PNNL-SA-58090



The Hanford 300 A Integrated Field Challenge

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

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Supported by DOE Office of Biological and Environmental Research (OBER) Environmental Remediation Sciences Division (ERSD)



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FREC-IFC Telecon November 20, 2007 Pacific Northwest National Laboratory U.S. Department of Energy

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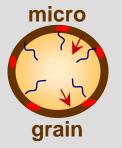
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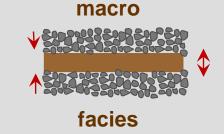


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Hanford IFC

Science Theme ~ Multiscale mass transfer processes influencing sorbed contaminant migration



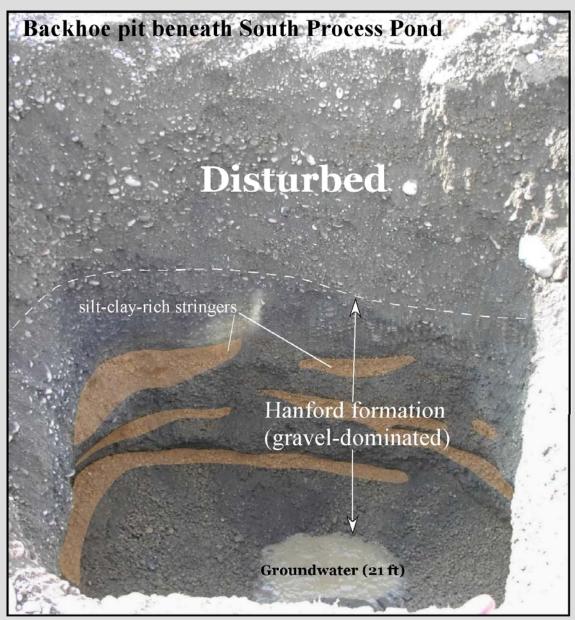


Associated Practical Issues

- 1. Accurate projection of dissipation times for groundwater plumes of sorbing contaminants
 - Sorbing solutes not equal
 - Concentrations at different scales
- 2. Optimal delivery of remediation reactants
 - Access
 - Kinetic formation and reaction
 - Persistence
- **3**. Practicality and effectiveness of remediation



Heterogeneity and Mass Transfer Domains in 300 A Vadose Zone Sediments





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Representative Facies from LFI Cores







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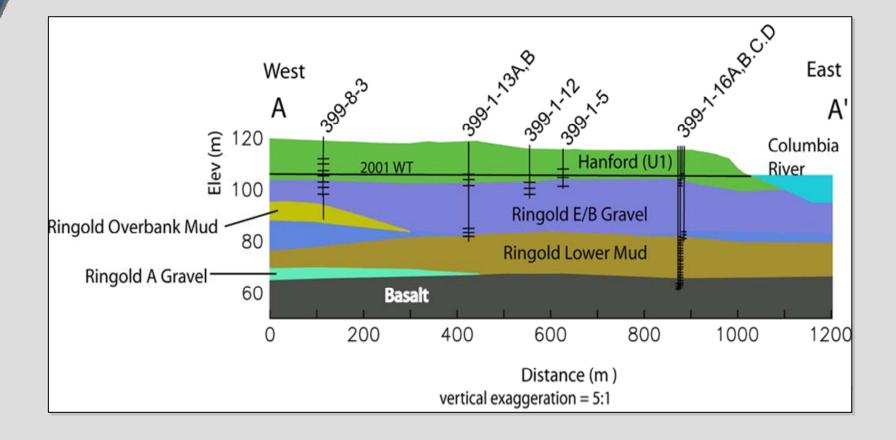
Primary Objectives

- Quantify the role of mass transfer in controlling U(VI) distribution under various geochemical, hydrologic, and remedial conditions
 - Vadose zone
 - Saturated zone
- Investigate in-situ microbiologic processes that couple with mass transfer to control U(VI) fate, transport, and remediation
- Create enduring field experimental data sets for model and fieldscale hypothesis evaluation
- Test and improve existing models of multi-reaction chemistry and multi-scale mass transfer by comparison to new, robust experimental field data
- Proactively transfer results to site for decision making and remediation



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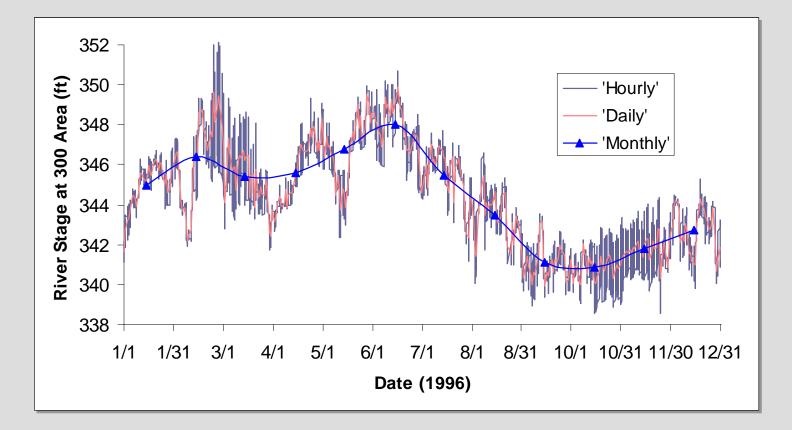
Geological Cross Section





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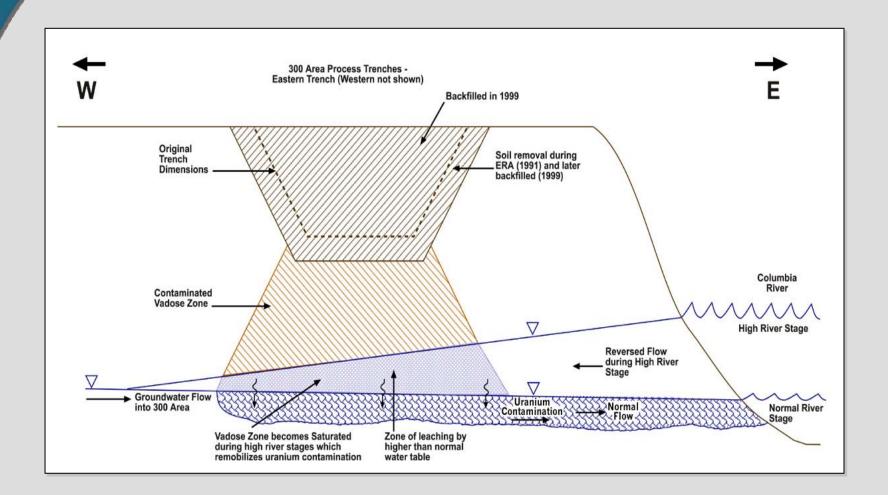
Hourly, Daily Average, and Monthly Average River Stage at the 300 Area in 1996





