Oak Ridge Integrated Field Research Challenge *(ORIFRC)*

Multiscale Investigations on the Rates and Mechanisms of Targeted Immobilization and Natural Attenuation of Metal, Radionuclide and Co-Contaminants in the Subsurface

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 20-23, 2009

OAK RIDGE National Laboratory

Team Members

Multi-disciplinary / Multi-institutional

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

*** Disciplinary lead**

Oak Ridge Integrated Field Research Challenge site

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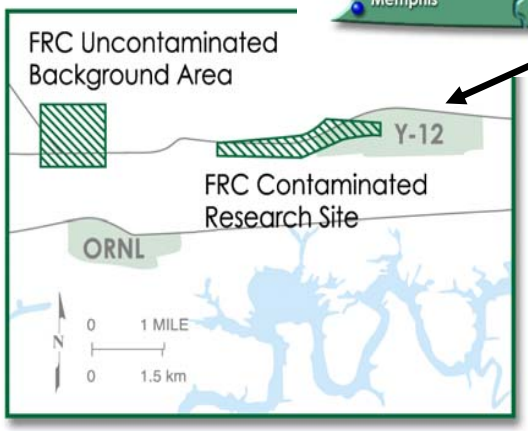
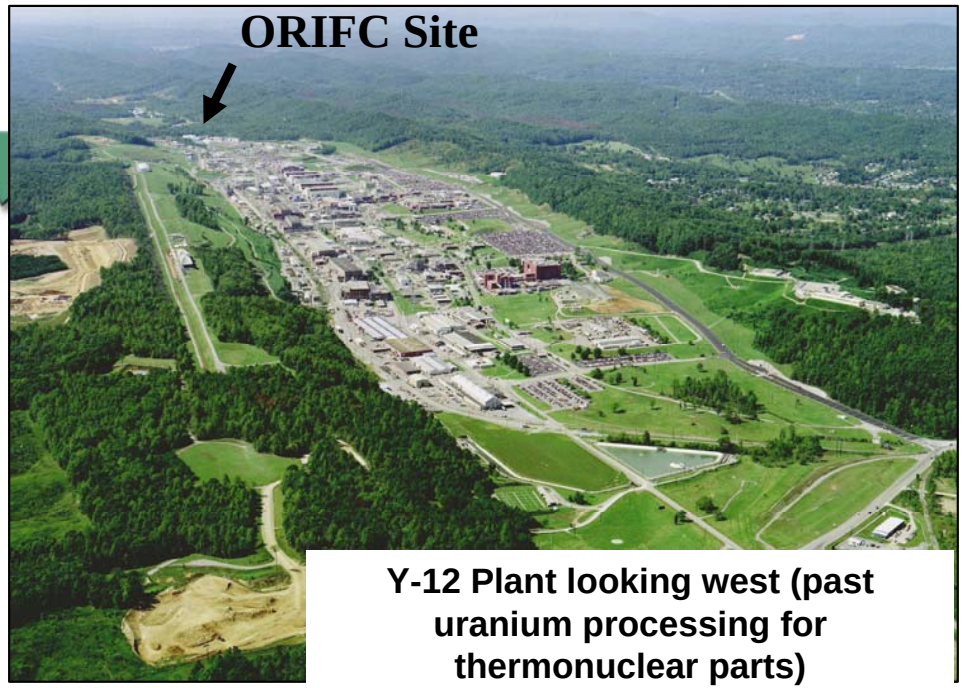




Y-12 Plant looking west (past uranium processing for thermonuclear parts)

