Oak Ridge Integrated Field-Scale Research Challenge
ERKP686

Multi-scale Investigations on the Rates and Mechanisms of Targeted Immobilization and Natural Attenuation of Metal, Radionuclide and Co-Contaminants in the Subsurface (project overview)

Environmental Remediation Science Program

Oak Ridge National Laboratory
Argonne National Laboratory
Florida State University
Georgia Tech
Lawrence Berkeley National Laboratory
Stanford University
University of Oklahoma
University of Tennessee

ERSP Annual PI Meeting
Lansdowne, Virginia
April 16-19, 2007

Oak Ridge National Laboratory

Team Members

- **Scientific Coordination:** Phil Jardine (PI), Dave Watson (Field Research Manager, ORNL) in consultation with project scientific disciplines.
- **Geophysics**: Susan Hubbard*, Ken Williams, J. Chen (LBNL), Greg Baker and David Gaines (UT), Les Beard (Battelle)
- **Geochemistry:** Scott Brooks*, Brian Spalding, Phil Jardine (ORNL), Ken Kemner and Shelly Kelly (ANL)
- **Microbiology:** Joel Kostka* and team (FSU), Tony Palumbo and Chris Schadt (ORNL), Joe Zhou and team (OU).
- **Biogeochemistry:** Craig Criddle* and Weimin Wu (Stanford), Baohua Gu (ORNL)
- **Hydrology:** Dave Watson* (ORNL), Jack Parker (ORNL/UT), Peter Kitanidis (Stanford)
- **Numerical Modeling:** Jack Parker* (ORNL/UT), Craig Brandt and Fan Zhang (ORNL), Peter Kitanidis (Stanford), Jian Luo (Georgia Tech)
- **Data Management:** Craig Brandt* (ORNL)

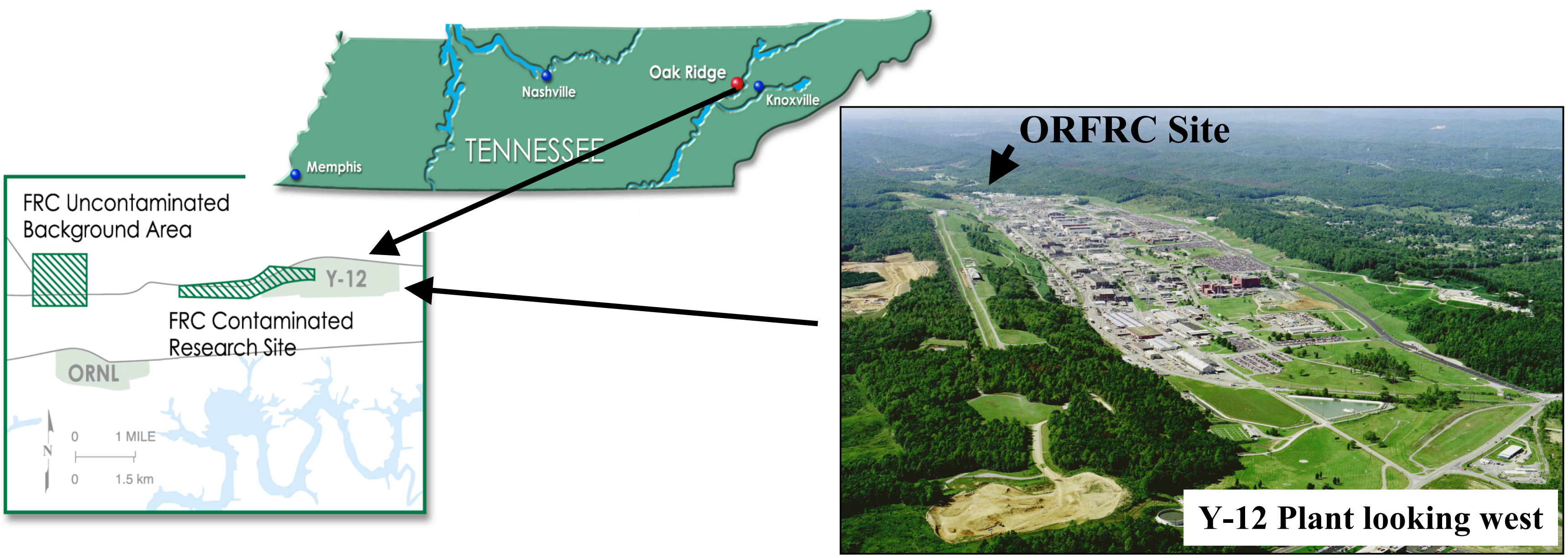
*** Disciplinary lead**

Agenda

- **Project Overview**: (speaker: Jardine) 15 min
- ***(Task B)*** **Natural Attenuation**: Rates and Mechanisms along pathways and within source zones (speaker: Watson) 20 min
- ***(Task C)*** **Targeted Manipulations**: Enhanced contaminant stability of source zones (speaker: Criddle) 20 min
- ***(Tasks A-C)*** **Geophysics**: Characterization and monitoring (speaker: Hubbard) 20 min
- ***(Tasks B & C)*** **Microbiology**: Characterization and monitoring as a function of scale (speaker: Kostka) 20 min
- ***(Task D)*** **Numerical Modeling**: Multi-scale flow and transport modeling, upscaling, and advanced pattern recognition (speaker: Parker) 15 min
- **Research Outcomes, Site Contributions, Data Management, and Opportunities**: (speaker: Jardine) 5 min