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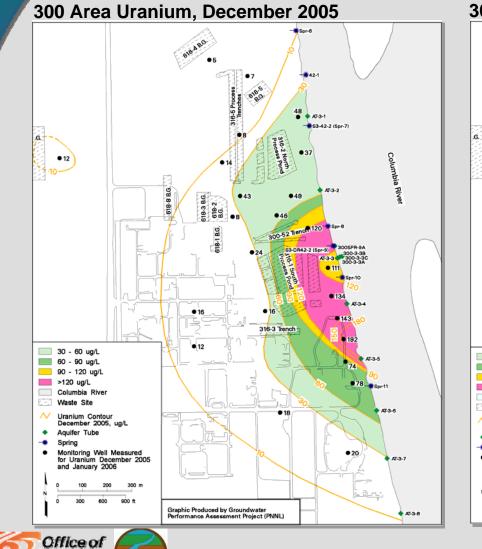
Vadose Zone Release Model





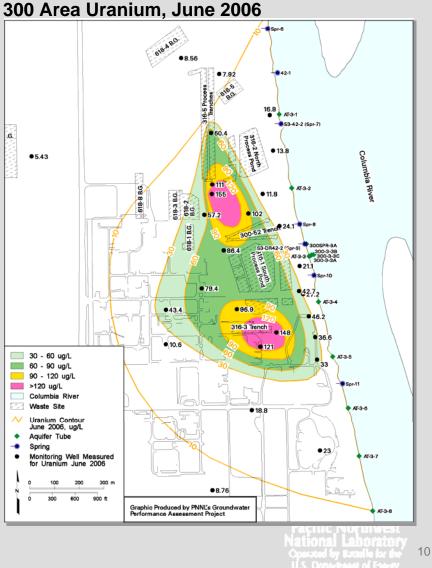
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Seasonal Dynamics of 300 A Uranium Plume



Science

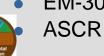
U.S. DEPARTMENT OF ENERGY



Approach

- Robust 3-D geostatistical characterization of the experimental domain
 - Borehole samples and geophysics
 - geo-, hydro-, chemo-, bio-, and U(VI)-facies
 - Correlative transfer functions with key process-specific parameters
- Field experimental campaigns based on 3 hypothesis at an integrated vadose zone-saturated zone site
 - Well field sufficient to sample heterogeneities
 - Infiltration experiments in vadose zone
 - Passive river stage experiments in capillary fringe
 - Injection experiments in saturated zone
 - Collaborative experiments with EM-20
- Modeling of different types
 - Stochastic-deterministic
 - STOMP, MODFLOW, and FLOTRAN by code originators
 - STOMP as the integrative project code
- Leverage broad data base and other site activities
 - ERSD
 - EM-30, EM-20





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Summary Accomplishments

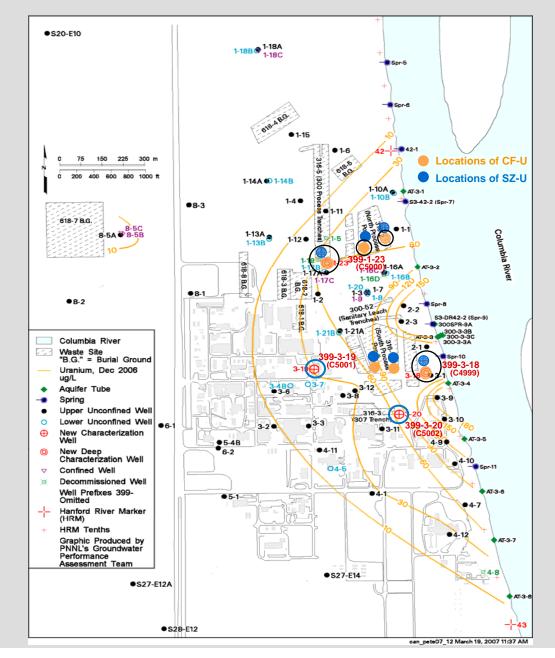
Subcontracts with external collaborators are being finalized.

- Project Management, Field Site Management, QA/QC, and Health and Safety Plans, and NEPA documentation completed.
- Web-Site under development with problems.
- Criteria established for site selection. Geophysical surveys to aid in site placement in SPP completed. Collaboration with EM-40 on SPP borehole provides data and "free well".
- Wells nearby SPP instrumented for continual monitoring to improve site hydrological model.
- Premodeling of tracer experiments performed to aid in site design and monitoring array configuration and spacing.
- Design of experimental site and monitoring system completed; drilling to begin in January.
- Characterization and Field Experimental Plans underway in response to final site design.
- Infrastructure for injection experiments identified and needed equipment is being procured.
- Data base management task initiated. Prototype data sets for geology, geochemistry, hydrogeology, and geophysics input to data base.
- Coordination with EM projects established.





Documented Locations of Solid-Associated U





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The 300-FF-5 Operable Unit

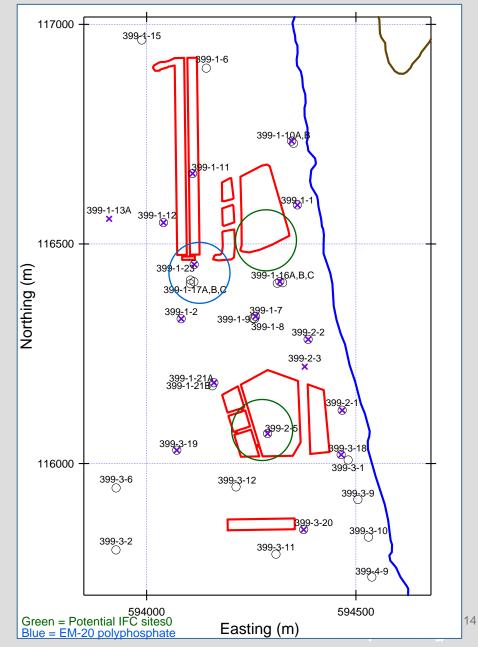
Locations of CERCLA monitoring wells (open circles) and wells instrumented for automated hourly measurements (purple dots)

- water levels
- temperature
- EC

IFC site selection strongly dependent on polyphosphate plume trajectory

- Model projections performed
- Continuous monitoring initiated around SPP to improve the hydrologic model



