



U.S. Department of Energy Office of Science

Environmental Remediation Sciences Program

INTEGRATED FIELD RESEARCH CHALLENGE SITE Hanford 300 Area



Hanford 300 Area

Multi-Scale Mass Transfer Processes Controlling Natural Attenuation and Engineered Remediation: An IFC Focused on Hanford's 300 Area Uranium Plume

J. Zachara*, PNNL; M. Freshley, PNNL; B. Bjornstad, PNNL; J. Christensen, PNNL; M. Conrad, PNNL; J. Fredrickson, PNNL; G. Hammond, PNNL; R. Haggerty, Oregon State U; P. Jardine, ORNL; D. Kent, USGS; A. Konopka, PNNL; P. Lichtner, LANL; C. Liu, PNNL; J. McKinley, PNNL; M. Rockhold, PNNL; Y. Rubin, UC/Berkeley; V. Vermeul, PNNL; R. Versteeg, INL; A. Ward, PNNL; C. Zheng, U. Alabama; and M. Thompson, DOE 300 A Site Steward

SCIENCE THEMES & CONCEPT

Hanford's 300 A Uranium Plume

- Large volumes of process water from fuel fabrication resulted in extensive U groundwater contamination.
- Highly contaminated waste pond sediments excavated, Cu- and U- containing vadose zone sediments remain.
- Prevalent U groundwater contamination remains above regulatory limits.
- Complex seasonal concentration trends associated with river stage changes and WT rise and fall.

South Process pond - Pit #2

- Significant ERSP research on 300 A sediments (~10 papers)
 - ◊ molecular speciation of adsorbed and precipitated species
 - ◊ adsorption/desorption and dissolution kinetics
 - ◊ bulk, surface, and microscopic mineralogy
 - ◊ mass transfer behavior and modeling

Hanford IFC Site Concept

- Wells spaced 10 m to allow cross-hole geophysical interrogation for inter-well properties determination.
- Injection experiments (10³ gallons) in the 6 m saturated zone under different seasonal gradients using site groundwaters.
- Passive experiments will exploit natural gradients and WT fluctuations.
- Continual water level monitoring at 12 locations to quantify boundary conditions.
- Overall goal is to understand system scale behavior and controlling role of mass transfer.
- Site supports a technical theme of multi-scale characterization and modeling.

IMPORTANT PROGRESS

EBF Profiles

- State of the art well field and monitoring system installed and tested ("well-field report").
- Extensive hydrologic testing completed including a non-reactive tracer injection that has modified the site hydrologic model.
- Significant progress made toward the site 3-D geostatistical model ("Characterization Plan")
 - ◊ Laboratory chemical, physical, and electrical measurements.
 - ◊ Surface, downhole and cross-hole geophysical measurements.
 - ◊ Inversion modeling and geostatistical analysis.
- Modeling Plan nearly completed.
 - ◊ Data collection sequences.
 - ◊ Sequential modeling goals.
 - ◊ Roles and responsibilities.
 - ◊ High quality samples obtained for microbiologic characterization that have shown surprising results.
 - ◊ Establishing collaborative strategy and experiments with PNNL SFA.
 - ◊ Completed low-water, multi-non reactive tracer and AT injection experiment in March 2009.

Borehole Summary Log for Well 399-2-9

- A report has recently been completed that summarizes all aspects of the IFC well field: "Borehole Completion and Conceptual Hydrogeologic Model for the IFC Well Field, 300 Area, Hanford Site".
- The figure to the left is an example of information summarized for each of the 35 IFC wells in the report, borehole log for each well are also included.
- A hydrogeologic model is developed for the site to inform reactive transport modeling.

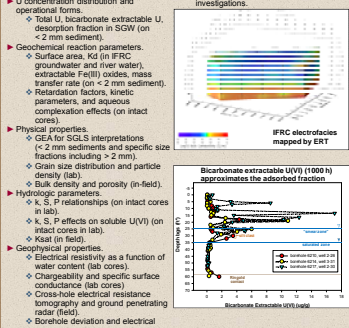
CHARACTERIZATION

IFRC Modeling & Parameterization Workflow

- A modeling plan is nearing completion that describes the various steps needed to model IFC experimental results in the most robust manner possible.
- The plan holds for development of a 3-dimensional site geostatistical model of hydrologic and geologic properties, geochemical reaction parameters (surface complexation and mass transfer), and U concentration distribution through a new inversion modeling technique.
- This complex model will be formulated through field and laboratory measurements performed by the Hanford IFC (green), and by the collaborating PNNL Science Focus Area (SFA) project (blue) investigating microenvironments and transition zones.
- A modeling team with expertise in multiple reactive transport codes, including MODFLOW, FLOTRAN, and STOMP, and inversion methods, are implementing the model using new characterization data and analysis of laboratory and field experimental results.
- Both Hanford IFC and PNNL research will support future planned field mobilization experiments and UV(U) mobilization experiments, and microbial ecology and biogeochemistry investigations.