Oak Ridge Integrated Field-Scale Research Challenge

Project Overview



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Purpose / Rationale

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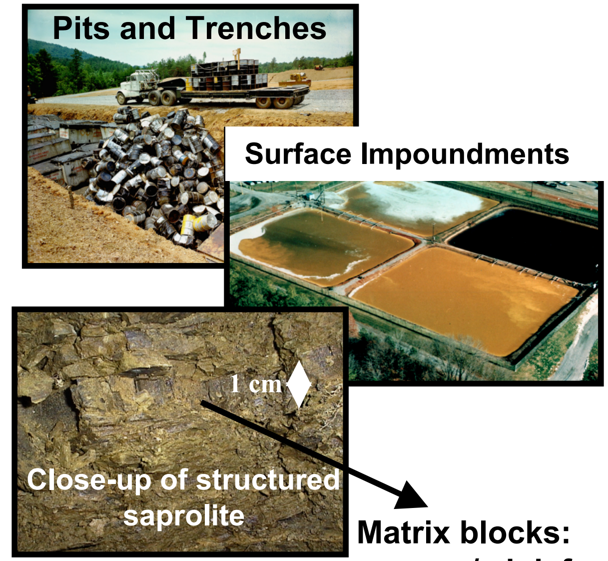
Historical disposal of wastes from the operation of three industrial plant sites on the Oak Ridge Reservation (ORR) has created extensive areas of subsurface inorganic, organic, and radioactive contamination (thousands of unlined trenches, pits, ponds).

These wastes have resulted in approximately 1,500 acres of contaminated groundwater on the ORR.

Much of the original contamination is now present as <u>secondary sources</u> within the soil-rock matrix outside of the original disposal sites.

The secondary source areas are extensive and encompass regions on the watershed scale (tens of km).



Matrix blocks: source / sink for contaminants

A significant limitation in assessing remediation needs of the secondary contaminant sources is the lack of information on the rates and mechanisms of coupled hydrological, geochemical, and microbial processes that control contaminant migration.

Contaminant fluxes emanating from the secondary sources are often so high as to prevent complete attenuation of the groundwater plumes.

Interventions such as source actions may be a prerequisite for effective and rapid natural attenuation (source actions such as: reduction of the soluble contaminant concentration at the source or controlling the flux from the source to groundwater by decreasing recharge)

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Goals

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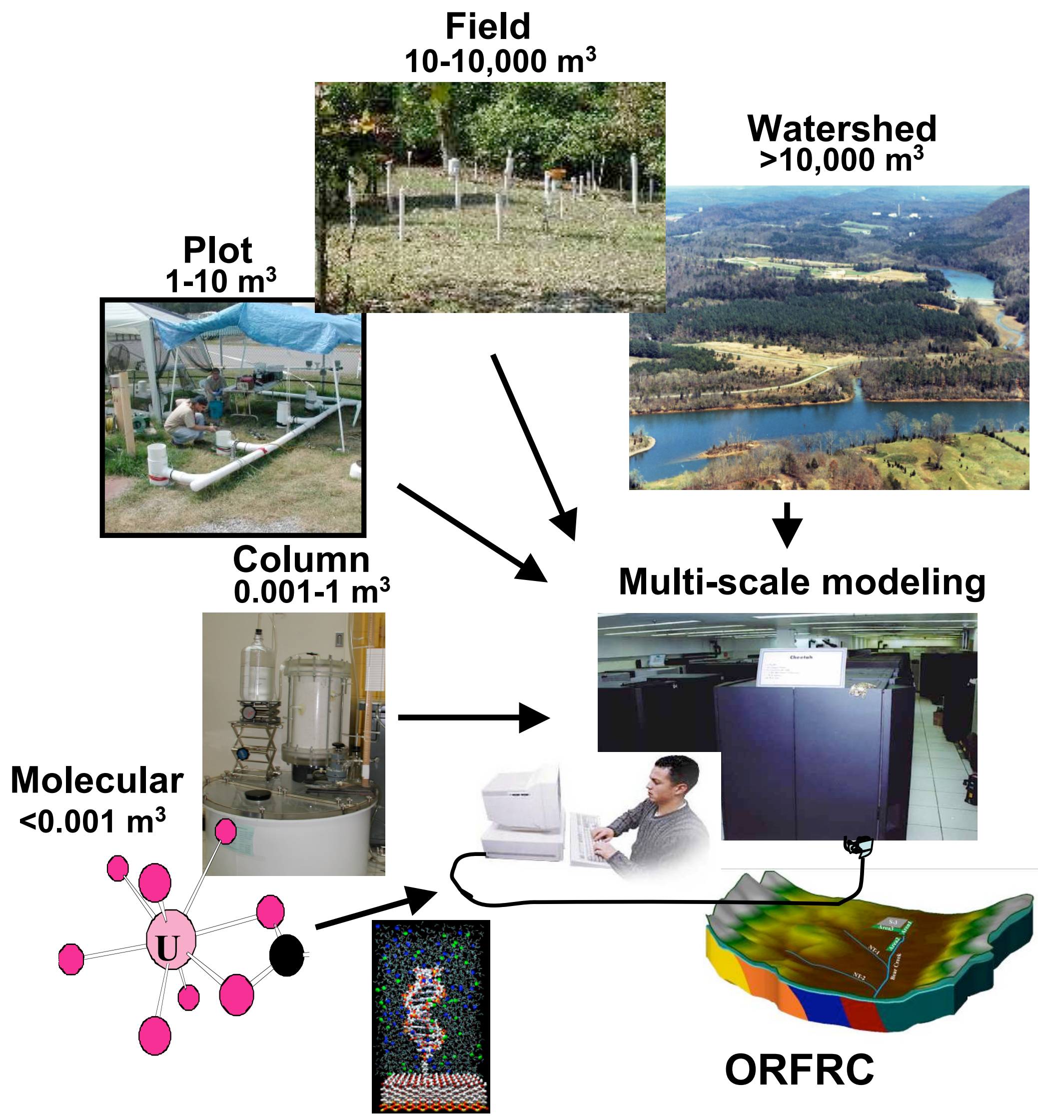