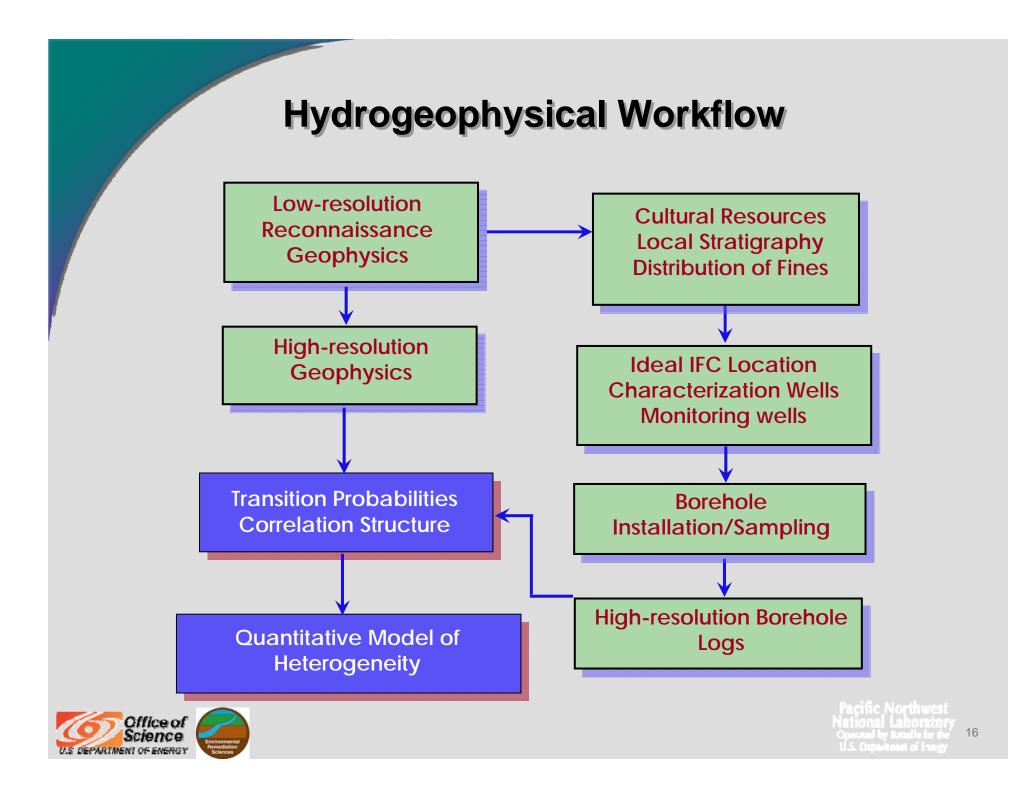


Criteria for Site Selection

- Seasonal changes in [U] ~ 2; [U]max > 50 ppb; no organic contaminants
- Proximate to previous excavations for which significant laboratory and characterization data exists
- Maximal amount of fines in saturated zone
- Near but out of the zone of influence of the EM-20 polyphosphate injection experiment
- Saturated zone thickness (Hanford formation) of 2-3 m
- Relatively flat Hanford-Ringold contact to minimize vertical gradients
- Located within coverage domain of existing groundwater monitoring domain
- Site location (and experiment timing) to allow relatively slow and predictable travel times



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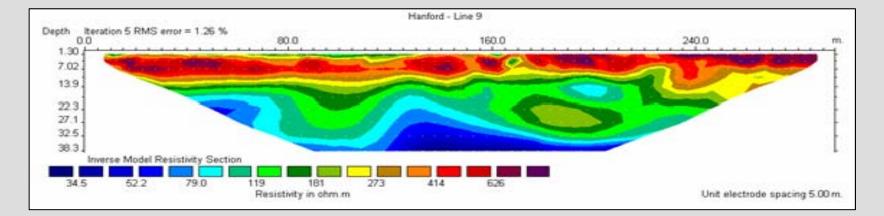
Geophysical Lines for Initial Characterization and Site Selection

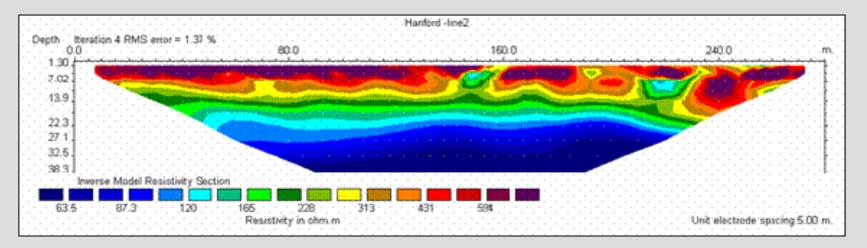




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IFC Site Location – Geophysical Exploration of the Hanford-Ringold Contact

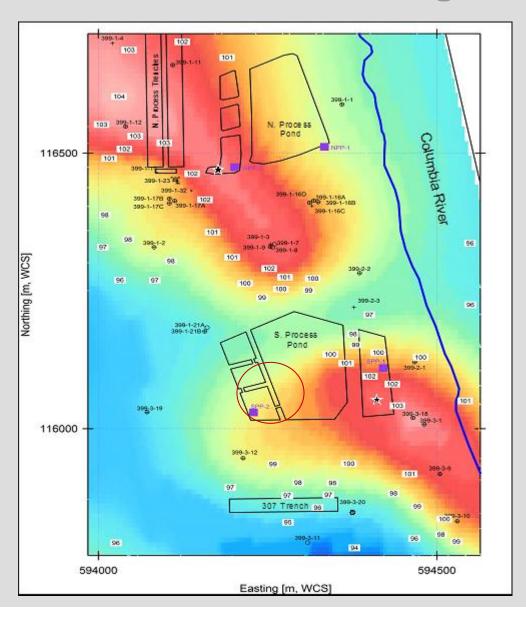






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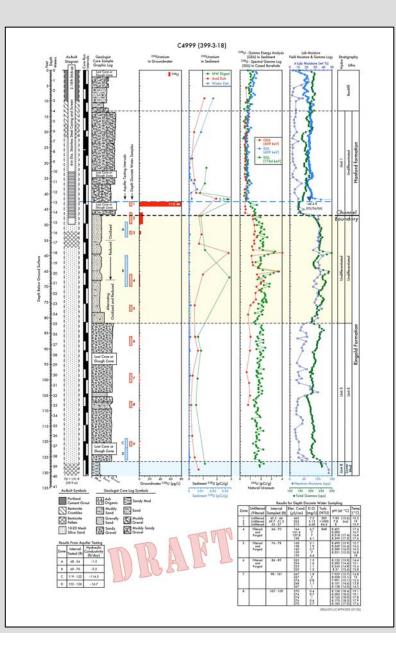
Shaded Relief of Hanford Ringold Contact





