The Hanford 300 A Integrated Field Challenge

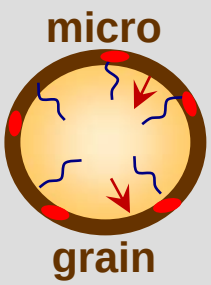
Supported by DOE Office of Biological and Environmental Research (OBER)
Environmental Remediation Sciences Division (ERSD)

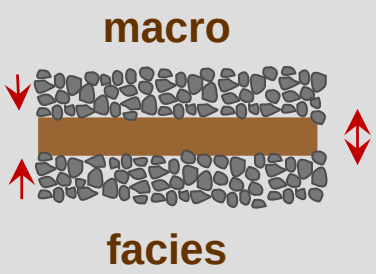
J. M. Zachara[1] and Colleagues
[1]Pacific Northwest National Laboratory, Richland, WA

August 29, 2007

Hanford IFC

Science Theme *~ Multiscale mass transfer processes influencing sorbed contaminant migration*





<u>Associated Practical Issues</u>

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

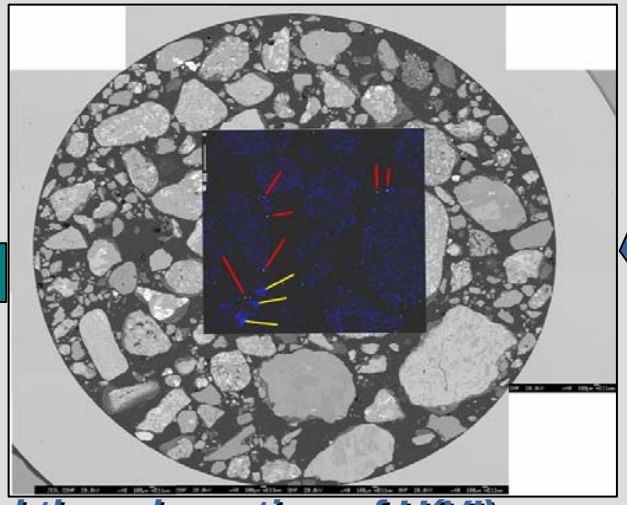
3. Practicality and effectiveness of remediation

Transport Behavior (Desorption/Sorption) in < 2 mm Process Pond Sediment is Kinetically Controlled

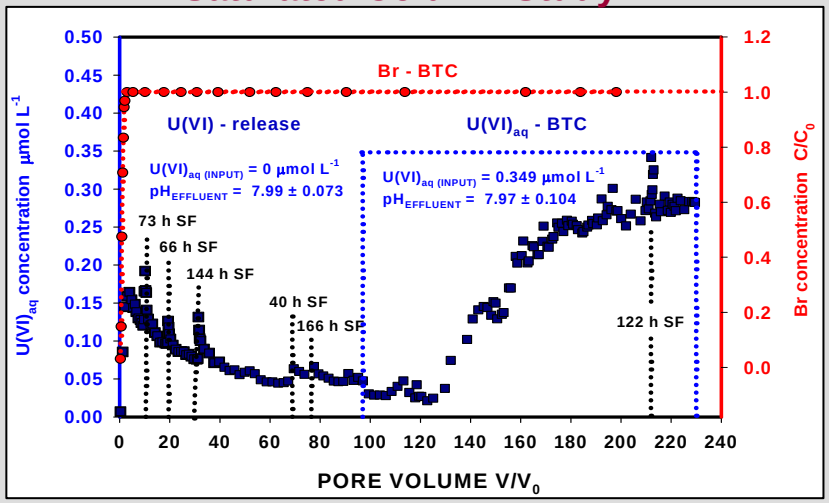
North Process Pond Pit 1 – 14 ft

Size Range (mm)	Mass Distribution (%)	U_{Total} (nmol/g)
Cobbles		
> 12.5	74.5	< 22
2.0 - 12.5	17.2	< 19
Sand		
1.0 - 2.0	2.64	26
0.5 - 1.0	2.34	< 18
0.25 - 0.5	0.78	< 21
0.149 - 0.25	0.33	37
0.106 - 0.149	0.19	< 23
0.053 - 0.149	0.20	< 23
Silt + Clay		
< 0.053	1.78	125

