



Hanford 300 A IFC

Office of Science/Environmental Remediation Sciences Division (ERSD) Supported Research at Hanford: The PNNL Scientific Focus Area (SFA) and Integrated Field Research Challenge (IFRC)

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River & Plateau Mtg
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Pacific Northwest
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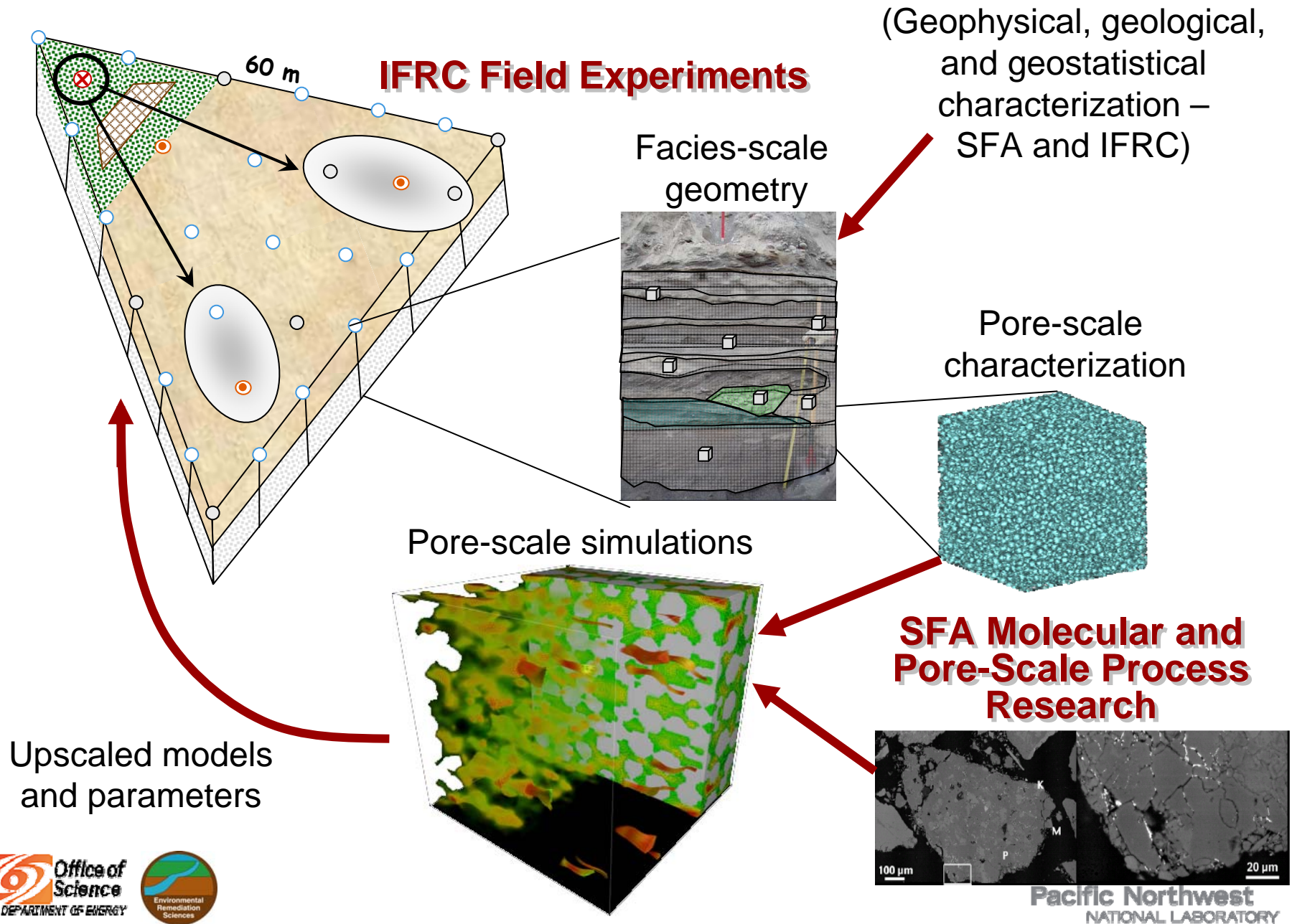
Environmental Remediation Sciences Division (ERSD)

- ▶ Located within Office of Biologic and Environmental Research (OBER) in the Office of Science.
- ▶ Focus on fundamental environmental science, fate and transport primarily (i.e., EMSP).
- ▶ Seek positive impact on clean-up progress through knowledge generation, process understanding, and advanced measurement/analysis techniques.
- ▶ Steward of PNNL's Environmental Molecular Sciences Lab (EMSL) a user facility with budget > \$25M.
- ▶ Traditional emphasis on environmental microbiology (e.g., NABIR).
- ▶ Primary product is peer reviewed publication and scientific insights to solve cleanup challenges.

Hanford Inspired, ERSD Funded Environmental Research at PNNL

	<u>Scientific Focus Area</u>	<u>Integrated Field Research Challenge</u>
Contact	www.pnl.gov/biology/sfa	www.ifchanford.pnl.gov
Funding	6.5M	3.0M
Science Theme	Role of microenvironments and transition zones on U, Tc, and Pu fate & transport	Influence of multi-scale mass transfer on U persistence and migration in groundwater
Scope	Lab based with model systems and Hanford samples. Some use of IFRC and other sites as field laboratories. Strong emphasis on biogeochemistry, coupled reactive transport processes, and molecular-microscopic-macroscopic interactions	Field-based with emphasis on i.) large scale processes and interactions controlling U concentrations in groundwater, ii.) characterizing controlling factors and their variation, iii.) field experiments of different type, and iv.) geostatistical and reactive transport models
Internal Investigators	12	7
External Collaborators	11	7
Origin	Reprogramming of competitively awarded ERSD projects in FY08	FY07 award in response to ERSD competitive proposal solicitation

PNNL ERSD Research



Scientific Focus Area (SFA) Concept

- ▶ Independent, competitively awarded projects (12), reformulated to be collaborative and to address a focused suite of state-of-science issues across different spatial and time scales.
- ▶ Guidance provided by BER/ERSD on SFA scope and unique expertise areas.
- ▶ Hanford-inspired science theme (microenvironments and transition zones), research topics, and contaminants.
- ▶ Close alignment with Hanford 300 Area Integrated Field Research Challenge, use of EMSL and other unique DOE capabilities.
- ▶ Team includes those with detailed knowledge of Hanford science issues (over 40 publications with Hanford Site impact).
- ▶ Concept allows PNNL to manage research team for maximum collaboration and synergistic impact – funding allocations made by PNNL science PI.
- ▶ Research to be sufficiently fundamental for application to other DOE contaminated sites and environmental problems.