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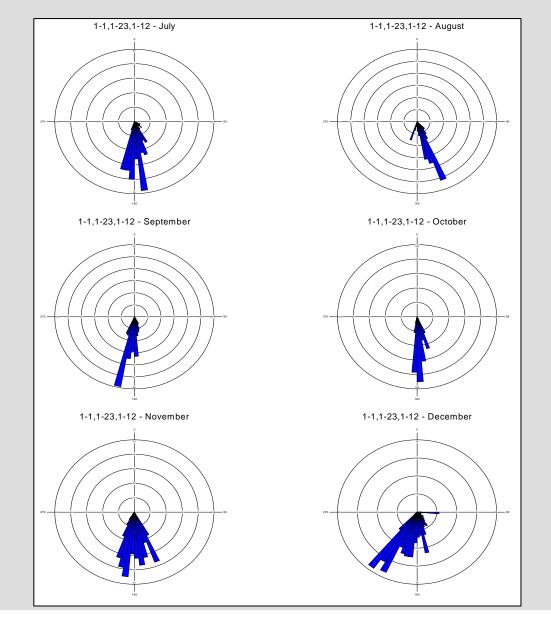
LFI Borehole Results East of SPP





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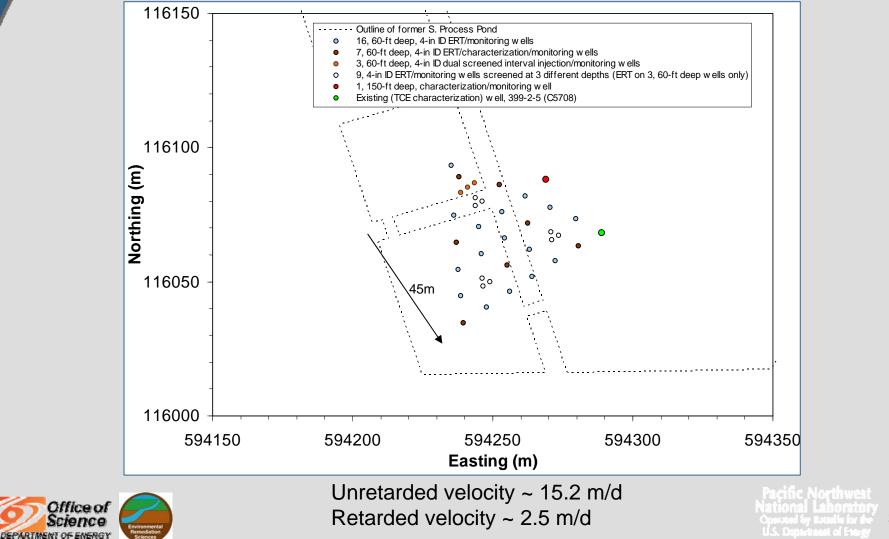
Seasonal Changes in Groundwater Flow Vectors



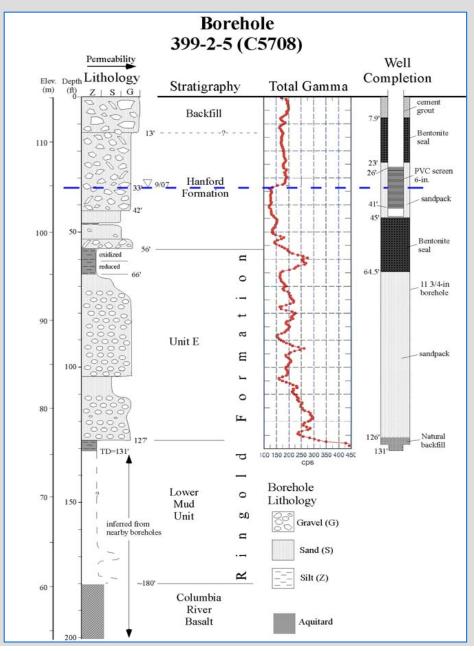


Planned Layout of Hanford 300 Area IFC **Well Array**

[Site installation to begin after Jan. 14, 2008]