To advance the understanding and predictive capability of coupled hydrological, geochemical, and microbiological processes that control *in situ* transport, remediation and natural attenuation of metals, radionuclides, and co-contaminants (i.e. U, Tc, NO_3) across multiple scales ranging from molecular to watershed levels.

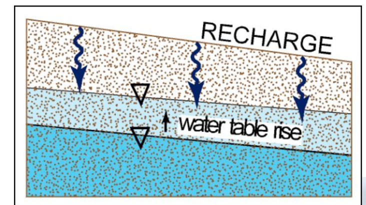
Provide multi-process, multi-scale predictive monitoring and modeling tools that can be used at sites throughout the DOE complex to:

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

(2) assess which sites are amenable to <u>natural attenuation</u> and which would benefit from source zone <u>remedial intervention</u>.

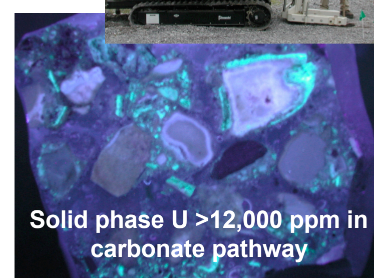
Objectives

(1) quantify recharge and other hydraulic drivers for groundwater flow and dilution of contaminants along flow pathways and determine how they change temporally and spatially during episodic events, seasonally, and long term.



(2) determine the rates and mechanisms of coupled hydrological, geochemical, and microbiological processes that control the natural attenuation of contaminants in highly diverse subsurface environments over scales ranging from molecular to watersheds.

(3) explore novel strategies for enhancing the subsurface stability of immobilized metals and radionuclides.


Solid phase U >12,000 ppm in carbonate pathway

(4) understand the long-term impacts of geochemical and hydrologic heterogeneity on the remobilization of immobilized radionuclides.

(5) improve our ability to predict the long-term effectiveness of remedial activities and natural attenuation processes that control subsurface contaminant behavior across a variety of scales.

Relevance

This project is intimately connected with the ORR Groundwater Strategy Document (DOE 2004) which emphasizes the need for timely and focused research investigations on natural hydrogeologic systems at the ORR to help evaluate the technical feasibility and cost-effectiveness of various remediation strategies including natural attenuation.

The results of our proposed research will have maximum impact on ORR groundwater remediation decisions since groundwater decisions are slated for 2012 – 2015, which is the same time period that our project ends.

> We will provide an enhanced scientific understanding of subsurface processes through improved multi-scale characterization and numerical modeling tools that are needed to predict contaminant fate and transport under a variety of remediation scenarios.

David Watson is an invited member of the ORR Groundwater Core Team that seeks to “facilitate the identification, funding, and implementation of high-priority science and technology investigations” as related to ORR site groundwater issues.

> This team works with state regulators, remediation contractors, and the DOE ORR Closure Project Core Teams to focus efforts on *the goal of site closure*.

Characterization data and research findings from the ORFRC will continue to be input into the Oak Ridge Environmental Information System (OREIS), which is the long-term repository for data generated by the ORR EM and compliance programs for use in decision making.