SFA Contaminant Emphasis – Hanford Drivers

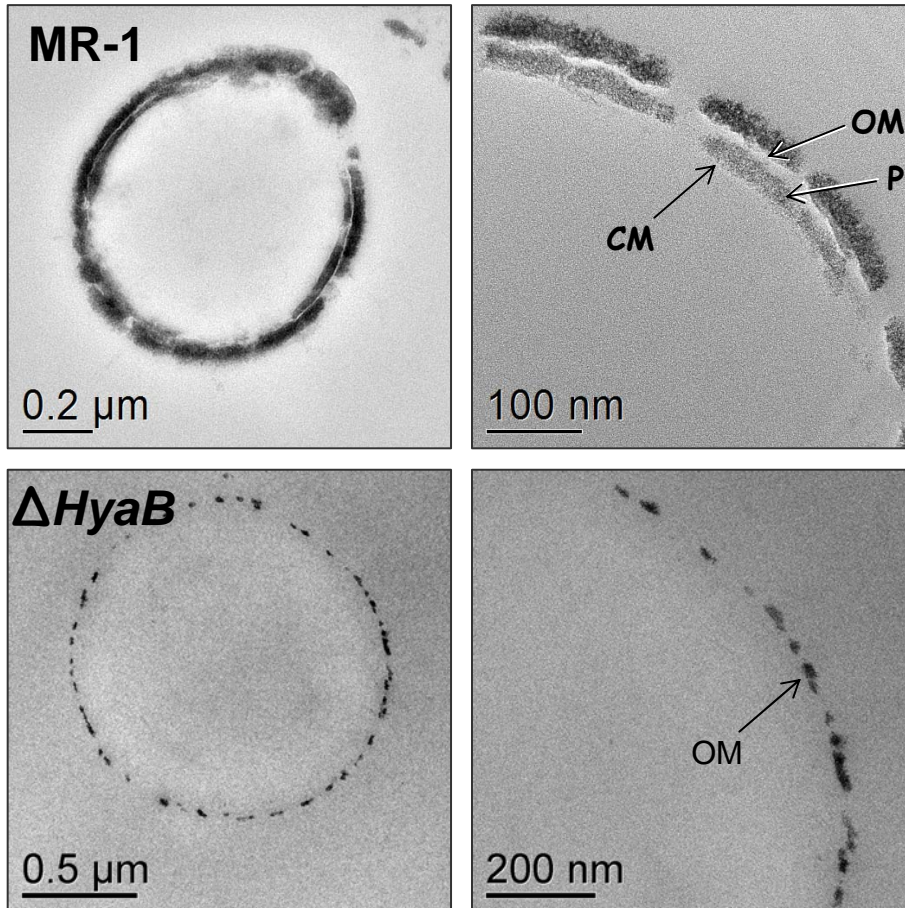
- ▶ Risk drivers on the Hanford site:
 - U, ^{99}Tc , ^{129}I , Cr, and CCl_4 – environmental mobility and persistence
 - $^{239,240,241}\text{Pu}$, ^{137}Cs , and ^{90}Sr – lower mobility but high radioactive toxicity

- ▶ Initial SFA research focused on U, ^{99}Tc , and Pu
 - Polyvalency with complex biogeochemistry
 - Past releases to soil
 - U = 202,703 kg, ^{99}Tc at 1390 Ci, and Pu at 400 kg
 - Long term concerns and scientific issues
 - Important science opportunities

[Cr investigated by LBNL-SFA; collaborative studies with PNNL SFA on CCl_4 mineral transformations supported by BES]

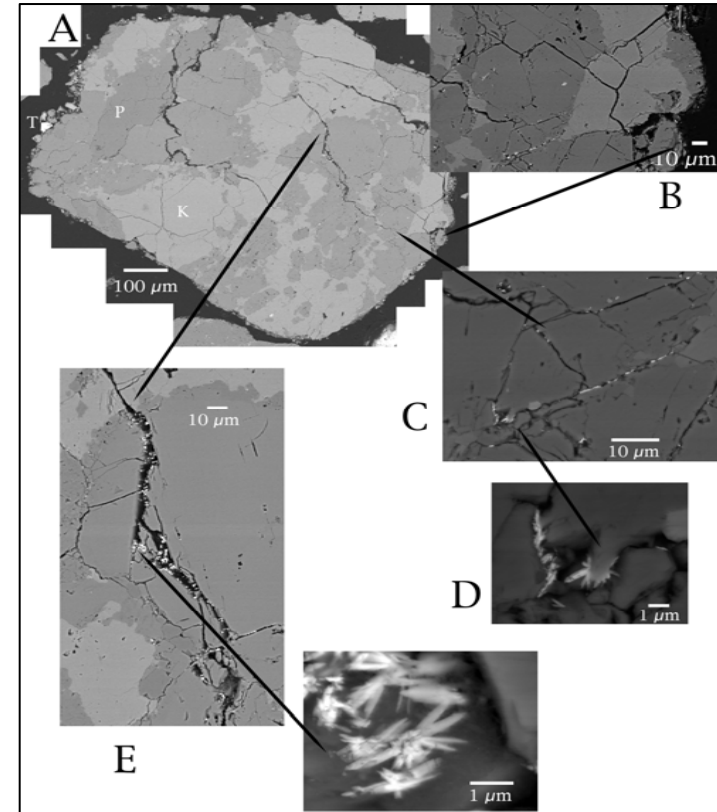
Microenvironments – Disproportionate Influence on Chemistry

Biogenic TcO₂



(Marshall et al. 2008, *Environ. Microbiol.*)