Site Description

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

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

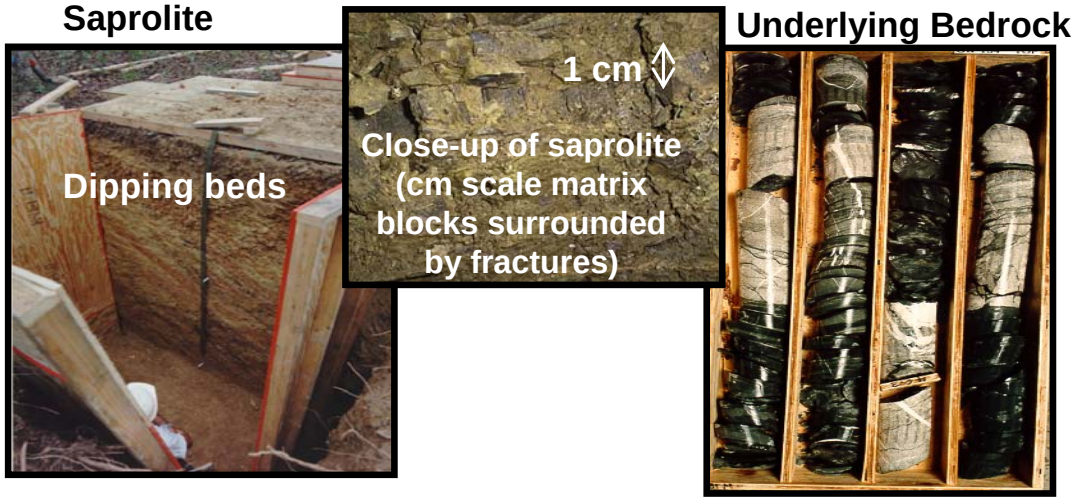
The subsurface media consist of fractured saprolite weathered from interbedded shale and limestone

High recharge and a system conducive to rapid preferential flow of water and solutes.

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

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

ORNL media consisting of interbedded fractured weathered shales and limestone



Remediation Dilemma for Secondary Sources

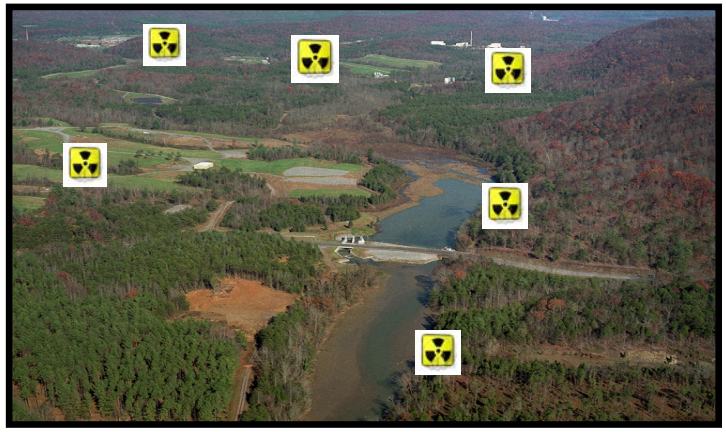
The problem is daunting since the spatial distribution of contaminants is massive

- **No feasible removal or immobilization technologies for large volumes of contaminated subsurface saprolites, bedrock, groundwater.**
- **Decisions are often made to leave contaminants in-place.**
- **Fundamental investigation, monitoring, and further evaluation of options are critical since contaminants left in-place continue to interact with the geosphere**

- ***Natural physical attenuation via dilution by recharge or diffusion into high porosity, low permeability matrix.***
- ***Natural biogeochemical attenuation via sorption, redox transformation, degradation, dissociation, and precipitation reactions.***
- ***Targeted manipulations to convert metals and radionuclides to more stable forms.***

