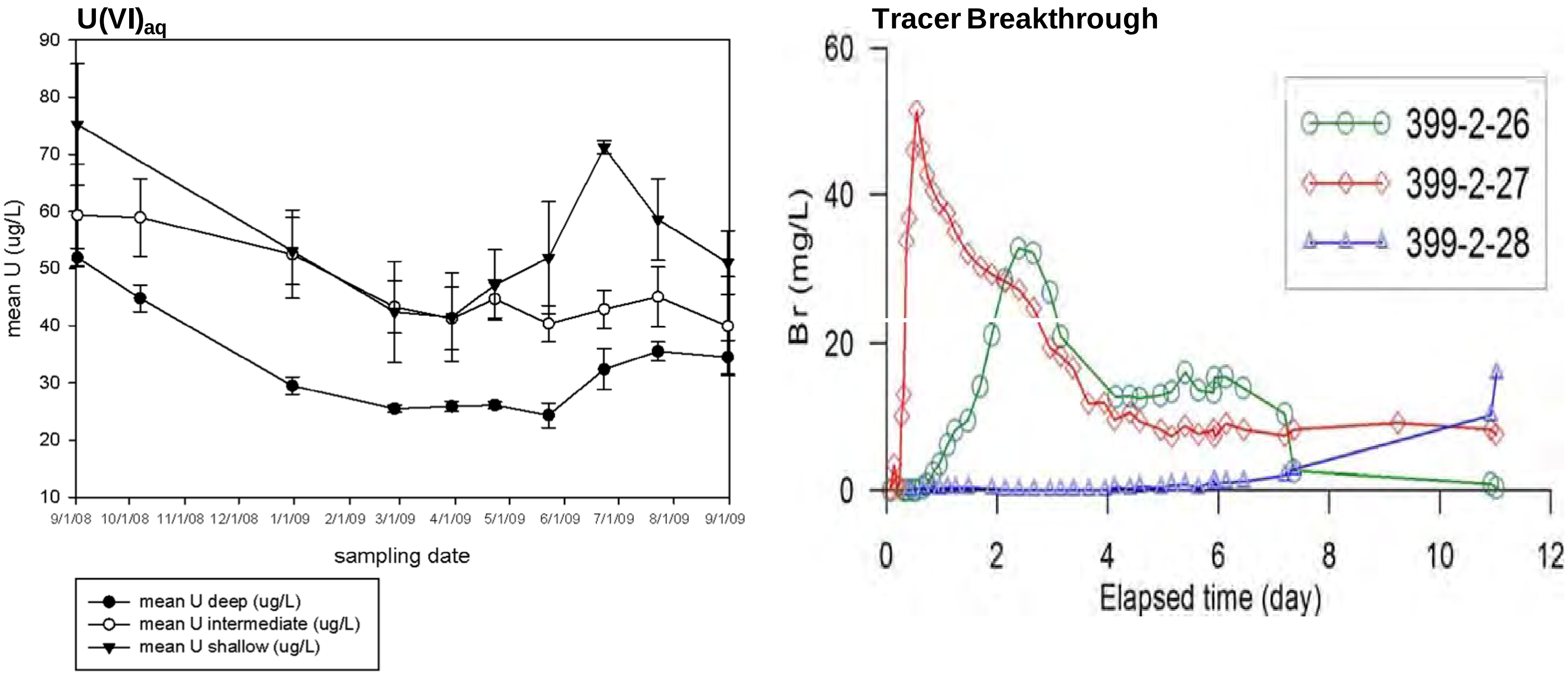


Figure 6. The direction and magnitude of borehole flow in wells 2-10 and 2-21 as measured by the electromagnetic borehole flowmeter. Positive values indicate upward flow, negative values indicate negative flow. Note that different date intervals were monitored for each well. Shown also are water level elevations in the wells and river stage elevations.



$U(VI)_{aq}$

Tracer Breakthrough

Figure 7. Average groundwater U(VI) concentrations measured in the depth discrete monitoring wells from 9/1/08 to 9/1/09 (left). Groundwater in the upper high K zone displays the highest U(VI) concentrations and the greatest seasonal variation. Tracer breakthrough curves measured in the west three-well cluster during the November 2008 tracer experiment (right). The most rapid transport is observed in the deep high K zone, followed by the upper high K zone.