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Site Description

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The Oak Ridge FRC is located in eastern Tennessee and contains contaminated and uncontaminated field facilities as well as on-site and off-site laboratory facilities.

At the contaminated site, 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 neutralized the waste and the site was capped in 1988.

The region receives ~1400 mm rainfall / y with 10% contributing to groundwater recharge and 40% contributing to surface water recharge. Water table 0 - 4 m from surface, with fringe ~ 1m.

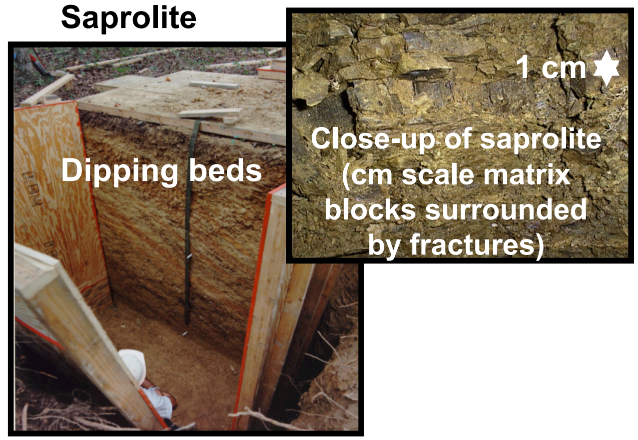
The subsurface media consist of fractured saprolite weathered from interbedded shale and limestone and is conducive to rapid preferential flow of water and solutes.

Fractures surround a low permeability, high porosity matrix which acts as a kinetically controlled source and sink for solutes.

It is the matrix porosity that serves as the "secondary contaminant source" whose aerial extent is massive (tens of kilometers).

ORNL media consisting of interbedded fractured weathered shales and limestone

Saprolite



Underlying Bedrock

Environmental Remediation Sciences

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Site Description

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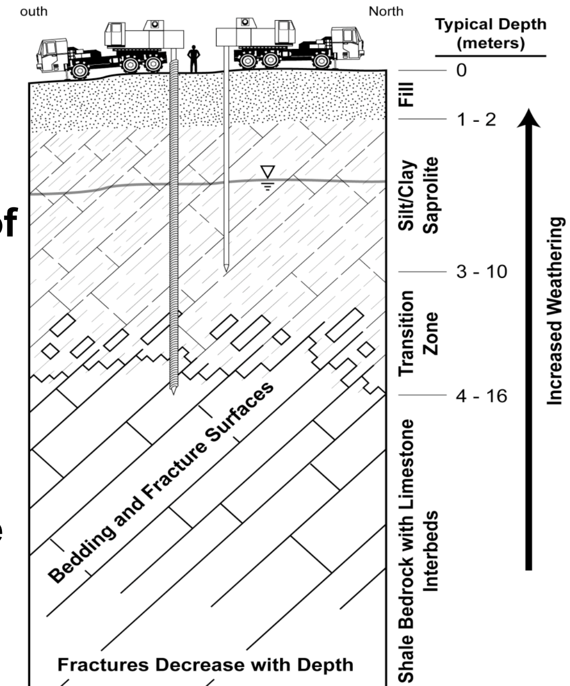
Preferred direction of contaminant plumes is along geologic strike. However, huge density effects exist and significant movement along bedding plane dip.

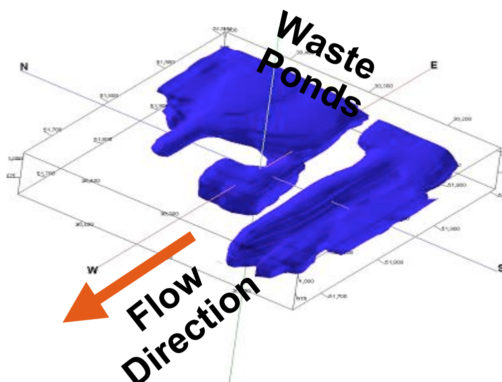
Both aqueous and solid phase geochemistry and microbiology are spatially and temporally diverse due to changing conditions of ionic strength, redox, pH, buffer capacity, nutrient and electron donor/carbon availability, and mineralogy. The system is highly reactive and in a state of non-equilibrium.

Subsurface processes are coupled and have been well characterized for many years, at this site and others, using interfacial surface spectroscopy techniques, geophysical and geochemical interrogation methods, and multi-scale manipulative experiments focused on interactive hydrological, geochemical, and microbial processes.