Geology Log for New SPP Borehole

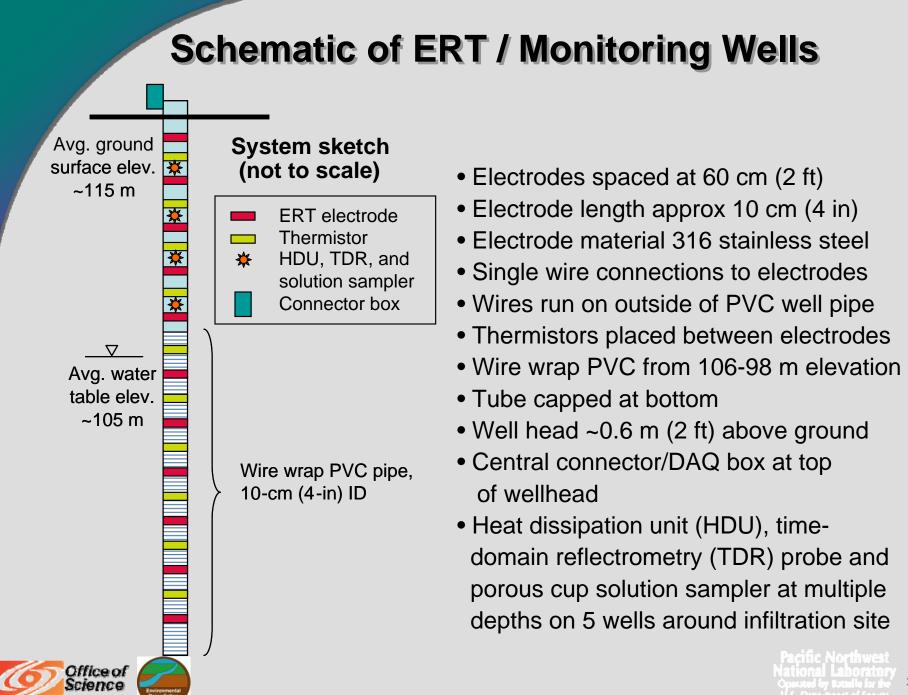


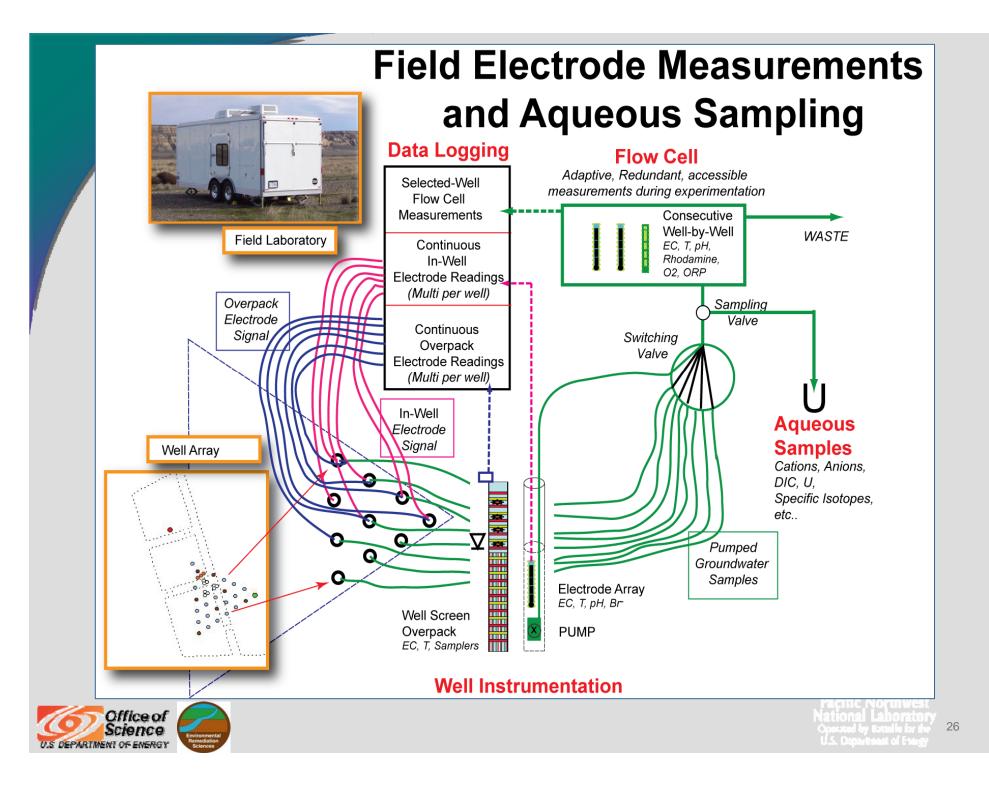


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Weak-Acid Extractable U(VI) from C5708

Formation	Notes	Depth (ft) 4.5-7	U(μg/g) 3.4
BF			
HF	vadose zone injection depth	15-17	1.03
		20-22	1.24
		23.5-26	3.09
	smear zone	28-31.5	5.17
	water table	32-33	3.29
	upper screen 🛓	33.8-36.8	0.99
	middle screen	40.8-42.8	0.93
	÷	45-47	0.64
	lower screen	49.5-51.5	0.56
RF	C.W. upper screen	55-56	1.43
	C.W. lower screen	73-75	0.58
		74-76	0.57





Deep Characterization Borehole

- Approximately 150' in depth to sample Ringold Formation
- Major focus is microbiological characterization (aspectic sampling)
- To be drilled and analyzed in collaboration with (shared support from) evolving PNNL SFA
- Located along east margin of IFC experimental site
- Seeking variance from WA. Dept of Ecology to permit two screened intervals in Ringold
- Will allow passive biogeochemical and mass transfer studies within Ringold transition zones
- Possible evaluation of diffusive mass transfer from the Ringold as a U source
- Details to follow as SFA planning evolves



Web-Site Status

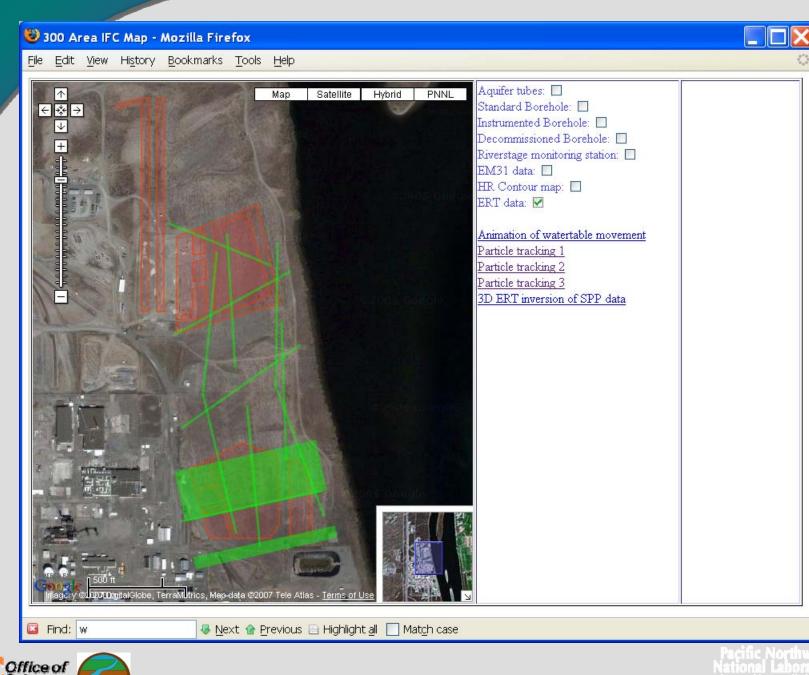
- The ORNL FRC Web-Site served as an initial model for the Hanford IFC Web-Site.
 Changes in organization and content were proposed for the Hanford IFC site to serve differences in targeted users and project needs.
- The Hanford IFC Web-Site was designed and populated with background information of different types including a bibliography and numerous project presentations; vita of project participants; project management, QA/QC, Health and Safety, and other plans; project planning documents including drilling specs and experiment premodeling; summaries of available data; site pictures and maps; etc.
- A number of issues have been encountered regarding PNNL policies on document clearance, and related subjects that have been problematic.
- The IFC Web-Site was submitted to the internal PNNL review committee 3 weeks ago. The Web-Site was denied for open release.
- The Hanford IFC Web-Site team has been expanded to include another technical editor and is responding to recommendations and criticisms from the committee. A revised Web-Site will be submitted to the internal review committee in approximately 3 weeks.
- We will evaluate our path forward after the second review.