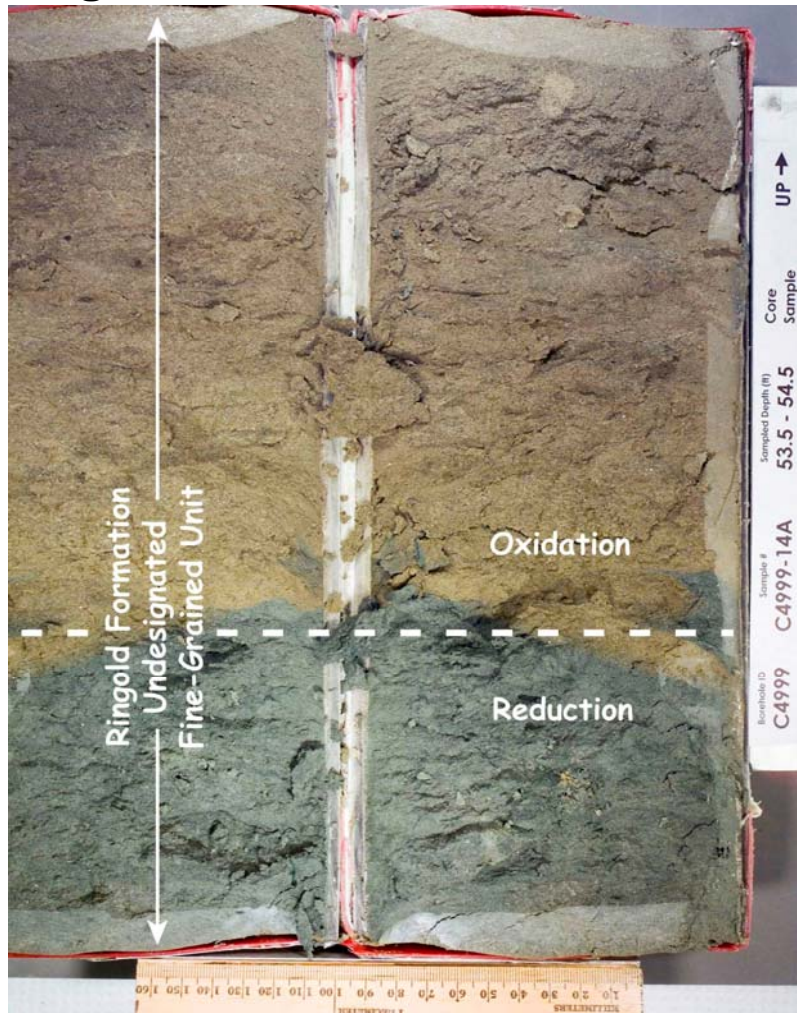
Intragrain U(VI) Precipitates



(McKinley et al. 2006, *GCA*)

Transition Zones – Exhibit Chem-Phys-Bio Changes Over Short Distances

Ringold Formation Redox Boundary



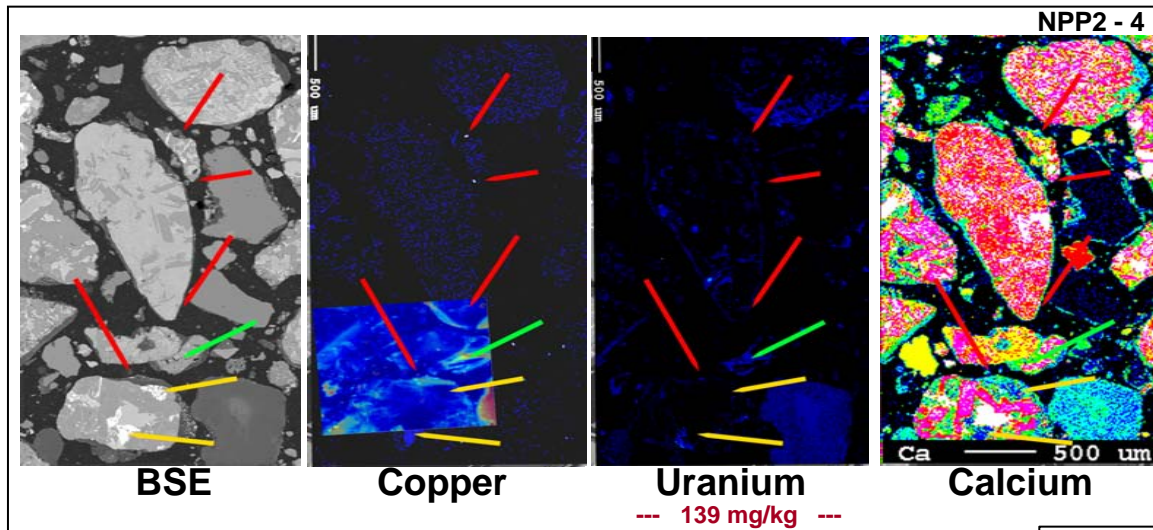
Columbia River Hyporheic Zone



(Moser et al. 2003, ES&T)

Microscopic Speciation Controls Macroscopic Release Behavior of U

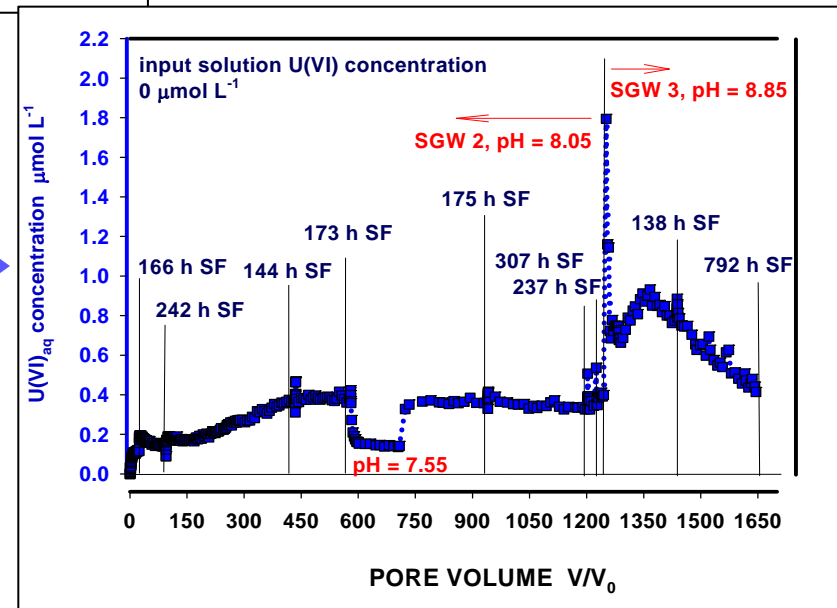
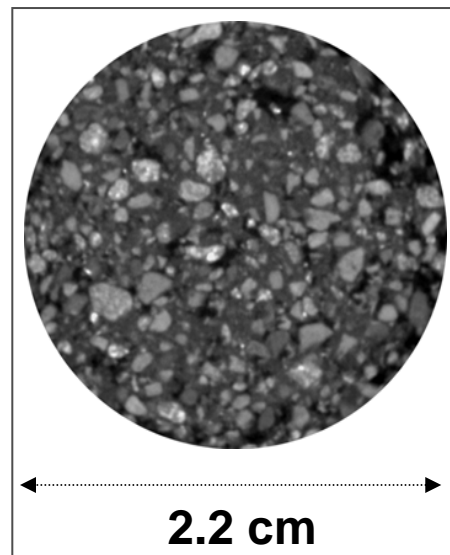
Metatorbernite (uranyl copper phosphate) in 300 A grain coatings