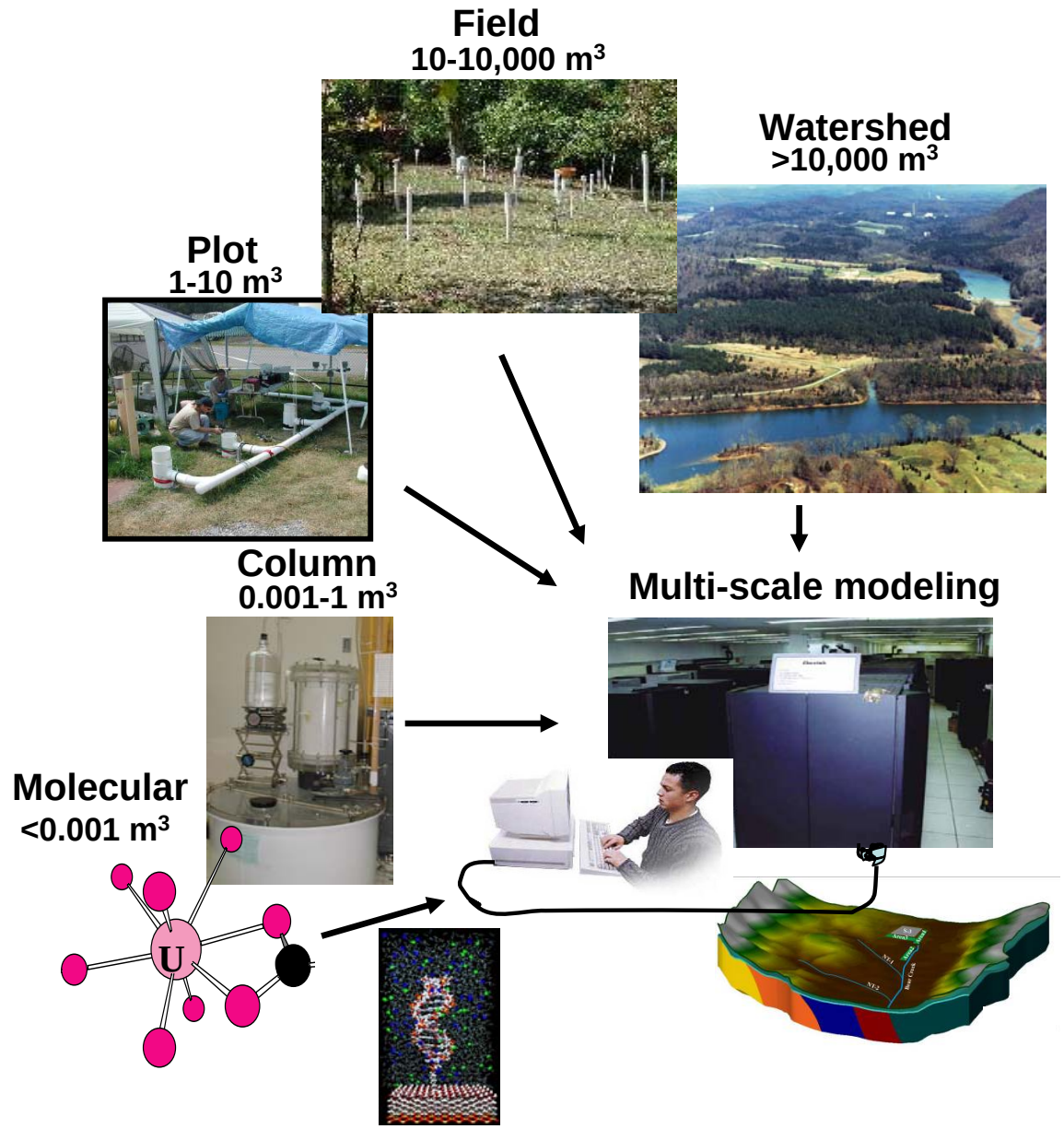
The focus of our research and presentations