Evolving Project Data Base

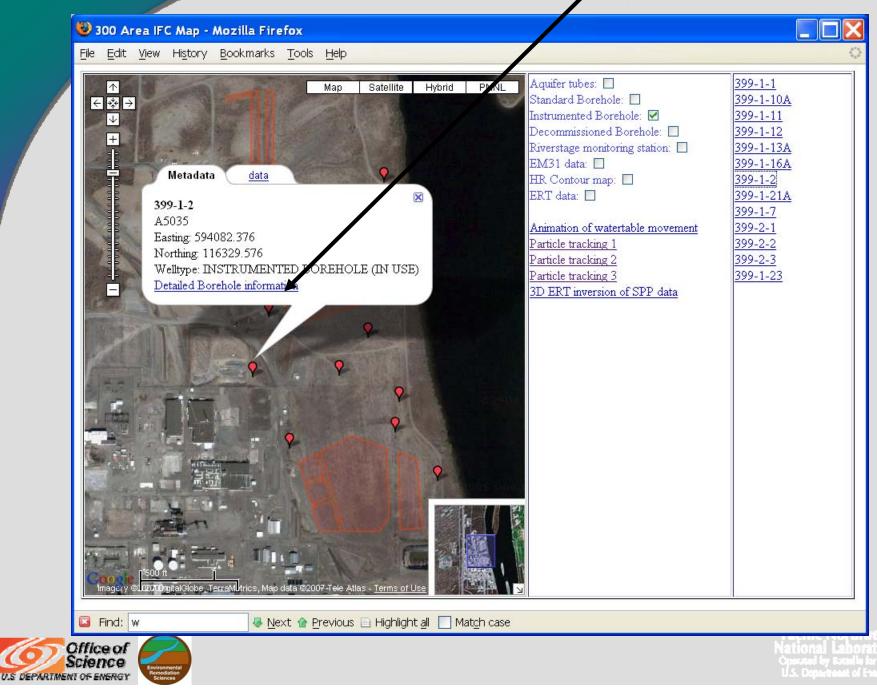
- Uses Google Map API (v2) to provide lightweight GIS access to data
- Not meant as a full fledged GIS, but good for rapid result distribution
- ► Has aerial imagery of 300-FF-5 as optional underlay
- Allows rapid oversight of data + access to data
- Currently being coupled to back end (HEIS, water level data) and new project-specific databases
- Coupling to test borehole database is operational (note: no logs/samples in this database yet)
- URL: <u>http://geophysics.inel.gov/IFC/ifcmap.htm</u>



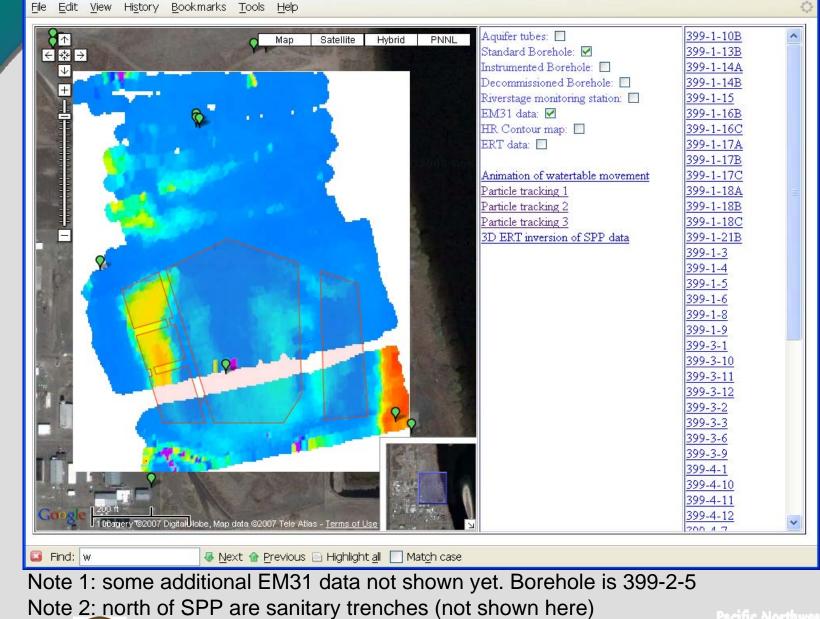


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Click here to query database for well



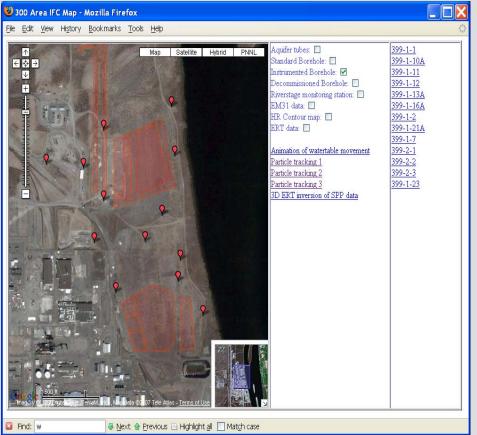
😻 300 Area IFC Map - Mozilla Firefox



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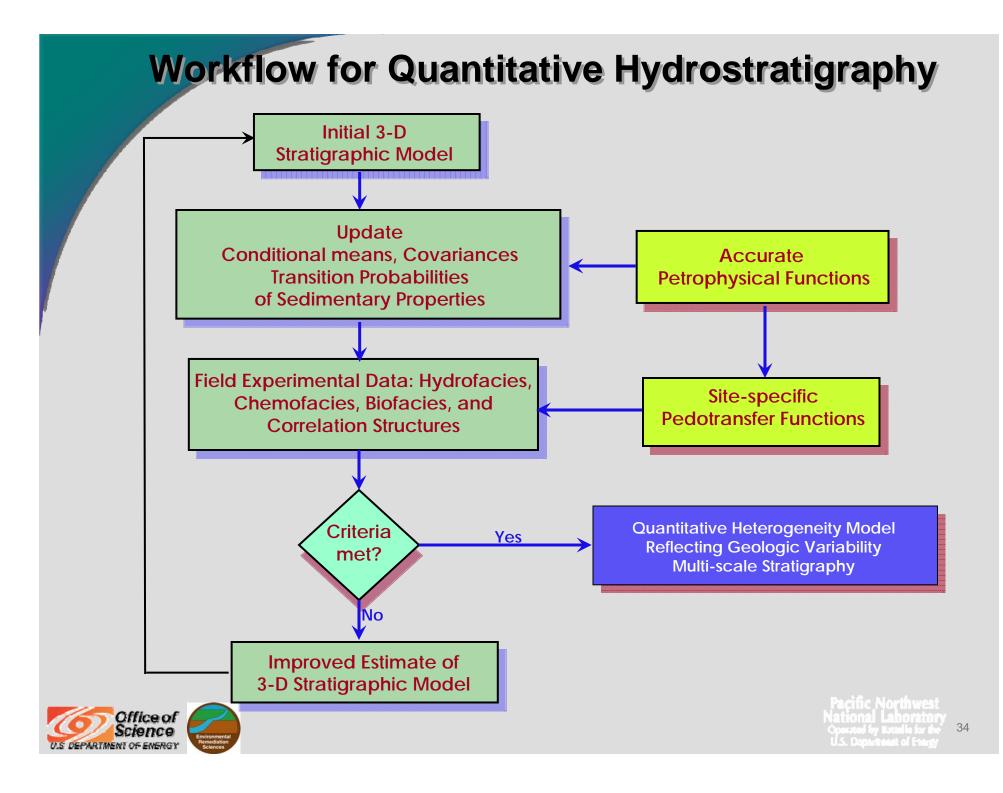
Water Level Data