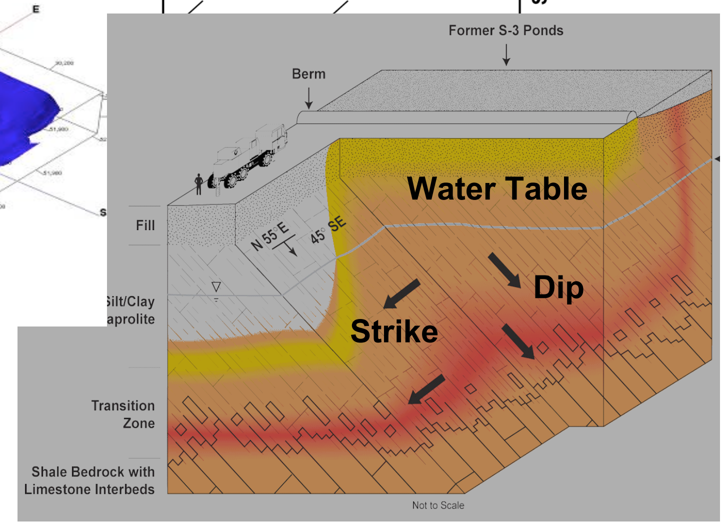


Conceptual models have been developed for secondary sources and a variety of shallow and deep plumes with variable hydrology, geochemistry, and microbiology.

The site is complex, but not complicated which lends itself as a tractable challenge for investigation coupled subsurface processes and their influence on NA and remediation.

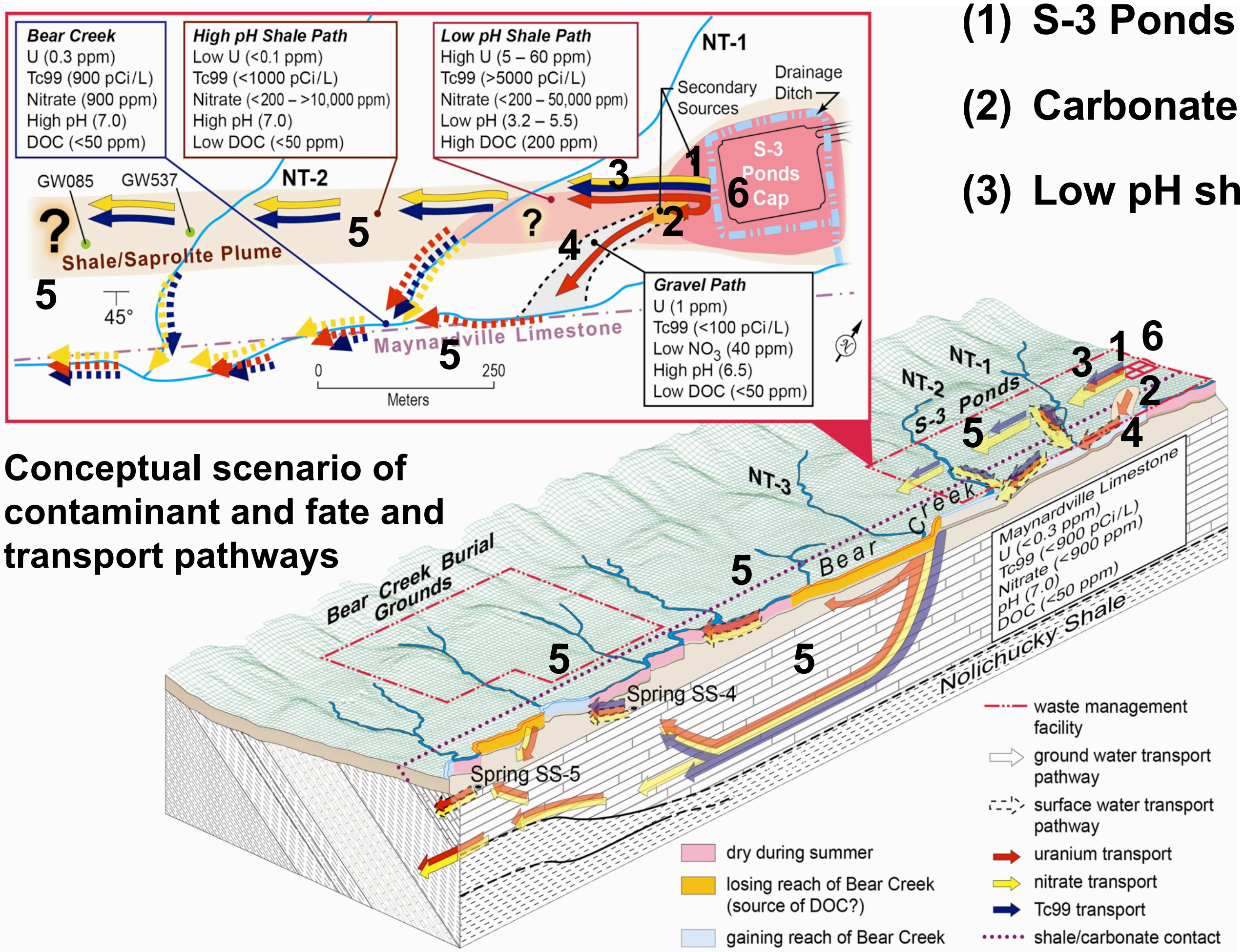


Observed conductivity plume model from geophysical surface resistivity (nitrate >3000mg/l)



Site Description

Focus on several <u>secondary source zones and flow pathways</u> in the ORFRC that represent a <u>range of scales</u>. These source zones and pathways contain numerous transition zones characterized by pronounced shifts in hydrology, geochemistry, and/or microbiology.