How to deal with DOEs legacy waste



Watershed scale problem

Our 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

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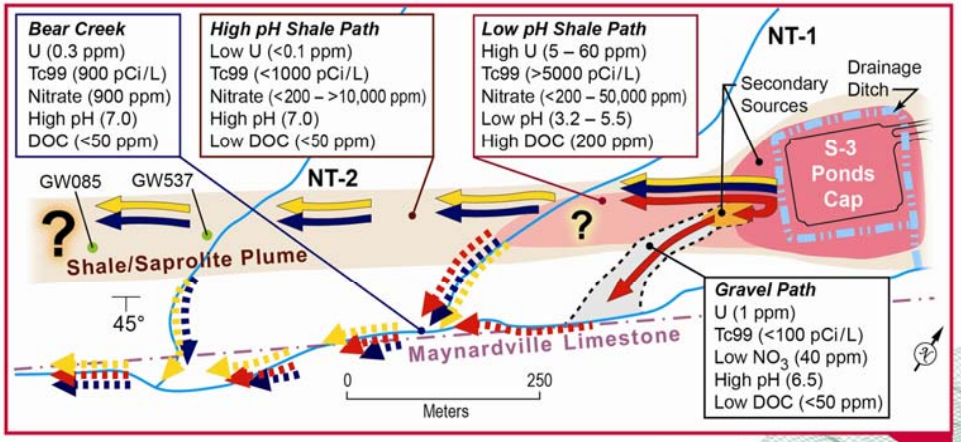
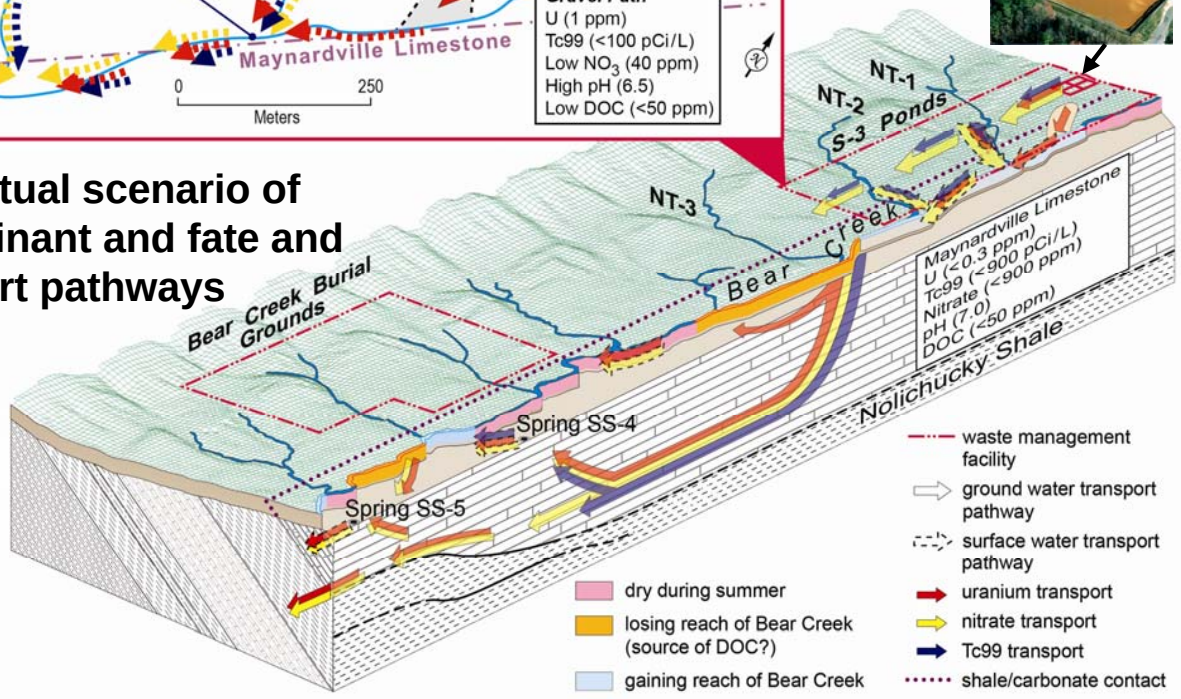
ORIFRC Research Site

OAK RIDGE National Laboratory

We focus on several *secondary source zones* and *major flow pathways* that represent a range of scales. These source zones and pathways contain numerous *transition zones* characterized by pronounced shifts in hydrology, geochemistry, and/or microbiology.

