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 IFC

U.S. Department of Energy — Office of Science
Integrated Field-Scale Subsurface Research Challenge
Oak Ridge Field Research Center

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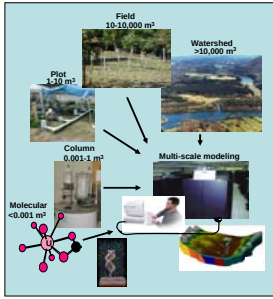
Abstract



The ORIFC is an integrated multi-disciplinary, multi-institutional research program that provides an improved scientific understanding and predictive capability of subsurface contaminant fate and transport at scales ranging from the molecular to the watershed

Seeks to quantify the influence of coupled processes on U, Tc, and nitrate migration along numerous distinct flow pathways and within transition zones throughout the watershed

Provides multi-process, multi-scale predictive monitoring and modeling tools to: (1) inform and improve the technical basis for remedial decision making, and (2) assess which sites are amenable to natural attenuation and which would benefit from source zone remedial intervention.

Site Description

The Oak Ridge IFC is located in eastern Tennessee, USA

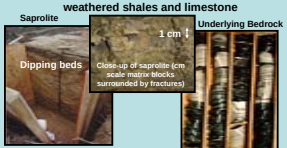
Unlined surface impoundments received acidic nitrate- and U-bearing waste from 1951 to 1983 at a rate of 2.5 million gallons/year. Attempts were made to neutralize the waste and the waste trenches were capped as a parking lot in 1988

Humid climate, ~1400 mm rainfall / y with 10% contributing to groundwater recharge and 40% contributing to surface water recharge.

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

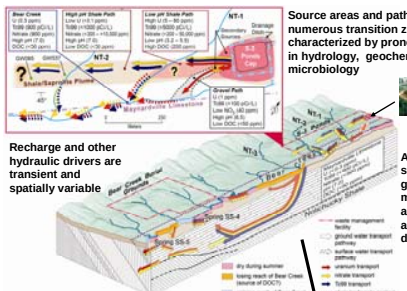
The problem scale is both local and at the watershed level

ORIFC media consisting of interbedded fractured weathered shales and limestone



Watershed Characterization

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

Advanced Geophysical Methods

Delineate Heterogeneity and Pathways
Link Geophysical Responses to Media Properties

Regional Data Acquisition

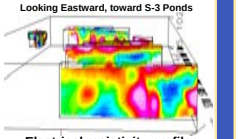
Surface geophysical data for 3-D characterization of media properties and contaminant plumes: Numerous coupled surface seismic refraction and electrical resistivity lines performed at various source areas, along plume transects, and the distal ends of plumes

Significant data has been processed for cross-correlation with traditional macroscopic measurements such as Geoprobe conductivity profiles, refusal, and water quality data.

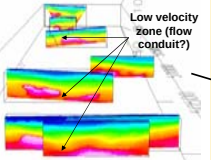
Development of a Bayesian approach to integrate multi-scale wellbore, crosshole, and surface datasets for quantitative watershed characterization

Site-wide model implementation

Looking Eastward, toward S-3 Ponds

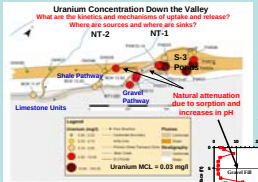


Electrical resistivity profiles (contaminant concentration)



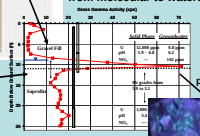
Seismic Refraction Profiles (media properties)

Quantifying Natural Attenuation Processes



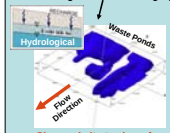
Rates and mechanisms of coupled hydrological, geochemical, and microbiological processes that control the natural attenuation of U, Tc, and NO_3

Highly diverse subsurface environments over scales ranging from molecular to watersheds



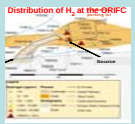
Recharge impacts Capillary fringe dynamics (contaminant dilution, biogeochemical changes)

U(VI) precipitate



X-ray Absorption Spectroscopy defines U solid phase speciation

Observed nitrate plume from geophysical surface resistivity



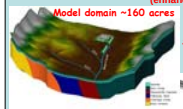
Stable isotope ratios in different solute and gaseous species within contaminant plumes of multiple scale (e.g., $^{15}N/^{14}N$ in nitrate, N_2O, and N_2). Physical vs. biogeochemical mechanisms

Permeable Environmental Leaching Capsules (PELCAPs) quantify *in situ* contaminant behavior (kinetics and mechanisms of attenuation / release)

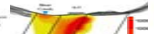
Bacteria community composition and Shannon Index cross-correlated with geochemistry, hydrology

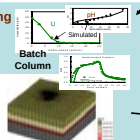
Multiscale, Multiprocess Modeling

(enhanced predictive capability)



Watershed





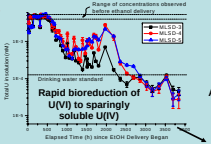
Targeted Immobilization Strategies

Field plot manipulations

(1)sustained bioreduction, (2) pH adjustment
(3) slow release electron donor (oleate), (4) organo-phosphate

Sustained bioreduction

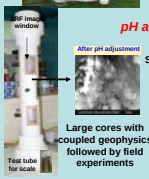
Impact of DO and NO_3 on U(IV) reoxidation



Rapid bioreduction of U(VI) to sparingly soluble U(IV)

Application of microarrays

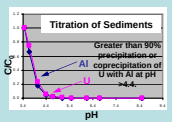
pH adjustment

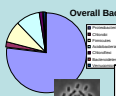


SEM of Al-U coprecipitates on ORIFC sediment (formation of sparingly soluble species)

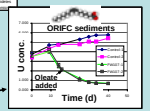
Contaminant speciation with interfacial surface spectroscopy

Large cores with coupled geophysics followed by field experiments





Oleate amendments



Bioreduction of U(VI) to U(IV)

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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