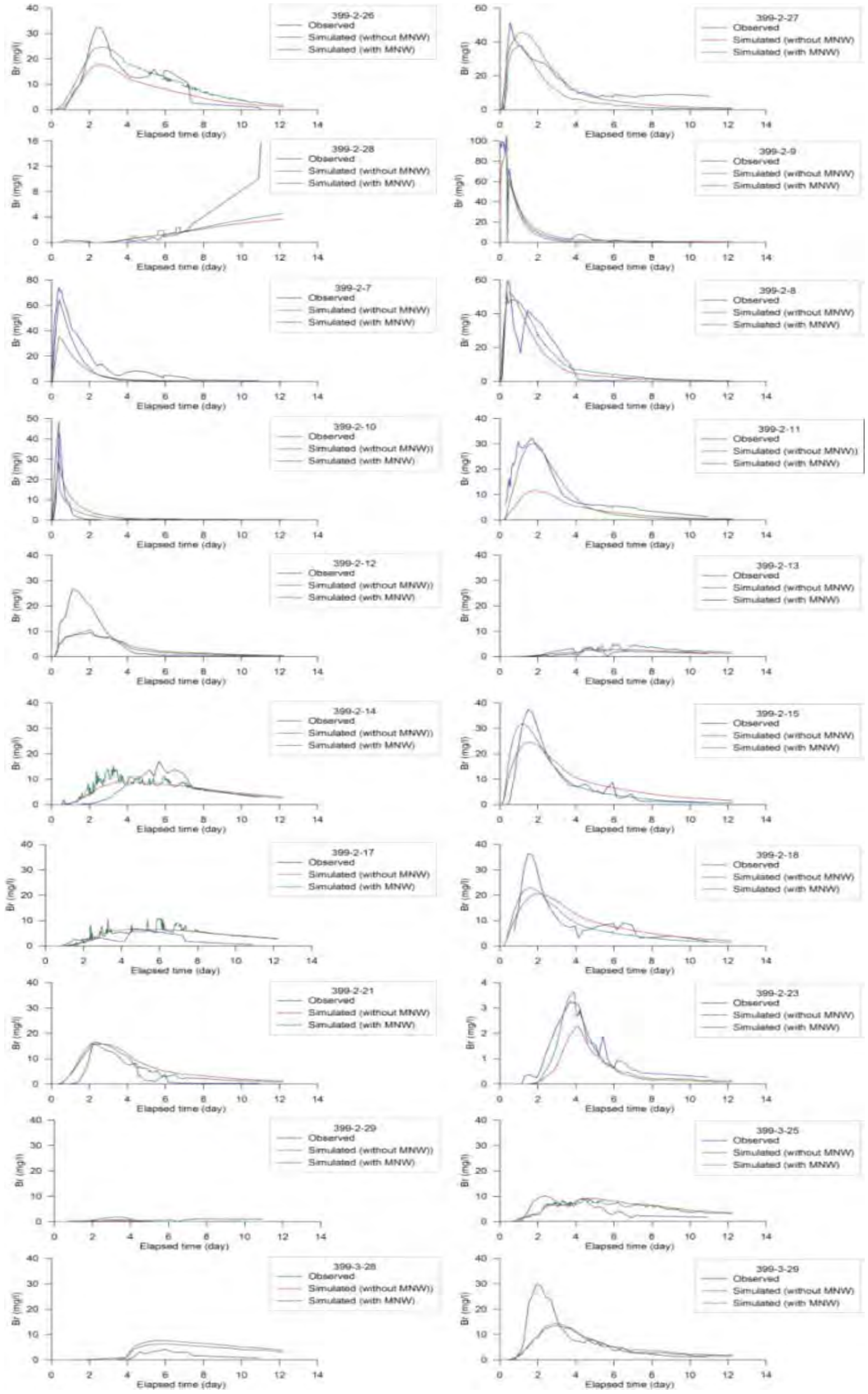


Figure 8. Simulated tracer (Br) breakthrough curves using MODFLOW for the March 2009 and tracer experiment. The multi-node well package (MNR) was used to compute the impact of well-bore flows on solute concentrations. MNR improves the simulation quality for some but not all wells.