Conceptual scenario of contaminant and fate and transport pathways

(1) S-3 Ponds secondary source

(2) Carbonate gravel secondary source

(3) Low pH shale/saprolite pathway

(4) Neutral pH carbonate gravel pathway

(5) Neutral pH Maynardville Limestone and shale / saprolite pathways

(6) Recharge pathway

Hypotheses Driven Approach

H1. *Geochemical Controls on Contaminant Attenuation*:

Microbial denitrification is the only geochemical mechanism for permanently decreasing NO_3^- flux (rate governed by pH and electron donor).

The master variable for U, and possibly Tc, attenuation is pH (i.e. aluminum hydrolysis and the presence of carbonates).

H2. *Recharge Controls on Hydraulics, Geochemistry, and Microbiology*:

Recharge immediately adjacent to the contaminant source is the main hydraulic driver for groundwater flow and dilution of contaminants

It is this recharge that dictates temporal and spatial variability of geochemical and microbiological processes in the saturated zone.

H3. *Secondary Source*:

The secondary source strength is of a magnitude that attenuation of contaminant plumes will not occur rapidly unless an action is taken (reducing concentration and/or by controlling recharge).

H4. *Transition Zone*:

Important from a remedial perspective since (1) characterized by steep gradients in subsurface properties, (2) transient in space and time, (3) most active zones with respect to geochemical and microbially mediated metal and radionuclide transformations.

H5. *Enhanced Stability*:

Enhanced subsurface stability of U and Tc can be achieved through remedial strategies that (1) maintain a favorable geochemistry and microbial ecology, (2) minimize biogeochemical heterogeneity, and (3) counteract or inhibit mechanisms of reoxidation and remobilization.

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Multi-Task Approach

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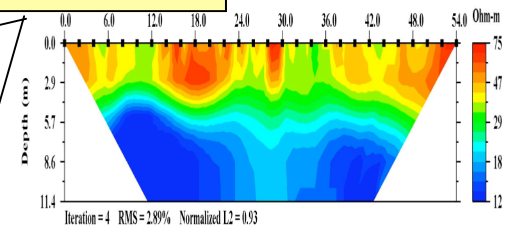
Link Geophysical Responses to Media

Task A – Geophysical Definition of Subsurface Heterogeneity within Pathways

Define Heterogeneity and Pathways



Task B – Quantifying Rates and Mechanisms of Natural Attenuation

Shale and Carbonate **Pathways**

Rates and Mechanisms of **Nitrate** Natural Attenuation

Rates and Mechanisms of **U and Tc** Natural Attenuation

Recharge Studies

Multi scale hydrobio-geochemical, geophysical studies

Recharge **Reduction** Test

Isotopes, coupled processes, geophysical monitoring of natural pathways

Task C – Enhanced Contaminant Stability Strategies for Source Control

Microbial Reduction Assessment

Oleate Slow Release Assessment

Metal Sequestration by Organo-**phosphate** Amendment

Long-term Immobilization of U and Tc by Subsurface **pH Adjustment**

Controlled **lab and field** investigations of coupled processes

Geophysical and coupled processes monitoring of manipulated zones

Site-wide modeling

Advanced Pattern Recognition

Local-scale Modeling

Task D – Multi-process and Multi-scale Numerical Modeling and Data Analysis

Up-scaling and Model Accuracy

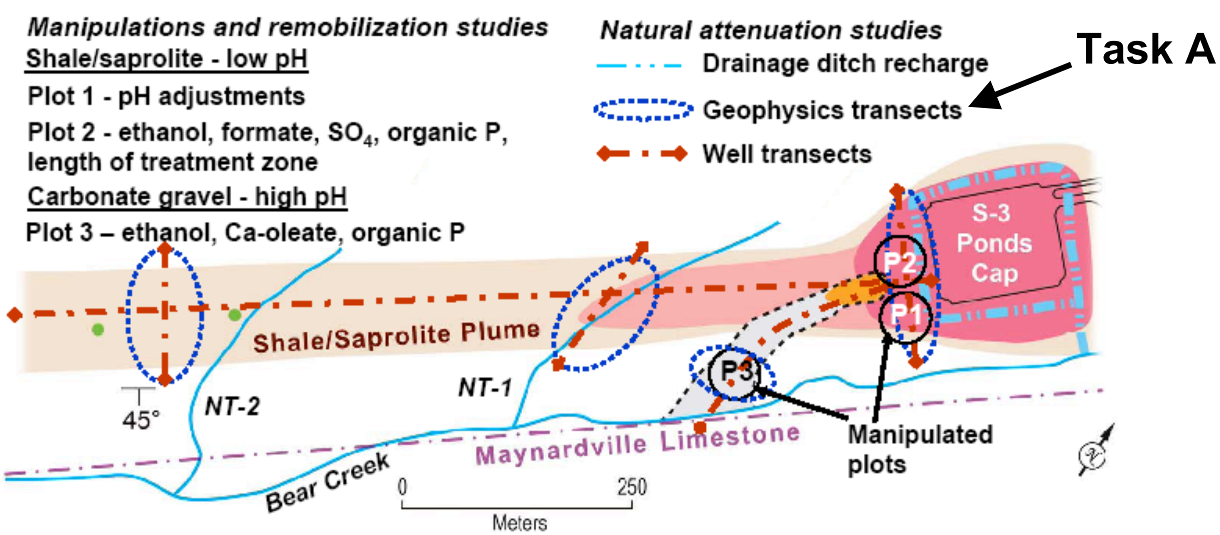
Predictive monitoring and modeling tools, scientifically informed choices on ground water remedial actions and stewardship, peer-reviewed publications