- ▶ Speciation model developed from EXAFS, S-XRD, and TEM/EPM
- ▶ Thermodynamic and kinetic model developed from studies of pure phase metatorbernite
- ▶ Column study integrates multiple pore-scale processes; provides data to evaluate whether metatorbernite controls solution concentrations from waste-impacted sediments

Column study to investigate mass-transfer controlled dissolution

(Catalano et al. 2006, Arai et al. 2007, Zachara et al. 2008)

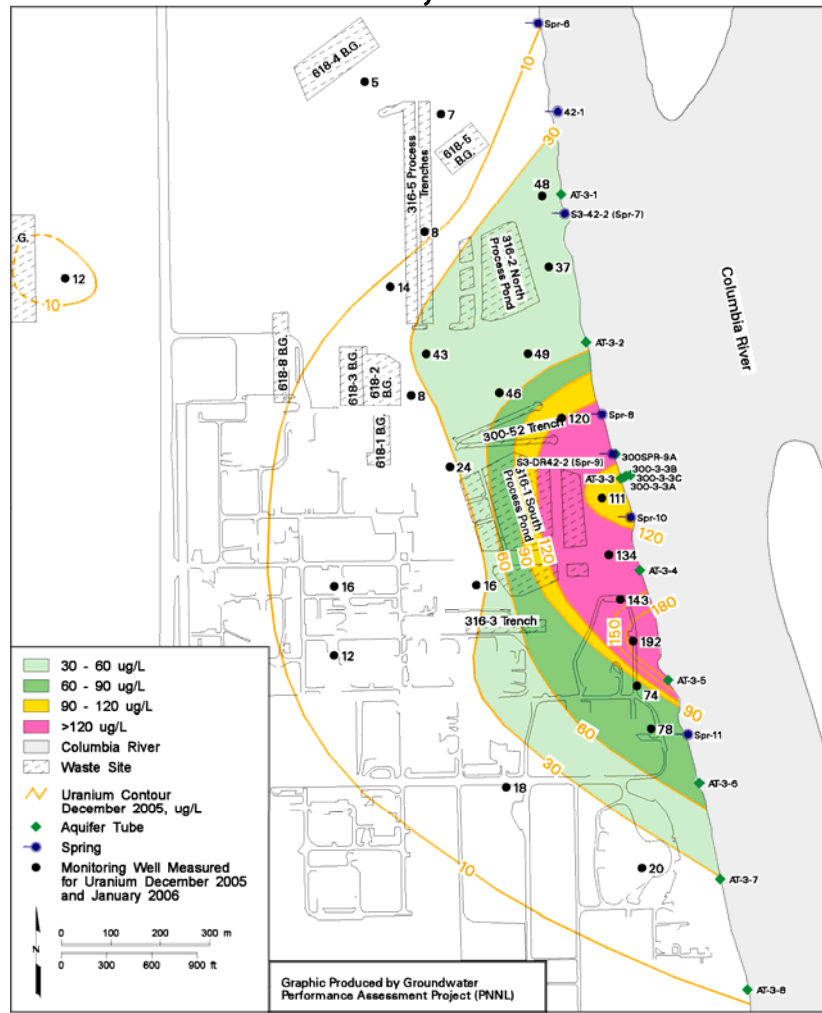


Example SFA Research Topics FY09 - FY10

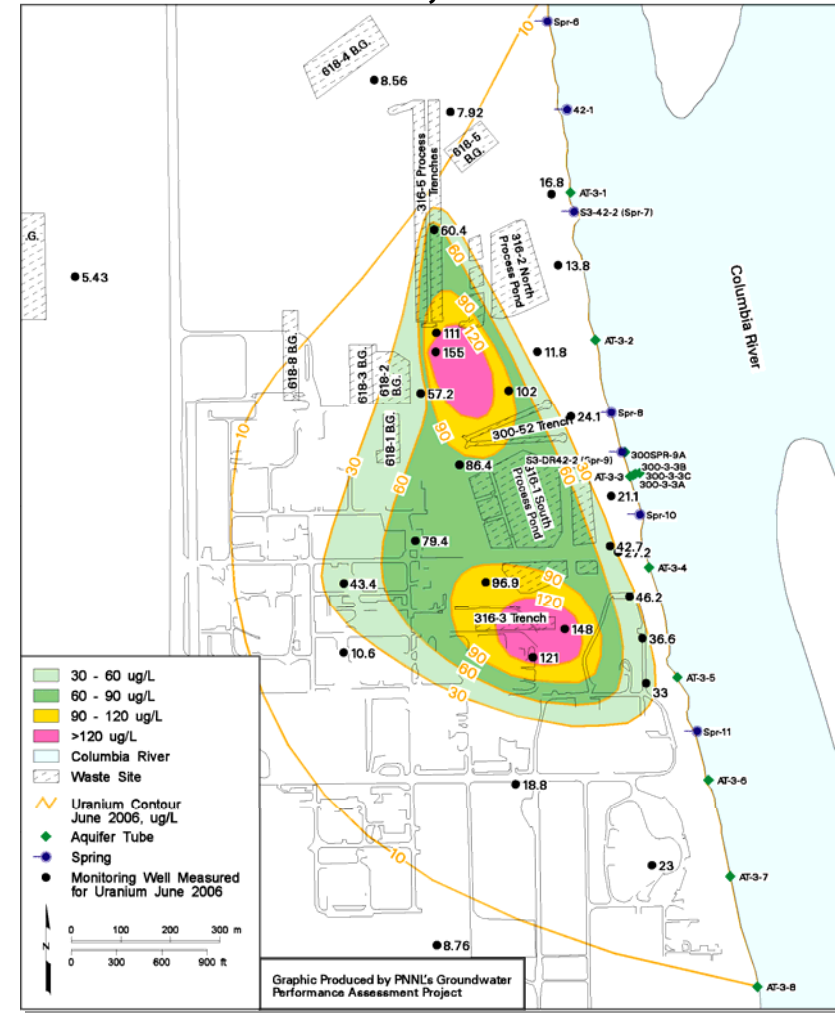
- ▶ Microbiology of 300 A unconfined aquifer.
- ▶ Biogeochemistry of 300 A microbial isolates toward Tc and U under microaerophilic conditions.
- ▶ Redox geochemistry of ^{99}Tc in 200 A and 300 A Ringold Formation sediments.
- ▶ Intragrain microscopic transport processes of U and Tc in different Hanford sediment facies.
- ▶ Reactive transport behavior and models of contaminant U in intact IFRC sediment cores.
- ▶ Pu molecular speciation and mobilization reactions in sediments beneath Z cribs.
- ▶ Pore scale and continuum reactive transport models.