Electron Microprobe U Abundance Map on Backscattered Electron Image

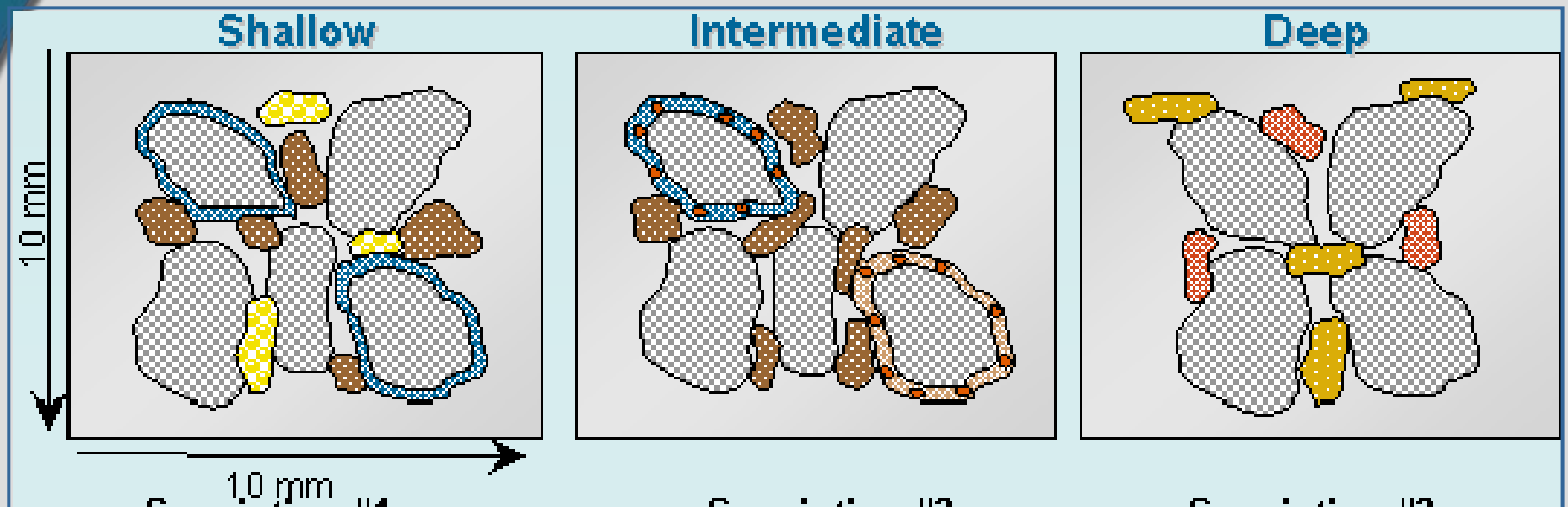


Saturated Column Study



The release of sorbed contaminant U(VI) and the adsorption of U(VI) from contaminated groundwater both show strong kinetic behavior

Conceptual Speciation Model Based on EXAFS, CLIFS, and Synchrotron XRD



Speciation #1

1.) Dispersed U(VI) coprecipitated in calcite
2.) Major copper grain coatings

Speciation #2

Discrete uranyl phosphate precipitates (metatorbermite, autunite) in grain coatings of:
1.) Malachite and
2.) Aluminosilicates

Speciation #3

1.) Adsorbed U(VI) in aggregates of clay sized phyllosilicates with variable Fe(III) oxide resulting from chlorite weathering