Ongoing Characterization

- U concentration distribution and operational forms.
 - ◊ Total U, bicarbonate extractable U, desorption fraction in SSOW (on < 2 mm sediment).
- Geochemical reaction parameters.
 - ◊ Surface area, Kd (in IFC groundwater and river water), extractable Fe(II) oxides, mass transfer rate (on < 2 mm sediment).
 - ◊ Retention factors, kinetic parameters, and aqueous complexation effects (on intact cores).
- Physical properties.
 - ◊ GSA for SCL interpretations (< 2 mm sediments and specific size fractions including > 2 mm).
 - ◊ Grain size distribution and particle density (lab).
 - ◊ Bulk density and porosity (in-field).
- Hydrologic parameters.
 - ◊ α, S, P relationships (on intact cores in lab).
 - ◊ α, S, P effects on soluble UV(U) (on intact cores in lab).
 - ◊ Ksat (in field).
- Geophysical properties.
 - ◊ Electrical resistivity as a function of water content (lab cores).
 - ◊ Changeability and specific surface conductivity (lab cores).
 - ◊ Cross-hole electrical resistance tomography and ground penetrating radar (field).
 - ◊ Borehole deviation and electrical conductivity (field).



SIGNIFICANT NEW FINDINGS

- Correlated heterogeneity in IFC-site saturated zone hydraulic properties.
- In-situ temperature monitoring allows characterization of groundwater flow paths.
- Enhanced connectivity of the Columbia River to the upper third of the saturated zone.
- Greater than expected groundwater-river coupling.
- Lower than expected groundwater travel times (< 3 m/d).
- Smear zone with significant labile, sorbed UV(U) inventory.
- Anomalous UV(U) adsorption to fines in all compartments.
- Significant mass transfer of contaminant UV(U) in intact, saturated zone sediments.
- Robust microbiologic community in the Hanford formation saturated zone.

FIELD EXPERIMENTAL PLANS

Field Electrode Measurements and Aqueous Sampling

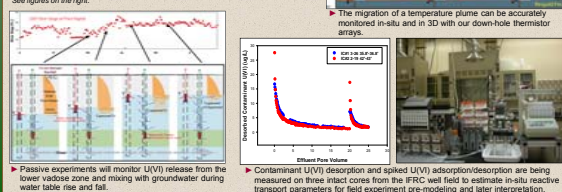
- The IFC site has continued monitoring of water levels, temperature, and specific conductance in wells around its periphery, and in the nearby Columbia River.
- 27 of the wells have been instrumented with downhole ERT electrodes and thermistors (see right).
- All wells contain dedicated downhole pumps that are routed through a manifold system to an on-site laboratory.
- Downhole pumps are computer activated at necessary times for plume sampling during movement.

Monitoring Well Schematic

- 10 cm ERT electrodes spaced at 60cm.
- Thermistors for temperature measurements placed between ERT electrodes.
- Electrodes and thermistors in sand pack in vadose zone and dangle in groundwater to allow removal.