Multi-scale in situ research





Conceptual scenario of contaminant and fate and transport pathways

(1) S-3 Ponds secondary source ***(pH 3.2, GW U ~ 60 ppm, Solid U ~ 1000 ppm)***

(2) Carbonate gravel secondary source ***(pH 7.0, GW U ~ 1 ppm, Solid U ~ 14,000 ppm)***

(3) Low pH shale/saprolite pathway ***(strike parallel)***

(4) Neutral pH carbonate gravel pathway ***(engineered platform for tank farm)***

(4) Neutral pH Maynardville Limestone pathway ***(deep karst bedrock)***

(6) Recharge ***(driven by transient and spatially variable rainfall)***

Approach

Task A: Define flowpaths and heterogeneities that control the fate and transport of contaminant plumes

- *Link geophysical responses to media properties, geochemistry, and hydrology*

- *Three dimensional characterization of local and watershed scale contaminant plumes*

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

- *Novel techniques to assess the impacts of coupled pH, redox conditions, microbial activity, reactivity, etc. on U, Tc, and NO_3 natural attenuation*

- *New methods to assess the impacts of recharge, antecedent moisture conditions on spatial and temporal plume dynamics*

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

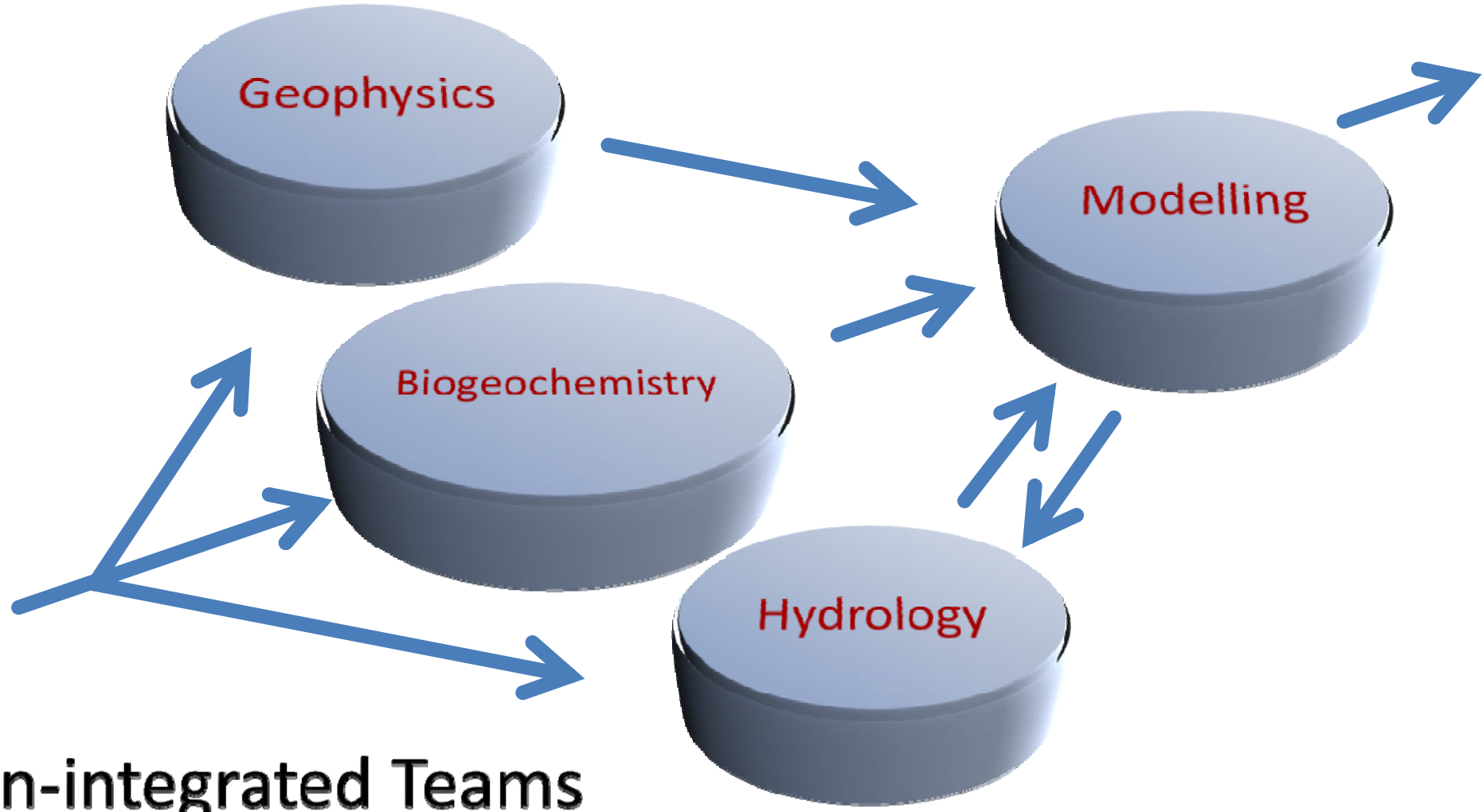
- *Targeted manipulation experiments*: sustained bioreduction, pH adjustment, organo-phosphate amendments, slow release substrate amendments. Assessing the *propensity for contaminant stability and remobilization.*