Heterogeneity and Mass Transfer Domains in 300 A Vadose Zone Sediments

Representative Facies from LFI Cores

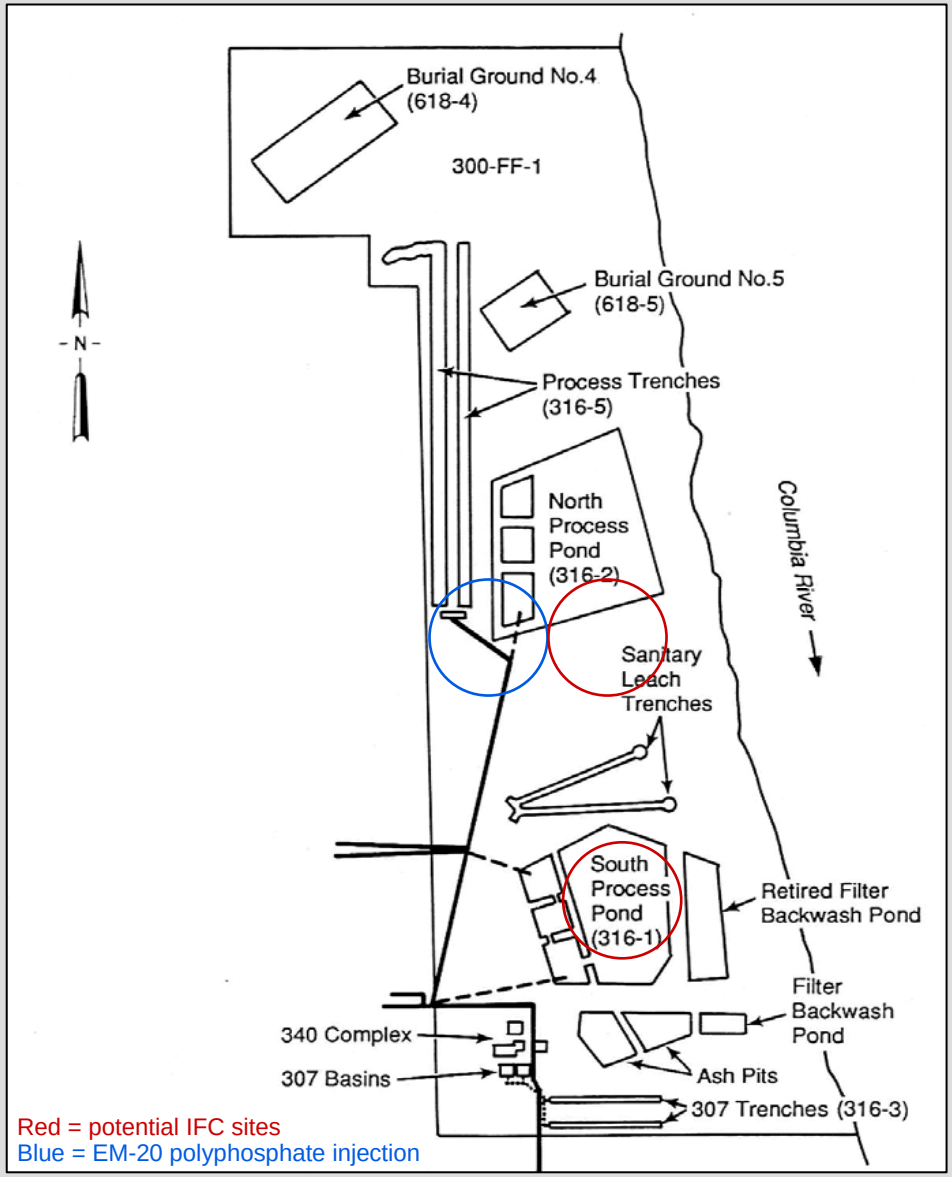
Primary Objectives

- 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 phosphate barrier performance and longevity
- Create enduring field experimental data sets for model and field-scale 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