EXPERIMENTAL CAMPAIGNS FOR MARCH 2009 - MARCH 2010

- Multi-tracer, cold water injection (completed March, 2009).
 - ◊ A combination of cold water and multiple non-reactive tracers were injected into the IFC site during a period of stable, toward-river groundwater flow.
 - ◊ Initial temperature and specific conductance were monitored down-hole, and samples were collected for laboratory analysis of bromide and deuterium.
 - ◊ The objectives were to define historical heterogeneities in water flow paths in the IFC site, and to quantify their effects on well-averaged tracer concentrations.
- Passive during rising water table (May - July, 2009).
 - ◊ Groundwater temperature and chemical composition including U will be monitored as river stage increases in response to spring snow-melt and causes water table rise into the smear zone.
 - ◊ Well-averaged groundwaters and near surface water from the upper 1' of the well will be sampled. Analytical results will be correlated with continuous monitoring data for river stage, potentiometric surface, and temperature and specific conductivity of all waters.
 - ◊ The objective is to determine UV(U) is mobilized from the lower vadose zone to groundwater during periods of high water table.
- Desorption Injection (September, 2009).
 - ◊ Upgradient groundwater with lower UV(U) concentration than found in the IFC site along with two non-reactive tracers and a salinity contrast will be injected during a stable hydrologic period with toward river groundwater flow.
 - ◊ Plume migration and mass transfer limited regions will be monitored in-situ by ERT and specific conductance, with laboratory analysis of the non-reactive tracers and U in sampled well waters.
 - ◊ The objective is to quantify the in-situ desorption kinetics of adsorbed contaminant UV(U) and how it varies in response to U concentration distribution and average water velocity.
- Adsorption Injection (March, 2010).
 - ◊ 300 U plume groundwaters from a nearby well will be injected that contain a higher concentration of dissolved UV(U), a 9°C temperature differential, and one non-reactive tracer into the IFC well field during a period of stable, toward-river groundwater flow.
 - ◊ Plume migration will be monitored by down-hole temperature and specific conductance measurements, with field potentiometry of pH, and robust (spatial, temporal) laboratory measurements of UV(U) and the target tracer. Groundwater compositional analysis may also be required depending on differences between the injected and in-situ groundwater.
 - ◊ The objectives are to investigate the in-situ retardation behavior of U by surface complexation, and the role of mass transfer in controlling the leading and trailing edges of the reaction plume.
 - *See figures on the right.



PLENARY SESSION CONTENT

Hanford 300 Area IFC
John Zachara, PNNL Lead PI and research team members

Geophysical Characterization of the Hanford IFC Site
Andy Ward (PNNL), Roelof Versteeg (INL), Timothy Johnson (INL), and Christopher Strickland (PNNL). (A joint contribution from the Hanford IFC and the PNNL-SFA)

The Method of Anchored Distributions (MAD) for Integration and Inversion of IFC Hydrogeological Data and for Establishing a Geostatistical Site Model
Yoram Rubin (UC Berkeley), Haruko Murakami (UC Berkeley), Xinyuan Chen (UC Berkeley), Heng Bai (UC Berkeley)

Hydrologic Characterization and Results from the First Tracer Experiment at Hanford's 300 Area IFC Site
Mark Rockhold (PNNL), Vince Vermeul (PNNL), Chris Murray (PNNL), and John Zachara (PNNL)

Variability in Adsorbed Uranium Concentrations in Saturated-Zone Sediments from the IFC Tracer-Test Well Array in Hanford's 300 Area Uranium Plume
Douglas Kent (USGS), Deborah Stoiker (USGS), and John Zachara (PNNL)

Modeling Field-Scale Uranium Mass Transfer at the Hanford IFC Site
Chunshuo Zheng (University of Alabama), Rui Ma (University of Alabama), Henning Prommer (CSIRO, Australia), Chongyan Liu (PNNL), Jarek Greskowiak (CSIRO, Australia), John Zachara (PNNL), and Mark Rockhold (PNNL)

Microbial Ecology in Subsurface Sediments from Hanford 300 Area Unconfined Aquifer
Allan Konopka (PNNL), Xuxu Lin (PNNL), David Kennedy (PNNL), Jim K Fredrickson (PNNL), and Rob Knight (University of Colorado) (A PNNL-SFA contribution)

CONTRIBUTIONS TO SITE CLOSURE

- System scale understanding of factors controlling groundwater UV(U) concentrations
- Understanding of site heterogeneities (physical, chemical, and biologic) and implications to reactive transport
- Robust multi-process models for assessment of remediation effectiveness.
- Comprehensive properties relationships and regressions for prediction.
- Documented multi-scale characterization and modeling approaches.
- Understanding of linked groundwater-river systems.

EXAMPLE OPPORTUNITIES FOR COLLABORATIVE RESEARCH

- In-situ adsorption/desorption experiments of various types.
- Laboratory to field comparisons.
- Evaluation of geophysical methods and inversion techniques.
- Mass transfer processes of different types at different scales.
- Microbiology of linked groundwater-river systems of low to high transmissivity.
- Geologic, hydrologic, geochemical, and biogeochemical modeling of different types.

MATERIALS AVAILABLE TO EXTERNAL INVESTIGATORS

- Historic UV(U)-contaminated source term materials (limited).
- Contaminated UV(U) vadose zone materials whose geochemical speciation and mass transfer properties have been determined (limited).
- Microbiology of linked groundwater-river systems of low to high transmissivity.
- Uncontaminated vadose zone and aquifer sediments from various locations.
- Circumneutral site groundwaters with variable UV(U), HCO₃⁻, and Ca concentrations.
- Core materials and grab samples from vadose zone and aquifer experimental plots.
- Ringold formation aquifer sediments of variable redox properties (limited).
- See website for details: <http://efc.hanford.pnl.gov>