Task D: Multiprocess and multiscale numerical modeling and data analysis

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

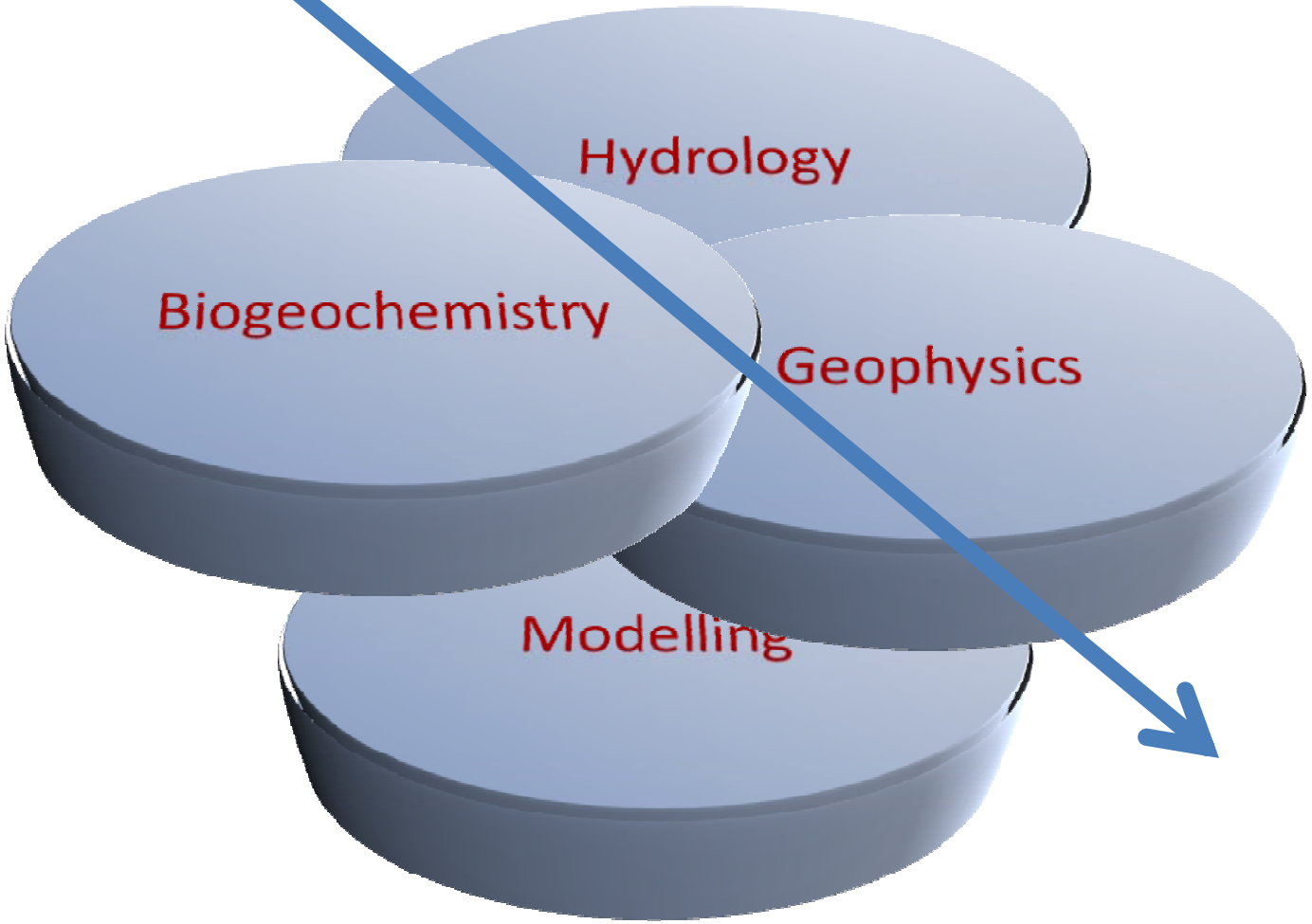
- *Site wide modeling* (HydroBioGeoChem) / upscaling and model accuracy (Tasks A-C results, geophysics, hydro-bio-geochemical processes).