Seasonal Dynamics of 300 A Uranium Plume

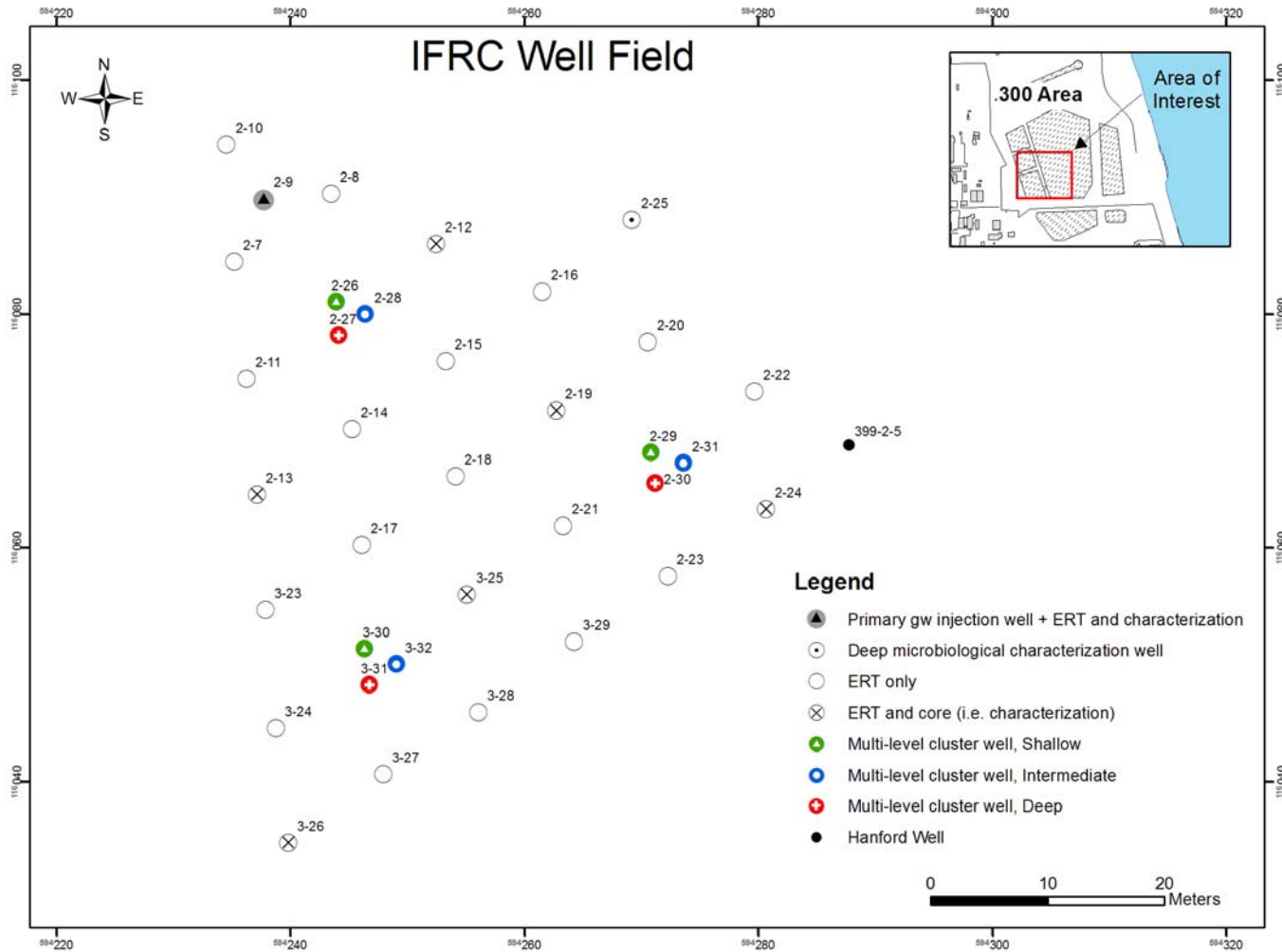
300 Area Uranium, December 2005



300 Area Uranium, June 2006



Hanford Integrated Field Research Challenge Site

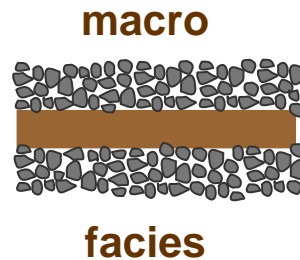
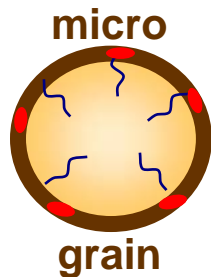


Coordinate System: Washington State Plane South, NAD83, in meters

Map Document: (C:\Documents and Settings\d3m608\My Documents\FY08\IFC\SurveyData\IFC_Basemap_rev3.mxd)
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Hanford Integrated Field Research Challenge Science Theme

