U.S. Department of Energy — Office of Science

Integrated Field-Scale Subsurface Research Challenge

Oak Ridge Field Research Center

Multiscale Investigations on the Rates and Mechanisms of Targeted Immobilization and Natural Attenuation of Metals, Radionuclides, and Co-Contaminants in the Subsurface: *The Oak Ridge IFRC*

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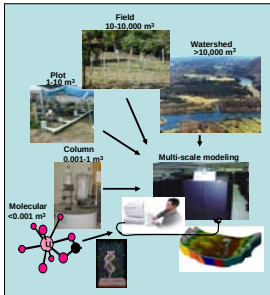
Project Goals



To advance the fundamental understanding and predictive capability of coupled processes that control *in situ transport and sequestration, remediation and natural attenuation* of metals, radionuclides, and co-contaminants (i.e. U, Tc, NO_3)

- *coupled processes*: hydrological, geochemical, and microbiological
- *multiple scales* ranging from molecular to watershed levels

Provide *multi-process, multi-scale predictive monitoring and modeling tools* that can be used at the ORIFC site and throughout the DOE complex

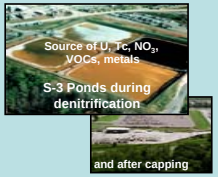
(1) inform and improve the technical basis for decision making

(2) assess which sites are amenable to natural attenuation and which would benefit from source zone remedial intervention

Site Description

The contaminated site of interest is on the Oak Ridge Reservation located in eastern Tennessee

The site consisted of unlined surface impoundments that received acidic nitrate- and U-bearing waste from 1951 to 1983 at a rate of 2.5 million gallons/year



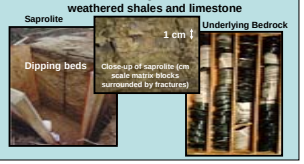
The subsurface media consist of fractured saprolite weathered from interbedded shale and limestone

Area has high recharge and a system conducive to rapid preferential flow of water and solutes.

The matrix porosity serves as a "secondary contaminant source" whose aerial extent is massive (tens of kilometers).

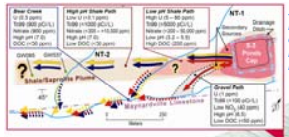
The problem scale is both local and at the watershed level.

ORIFC media consisting of interbedded fractured weathered shales and limestone



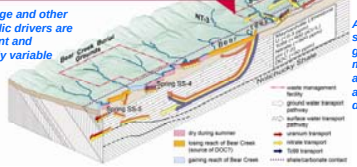
ORIFRC Research Site

Multi-scale *in situ* research along pathways and transition zones



Source areas and pathways contain numerous transition zones characterized by pronounced shifts in hydrology, geochemistry, and/or microbiology

Recharge and other hydraulic drivers are transient and spatially variable



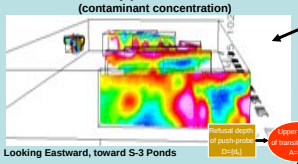
Aqueous and solid phase geochemistry, microbiology are spatially and temporally diverse

Quantifying Watershed Processes

Geophysical Quantification of Indigenous Plume-Scale Flow Architecture and Recharge Processes

Regional Data Acquisition

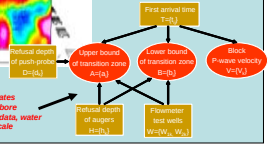
Electrical resistivity profiles down the watershed (contaminant concentration)



Looking Eastward, toward S-3 Ponds

Geophysics used to identify the 3-dimensional characteristics of the subsurface plume and media structure.

Geophysical signature quantified through cross-correlated with traditional macroscopic measurements such as Geoprobe conductivity profiles, refusal, and water quality data.



Novel Bayesian Estimation Framework incorporates surface seismic refraction arrival times and wellbore measurements (e.g. depth of refusal, flowmeter data, water quality) to estimate zonation at the watershed scale

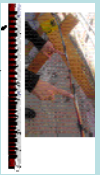
Quantifying Natural Attenuation Processes

(linking geophysical signatures with observed natural watershed processes)

Rates and mechanisms of coupled hydrological, geochemical, and microbiological processes that control the natural attenuation of U, Tc, and NO_3

Hydrologic impacts

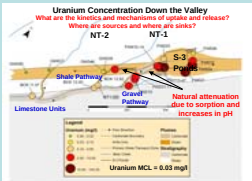
Time-lapse geophysical datasets to quantify recharge through the vadose zone

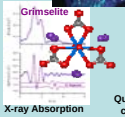


Recharge impacts capillary fringe and plume dynamics (dilution, biogeo-chemical changes)

Observed nitrate plume from geophysical surface resistivity

Geochemical impacts

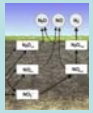




X-ray Absorption Spectroscopy defines U solid phase speciation

Quantify *in situ* contaminant behavior

Microbial impacts



Denitrification rates and mechanisms via stable isotope ratios in different solute and gaseous species within contaminant plumes of multiple scale (e.g., $^{15}N/^{14}N$ in nitrate, N_2O, and N_2).

Bacteria community composition and Shannon Index cross-correlated with geochemistry, hydrology across watershed

Multiscale, Multiprocess Site Wide Modeling

(enhanced predictive capability)

High-performance computing

Targeted Immobilization Strategies

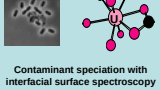
Field plot manipulations

Geophysical monitoring coupled with spatially intensive borehole macroscopic measurements

Sustained U(VI) reduction using Slow Release Substrate

Slow Release Substrate

Quantifying coupled hydrological, geochemical, and microbial processes



Contaminant speciation with interfacial surface spectroscopy

pH adjustment

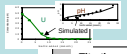
Slow titration of high indigenous solid and solution Al results in metastable polymers that scavenge U and Tc

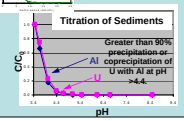
Large cores with coupled molecular analyses to define rates and mechanisms of U and Tc sequestration

SEM of Al-U coprecipitates on ORIFC sediment (formation of sparingly soluble species)

Experimental upscaling to field

Multiscale coupled processes modeling





Summary (also see task specific posters)

Project providing an improved conceptual and predictive understanding of manipulated and natural contaminant attenuation rates and mechanisms and the long-term effectiveness of remedial activities relevant to *in situ* remediation and stewardship at DOE sites.

Predictive monitoring and modeling tools to be used at sites throughout the DOE complex to inform and improve the technical basis for decision making, and to assess which sites are amenable to natural attenuation and which would benefit from source zone remedial intervention

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