Traditional Site Characterization/Monitoring



- Non-integrated Teams
- Non-integrated Analysis
- Non-integrated Incorporation into Modeling

Our Approach



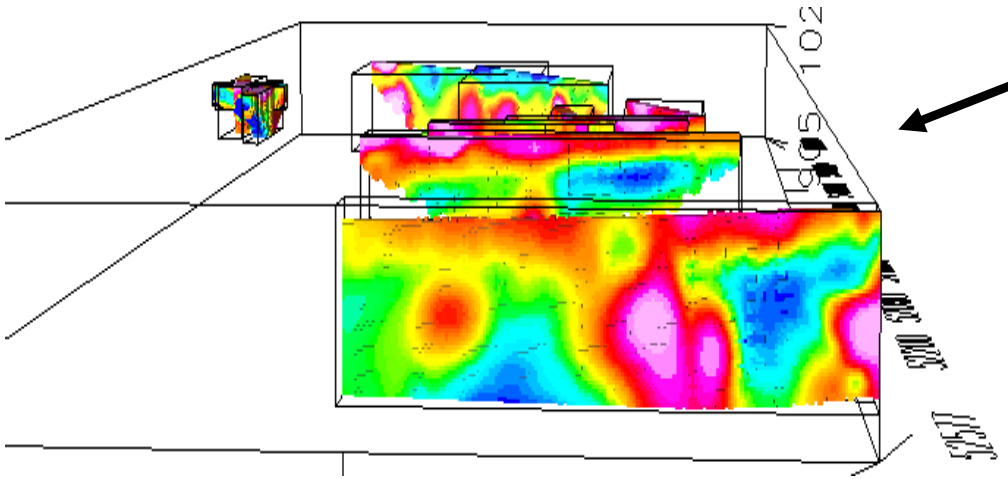
- Integrated Teams
- Integrated Analysis
- Integrated Incorporation into Modeling