Multiscale mass transfer processes influencing sorbed contaminant migration

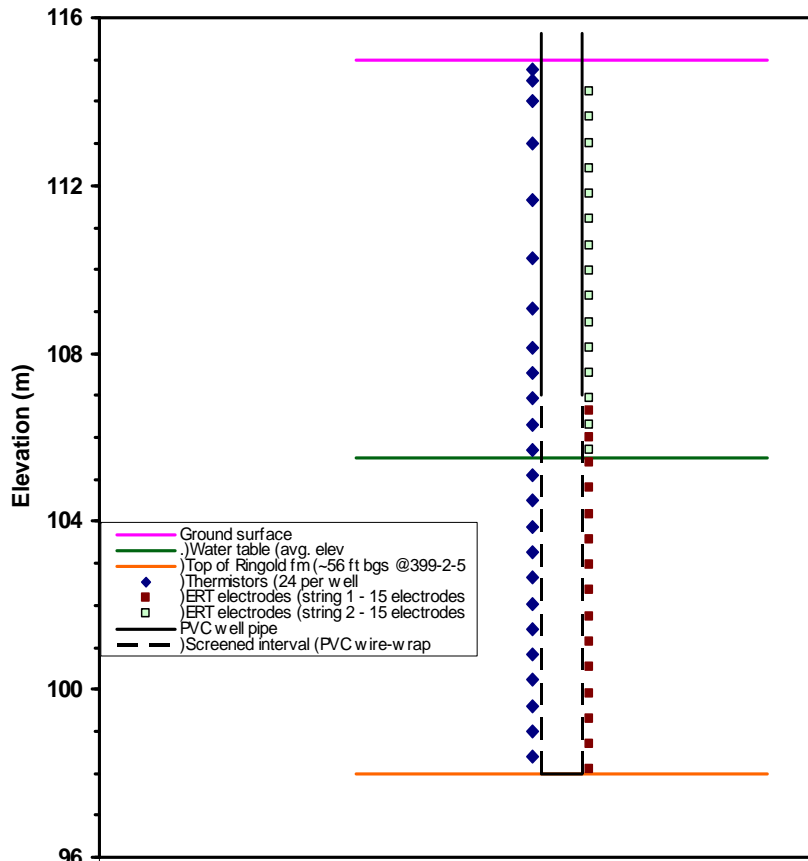


- ▶ Mass transfer is controlled by diffusion, and is influenced by the path length, tortuosity, thickness, and surface charge of immobile, water-filled pore space.
- ▶ It controls contaminant release at the particle scale from intragrain domains to porewater, and at the aquifer scale from fine-textured to coarse-textured aquifer facies.
- ▶ Kinetic behavior results, as well as long-term contaminant resupply after remedial activities.

Associated Practical Issues

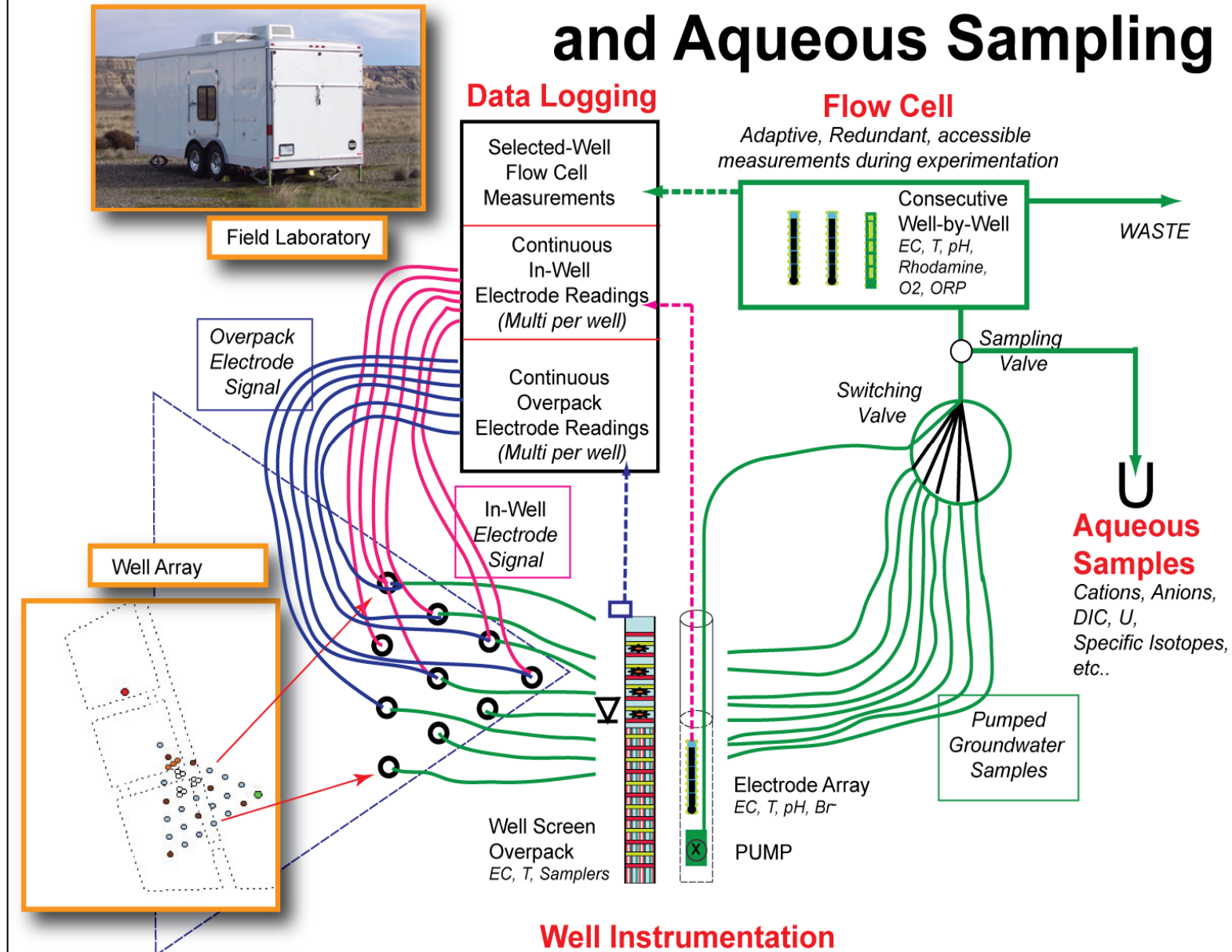
1. Accurate projection of dissipation times for groundwater plumes
2. Optimal delivery of remediation reactants
3. Effectiveness of remediation