- 13 instrumented wells in 300 area
- Processed to give groundwater direction by fitting water levels to best plane
- Excellent correlation to river stage
- Confirms general understanding, also points to variability in flow direction
- Basis for site 3-D hydrologic model









High Resolution Geophysics

- 1. Cross-hole ERT
 - Interpolate lithology between wells (design to allow with stationary downhole electrodes)
 - Track recharge and plume movement (water content, and salt contrast)
- 2. Cross-hole radar
 - Monitor changes in water content
- 3. High resolution seismic (surface)
 - Lithology and porosity
 - Experimental plot (1/08)
 - Larger scale features (spring 08)



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Geochemical and Mineralogic Characterization

- Characterization wells (continuous cores by sonic, 420' of core depending on recovery)
 - Sand and silt/clay mineralogy
 - Extractable Fe forms (DCB, am-ox), calcium carbonate or TIC
 - Total, extractable, and isotopically exchangeable U
 - U adsorbability and adsorption/desorption mass transfer
 - Particle size distribution (< 2 cm, > 2 cm)
 - Surface area of < 2 mm fraction
- Monitoring wells (1-2' grab samples by Becker hammer, ~ 840 samples)
 - Various U forms (total, acid- and bicarbonate-extractable)
 - Extractable Fe or other surrogate of U adsorptivity
 - Particle size distribution of < 2 cm materials



Microbiologic Characterization of Deep Borehole Sediments

Microscopic

- Direct counts (w SYTO and PI) and active cells (TDR)
- Culture independent
 - Phylogenetic diversity/richness
 - Real time PCR for specific functional groups
 - Phylo & functional gene arrays (Geochip)
- Culture dependent
 - Liquid MPN's with various TEA's
 - Filter based cultivation
 - Isolation & characterization of representative cultures
- Activity analysis
 - Targeted incubations w/ radiolabeled substrates



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Materials Available for ERSD Researchers

Existing

- Historic, highly contaminated sediments from process ponds (small masses available)
- Excavation samples from the vadose zone at two locations in the North Process Pond (NPP) and two locations in the South Process Pond (SPP). Samples contain various U(VI) speciation states, including adsorbed, precipitated, and surface complexed phases (variable sample masses are available).
- Uncontaminated vadose zone and aquifer sediments from the EM-40 Limited Field Investigation (LFI)
- Low-level contaminated samples from SPP C5708

To be Collected in Jan - Feb

- Becker-hammer grab samples from monitoring well installation in the Hanford formation screened to < 2 cm (~ 100 samples saved for ERSD researchers)</p>
- 4" continuous sonic core samples from 7 characterization boreholes in Hanford sediment (~ 75-100' saved for ERSD researchers)
- 4" continuous sonic, aseptic core samples from one -150' characterization borehole through the Hanford and Ringold formations (select sample aliquots and undisturbed cores will be saved for ERSD researchers)

<u>Other</u>

An excavation will be opened below backfill (~15 ') to allow bulk sample collection and in-situ structural analysis



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IFC Experiments

U(VI) fluxes from the vadose zone

- Scale-dependent mass transfer, geochemical kinetics (adsorption/desorption) and water pathway effects on U(VI) fluxes to groundwater
 - Infiltration experiments with varying water application rates, volumes, and composition (pH, HCO₃, Na/Ca)
 - Passive experiments to explore rising and falling water table effects on U(VI) solubilization and release from lower vadose zone

► U(VI) concentration dynamics within the groundwater plume

- Scale-dependent mass transfer involved in forward (adsorption), backward (desorption), and steady-state (isotopic exchange) reaction processes in flow paths with different trajectories and residence times
 - Injection experiments with varying HCO₃ and U(VI) concentrations, and U(VI) isotopic ratios
 - Passive experiments follow vadose zone pulses, or inland riverwater groundwater gradients

Optimized and sustained remediation strategies

- Evaluate role of mass transfer and microbiological processes on different forms of phosphate used to precipitate and immobilize U
 - Injection experiments with polyphosphate, Ca-citrate/PO₄³⁻, organic P with HCO₃
 - In collaboration with EM-22 and team



Large Column Experiment with NPP1-14 to Investigate Scaling of Reaction and Mass Transfer Parameters