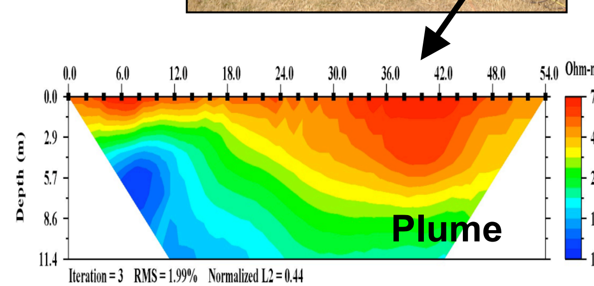
Approach

<u>Task A: Define flowpaths and heterogeneities that control the fate and transport of contaminant plumes</u>

- **Associate geophysical response (e.g., electrical, seismic, radar, SP) to media and plume properties.**
- **Monitor geophysical and hydro-bio-geo-chemical changes across plume cross-section over time to quantify impact of <u>short and long-term recharge</u> events and <u>remedial manipulations</u> on plume attenuation.**

Location of research tasks at ORFRC





Approach

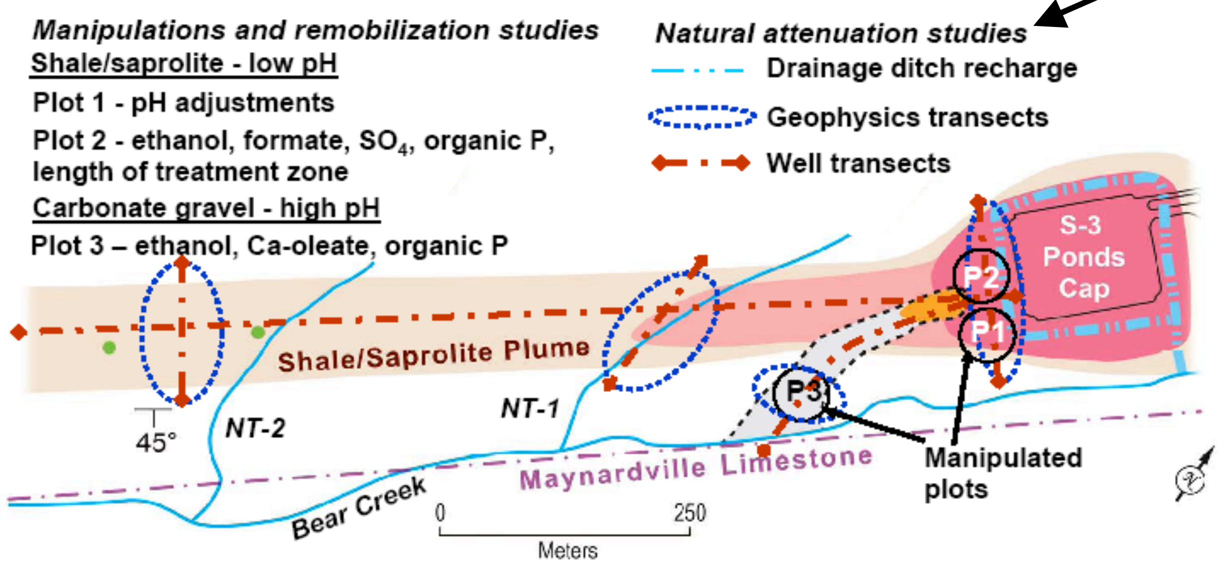
<u>Task B: Define and quantify natural attenuation rates & mechanisms across the Bear Creek watershed</u>

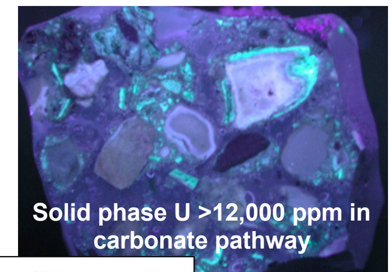
- **Impacts of coupled pH, redox conditions, microbial activity, reactivity, etc. on U, Tc, and nitrate <u>natural attenuation</u> (isotopes, spatial and temporal variability along pathways and transition zones).**