Schematic of ERT / Monitoring Wells

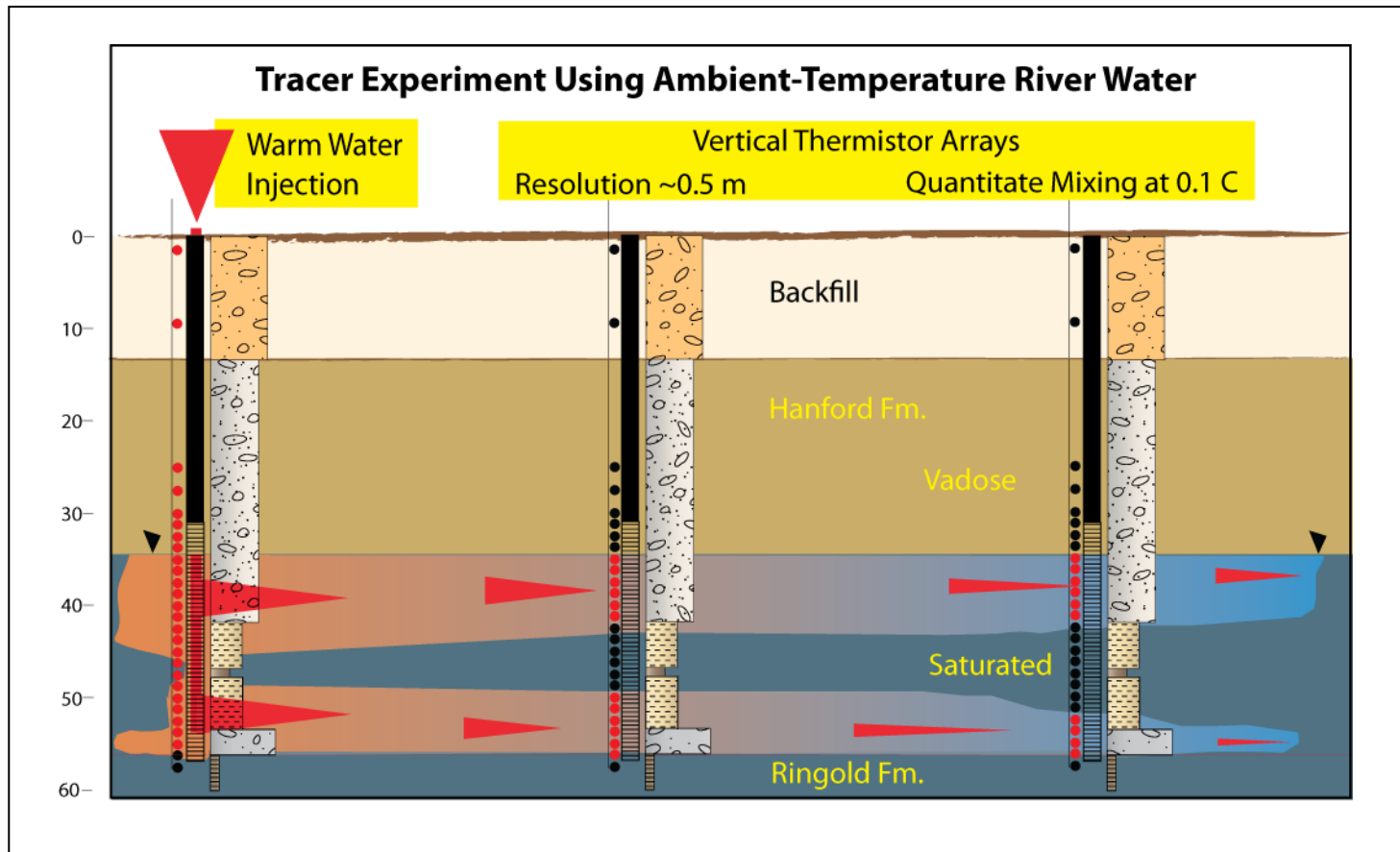


- ▶ Electrodes spaced at 60 cm (2 ft)
- ▶ Electrode length approx 10 cm (4 in)
- ▶ Electrode material 316 stainless steel
- ▶ Single wire connections to electrodes
- ▶ Wires run on outside of PVC well pipe
- ▶ Thermistors placed between electrodes
- ▶ Wire wrap PVC from 106-98 m elevation
- ▶ Tube capped at bottom
- ▶ Well head ~0.6 m (2 ft) above ground
- ▶ Central connector/DAQ box at top of wellhead
- ▶ Heat dissipation unit (HDU), time- domain reflectometry (TDR) probe and porous cup solution sampler at multiple depths on 5 wells around infiltration site

Field Electrode Measurements and Aqueous Sampling



Defining Heterogeneities in Hydrologic Properties and Flow



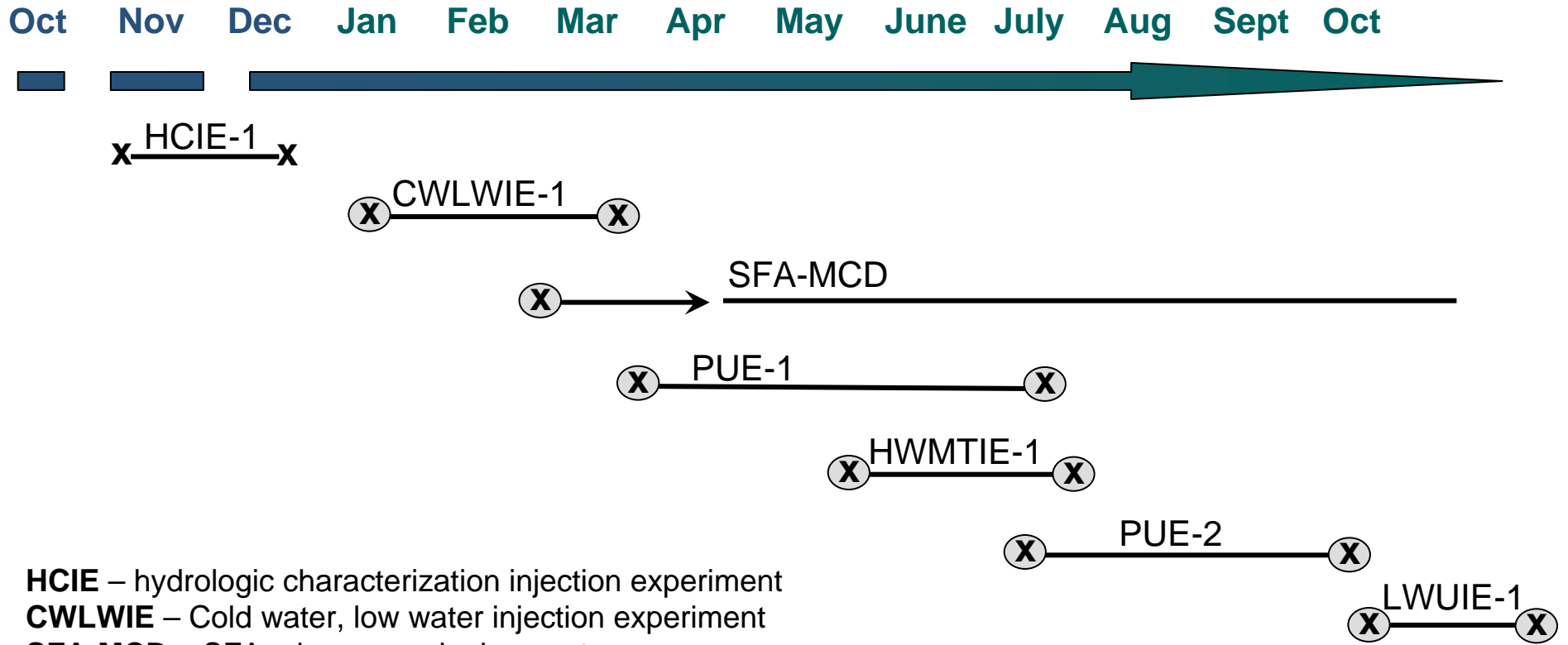
IFRC Experiments

- ▶ 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 HCO_3 and U(VI) concentrations, and U(VI) isotopic ratios
 - Passive experiments follow vadose zone pulses, or inland river water – groundwater gradients
- ▶ 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, HCO_3 , Na/Ca)
 - Passive experiments to explore rising and falling water table effects on U(VI) solubilization and release from lower vadose zone
- ▶ 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/ PO_4^{3-} , organic P with HCO_3
 - In collaboration with EM-20 and team