Unseived Sediment with River Cobble and Mud



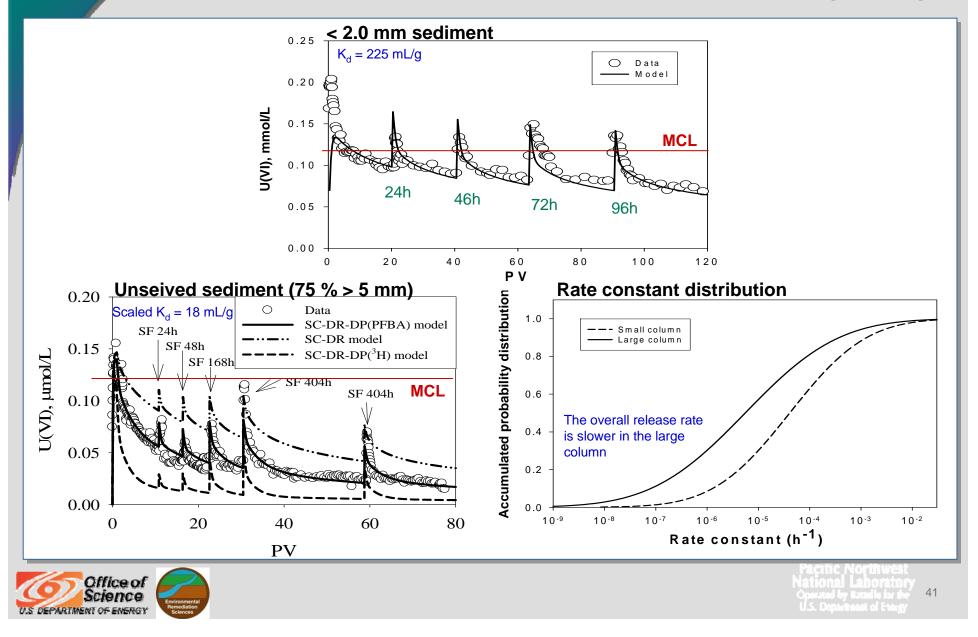
The 80 kg Column



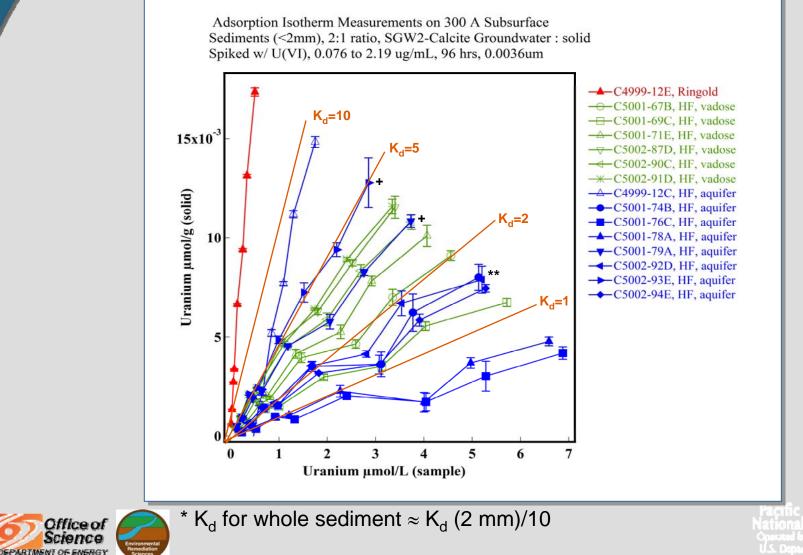
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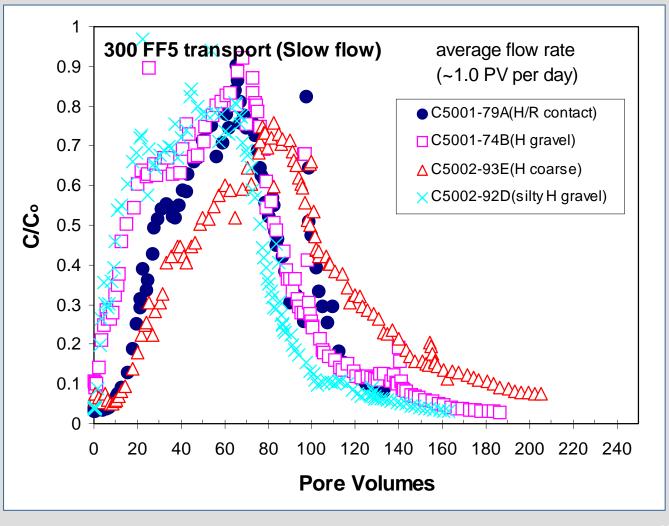
Small and Large Column Results and Rate Constant Distribution from the Distributed Rate Model (DRM)



U(VI) Adsorption Isotherms on Uncontaminated **300** A Vadose Zone and Aquifer Materials from **LFI** Cores



One Dimensional Reactive Transport Behavior of U in 300 A Aquifer Sediments





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Injection Experiments for Hanford IFC

Subsurface Zone (Number of Experiments)	Experiment	Priority/FY	Targeted Process
Saturated Zone	Tracer 2 Flowpaths	1, FY08, FY09	Saturated zone flowpaths, dispersion. Multiple tracers (PFBA, D ₂ O, fluorescent organics, Br ⁻), salinity contrast.
	²³³ U 2 Flowpaths	2, FY09	Uranium isotope exchange along flowpath. Multiple tracers, <i>in situ</i> groundwater (if permittable).
	- Uranyl 2 Flowpaths	2, FY09, FY10	Uranium desorption along flowpath. Multiple tracers, groundwater from appropriate source, including two solution compositions.
	+ Uranyl 2 Flowpaths	2, FY10	Uranium adsorption along flowpath. Multiple tracers, groundwater from appropriate source, including two solution compositions.
	River Water 1 Flowpath	3, FY10	Complexation Effects. Multiple tracers.
	Polyphosphate	TBD	Mass transfer effects on autunite and apatite precipitation (TBD).
Vadose Zone	Tracer 1 Flowpath	1, FY08	Vadose zone flowpaths, dispersion, mixing with GW. Multiple tracers, low ionic strength, salinity contrast.
	- Uranyl 2 GW Flowpaths	2, FY09	Dissolution and/or desorption within the vadose zone and migration within the saturated zone. Multiple tracers, high ionic strength, variable injection rate.
	+ Uranyl 2 GW Flowpaths	3, FY10	Adsorptive retardation within the vadose zone and migration within the saturated zone. Multiple tracers, variable solution compositions (U, HCO_3), variable injection rate.
Capillary Fringe	One-Well Injection 2 Flowpaths	TBD	Depth of influence on saturated zone, vertical dispersion along flowpath. Multiple tracers, variable solution compositions.
	Multi-well Injection 2 Flowpaths	TBD	Depth of influence on saturated zone, vertical dispersion along flowpath. Multiple tracers, variable solution compositions.



Major FY08 Goals and Milestones

- Open an operational web-site with data base linkage
- Distribute multi-year characterization and experiment plan
- Complete well-field installation and testing
- Make significant progress toward 3-D geostatistical model of experiment site
- Integrate modeling team
- Perform two tracer injection experiments
- Develop initial publication plans



IFC Experiments will Help Resolve Lingering Conceptual Model Uncertainties Over Next 2-3 yr

Seasonal concentration dynamics

- Predominance of hydrologic (dilution, trajectory, elevation) versus geochemical effects (water composition change, residence time)
- In-situ retardation and Ringold channel effects

Sources of sustained U in groundwater

- Contributions of upper/lower/ and smear zones (direct measurements of flux and groundwater mixing)
- Aquifer solids (measurements of sorbed U and its exchangeability along different flowpaths with different chemistry)
- Slow dissipation of plume
 - Field scale desorption rates from smear zone and aquifer (measurements and modeling of mass transfer at different scales)
 - Fluxes to river (U isotopic specific measurements IFC/ERSP, higher density monitoring and improved modeling)



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Anticipated Outcomes

- Outstanding, multidisciplinary collaborative effort that significantly advances science
 - Characterization, experiment design, interpretation
 - Basic underpinnings of EM-20 activities
- Enduring and accessible field experiment data sets for hypothesis and model testing
- Improved linked multi-scale mass transfer/biogeochemical models for reactive contaminants
- New conceptual understanding of mass transfer processes at different scales influencing field behavior
 - Desorption, dissolution, dissipation
 - Effective reaction kinetics
 - Contaminant immobilization



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