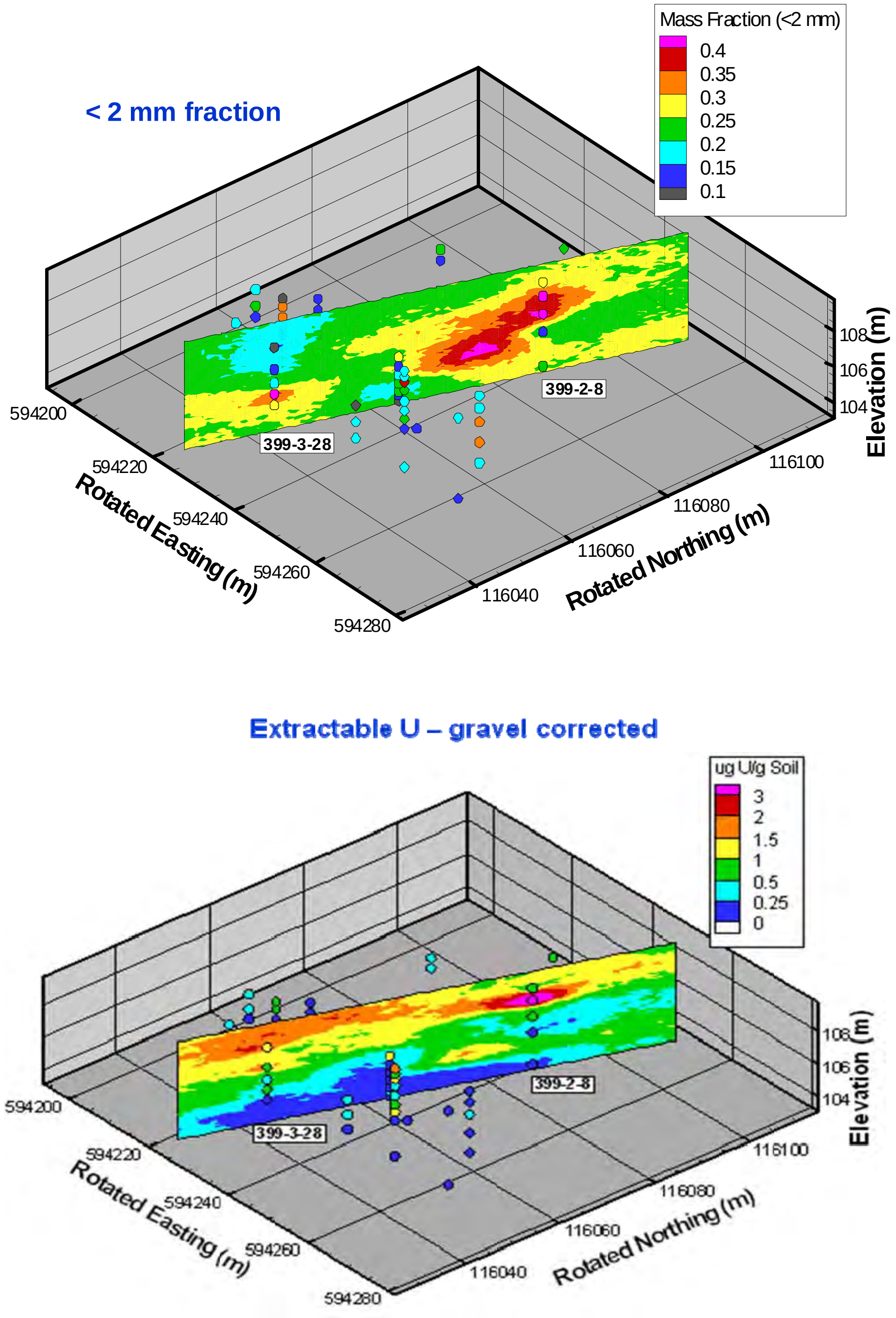


Figure 9. Geostatistical distributions of the reactive < 2mm sediment fraction (left) and bicarbonate extractable U(VI) (labile U) over the elevation interval 103-109 m. The sample set was approximately 200. This depth interval spans the zone of water table variation in the lower vadose zone and the upper high K region in the saturated zone. There are localized regions of correlation between reactive fines and extractable U, but the global correlation is not strong. The extractable U concentrations are influenced by both geochemical reaction in the sediment, and historic operational details.