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
  - ASCR
  - NRC

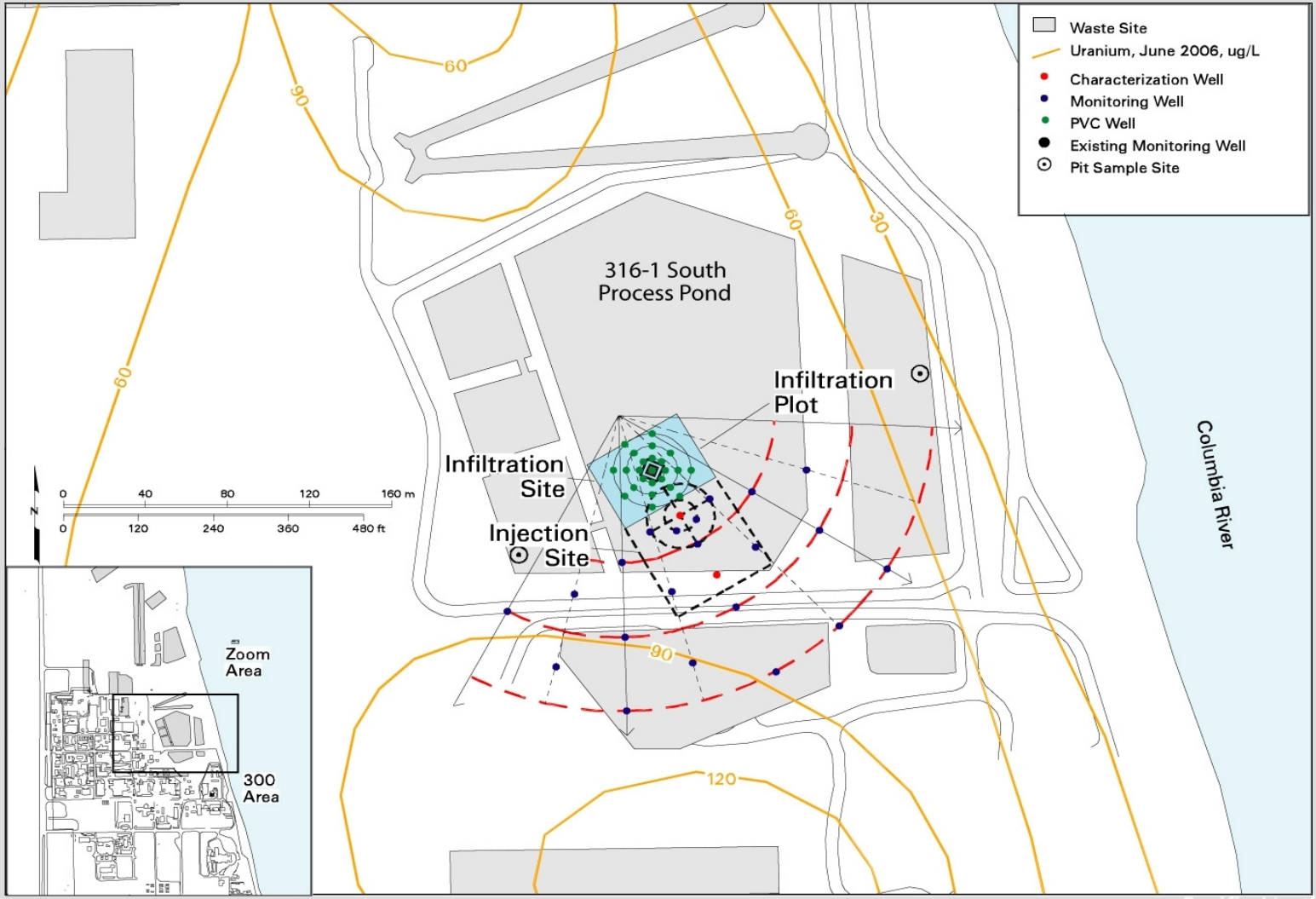
The 300-FF-1 Operable Unit



IFC site selection strongly dependent on polyphosphate plume trajectory

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

IFC Experimental Site in South Process Pond Footprint



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, HCO_3, 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 HCO_3 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/PO_4^{3-}, organic P with HCO_3
    - In collaboration with EM-22 and team

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)

Status

- Subcontracts with external collaborators are being finalized to begin 10/1/07.
- Criteria established for site selection.
- Geophysical surveys to aid in site placement and development underway in SPP.
- Three wells nearby the SPP (399-3-18, 399-3-19, and 399-3-20) are being 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. Additional modeling with newly determined adsorption parameters planned.
- Drilling specifications and experimental site design sent to Fluor for costing and bids, with drilling to commence in September.
- Project Management, Field Site Management, QA/QC, and Health and Safety Plans, and NEPA documentation completed.
- Characterization and Field Experimental Plans underway.
- Data base management task initiated. Prototype data sets for geology, geochemistry, hydrogeology, and geophysics identified for structuring data base.
- Coordination with EM projects established.

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