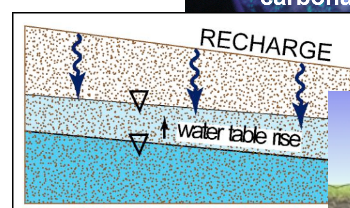
- **Impacts of recharge on geochemistry, contaminant dilution, O_2, carbon source, microbial activity, etc. on spatial and temporal plume dynamics.**

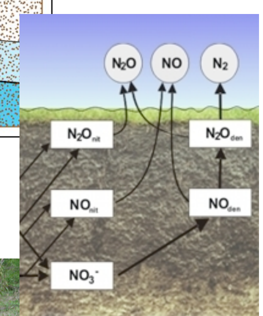
Location of research tasks at ORFRC

Task B







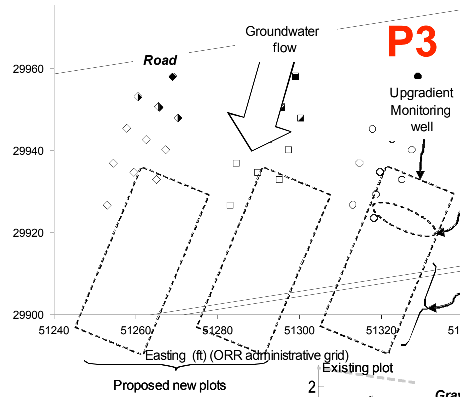


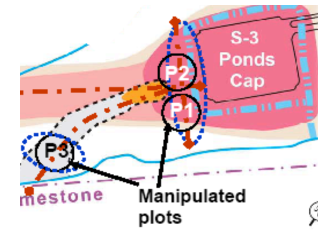
Approach

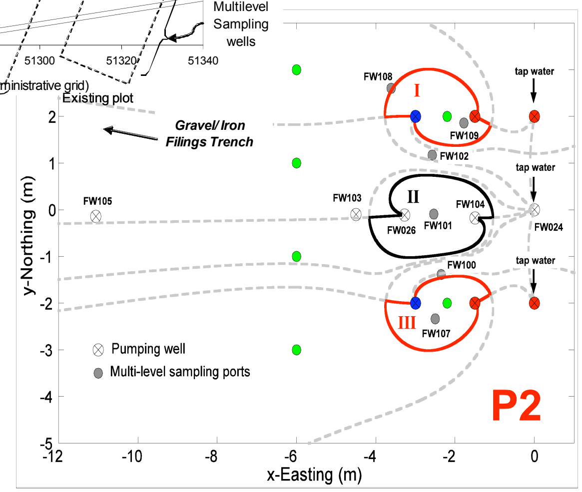
<u>Task C: Quantitative *in situ* immobilization strategies within secondary sources of the saprolite and carbonate units (U, Tc, nitrate)</u>

Targeted <u>manipulation</u> experiments: (1) sustained bioreduction, (2) pH adjustment, (3) organo-phosphate amendments, (4) slow release oleate amendments.

Monitor multi-scale hydrobiogeochemical and geophysical changes and propensity for contaminant remobilization.



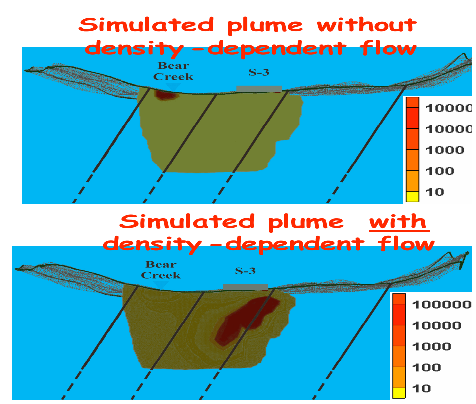


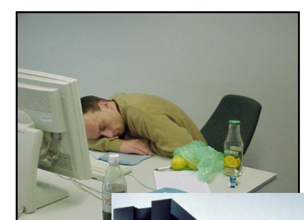


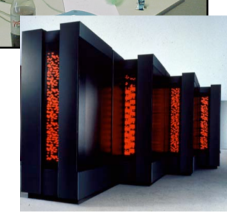
<u>Task D: Multiprocess and multiscale numerical modeling and data analysis</u>

Local plot scale modeling (Task C results) and Advanced Pattern Recognition techniques (Task B results).

Site wide modeling (HydroBioGeoChem) / upscaling and model accuracy (Tasks A-C results).







Presentations to follow: ***Detailed Research Tasks and Descriptions***

- ***(Task B)* Natural Attenuation**: Rates and Mechanisms along pathways and within source zones (speaker: Watson) 20 min
- ***(Task C)* Targeted Manipulations**: Enhanced contaminant stability of source zones (speaker: Criddle) 20 min
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