Hanford IFRC Preliminary Field Experimental Plan for FY09

FY09

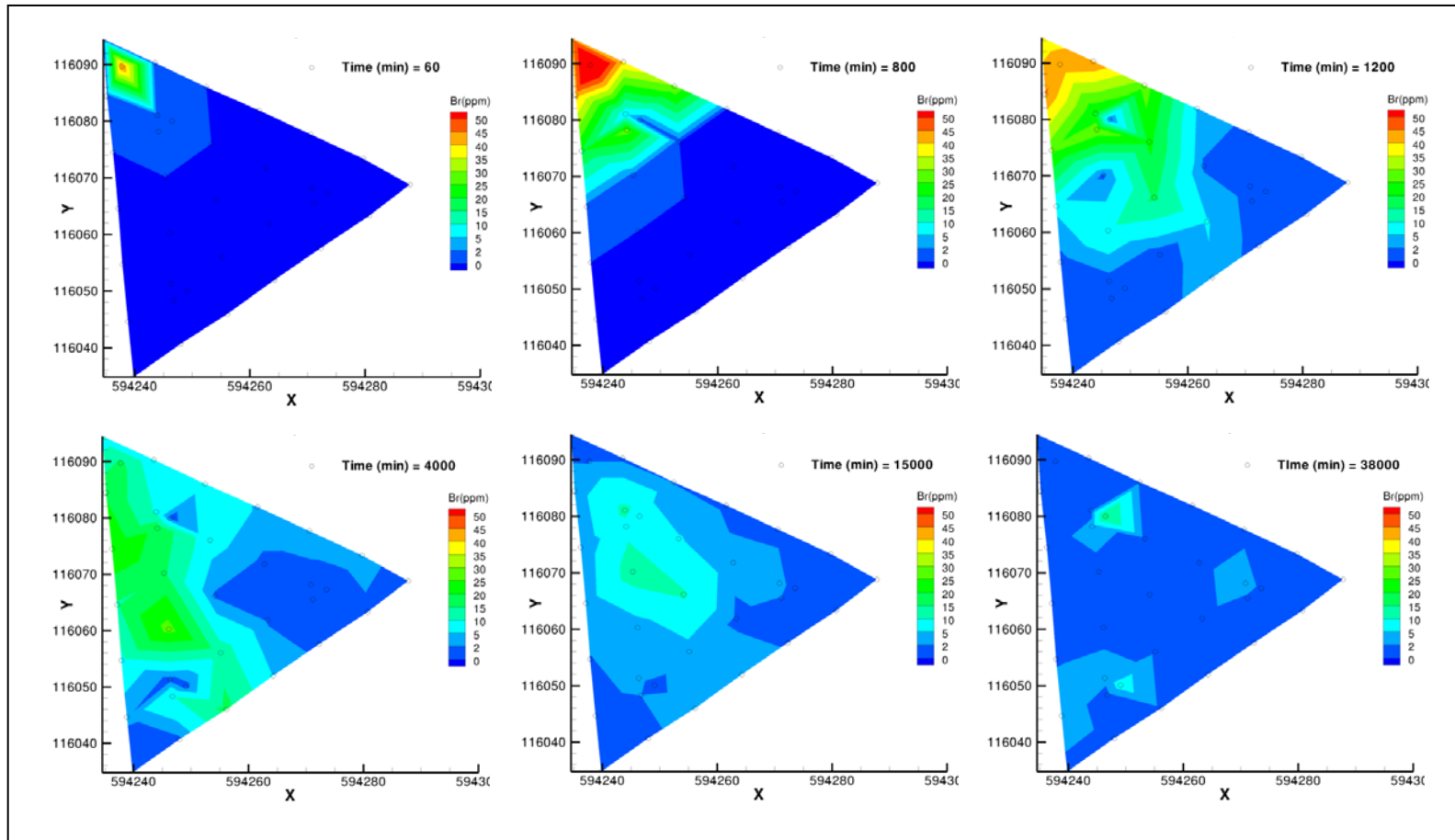
FY10



- HCIE** – hydrologic characterization injection experiment
- CWLWIE** – Cold water, low water injection experiment
- SFA-MCD** – SFA microcosm deployment
- PUE** – Passive uranium experiment
- HWMTIE** – High water multi-tracer injection experiment
- LWUIE** – Low water uranium injection experiment



Results of November 2008 Tracer Test for Hydrologic Characterization



Linkage of SFA/IFRC Research to Site Remediation, Closure, and Monitored Natural Attenuation

- ▶ Operational model for infusion of DOE science into site remediation and closure decisions
 - In-ground contaminant status and behavior
 - Understanding of processes and specific sites
 - Evaluation and testing of new models and measurements techniques
 - Knowledge to reduce uncertainty
 - Websites updated every 6 mo. to identify key findings and new publications

- ▶ 300 A site is representative of Hanford River Corridor locations
 - Applicability of conceptual and numeric models to other locations

- ▶ Scientific context for evaluation of remediation strategies and concepts
 - Critical characterization needs
 - MNA versus active approaches
 - Expectations for long-term remediation efficiency

Summary

- ▶ ERSD has invested significant research funding at PNNL to investigate "Hanford inspired" fundamental environmental science issues associated with contaminant fate and mobility, environmental microbiology, and advanced subsurface characterization and modeling.
- ▶ SFA and IFRC research projects are independent, but are closely linked and synergistic. Both are bringing international scientific expertise to Hanford.
- ▶ Scientific targets have been selected for research that are consistent with ERSD's Strategic Plan and that will yield useful knowledge to forecast contaminant migration and transformation at the Hanford site.
- ▶ ERSD seeks impact from the research funding in ways consistent with the Office of Science mission. Meaningful collaborations with EM are sought.