Snap shot of what has been done

Quantifying Watershed Processes

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

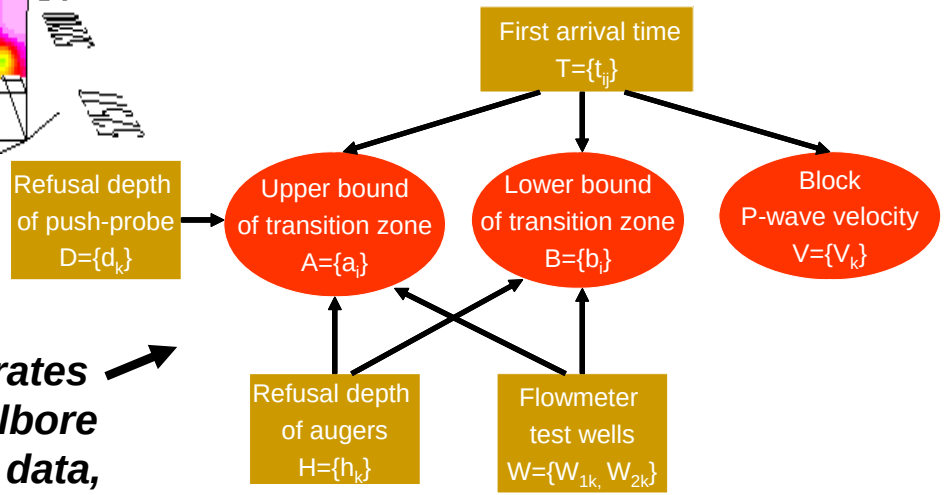
Electrical resistivity profiles down the watershed
(contaminant concentration)



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 integration 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

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Quantifying Natural Attenuation Processes: *Recharge Impacts*

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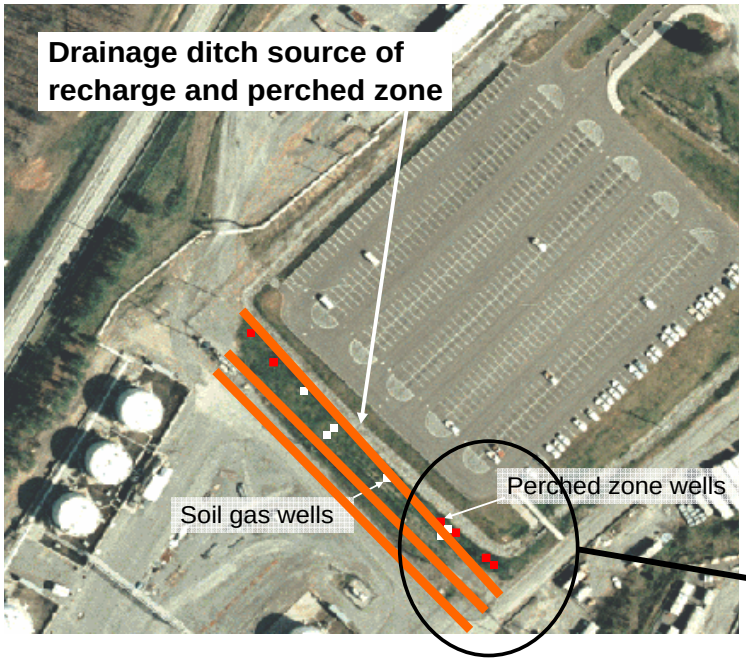
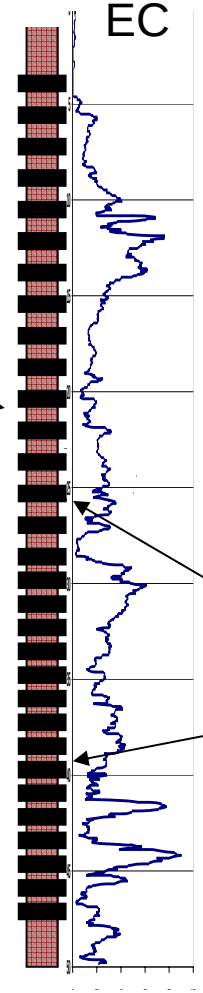
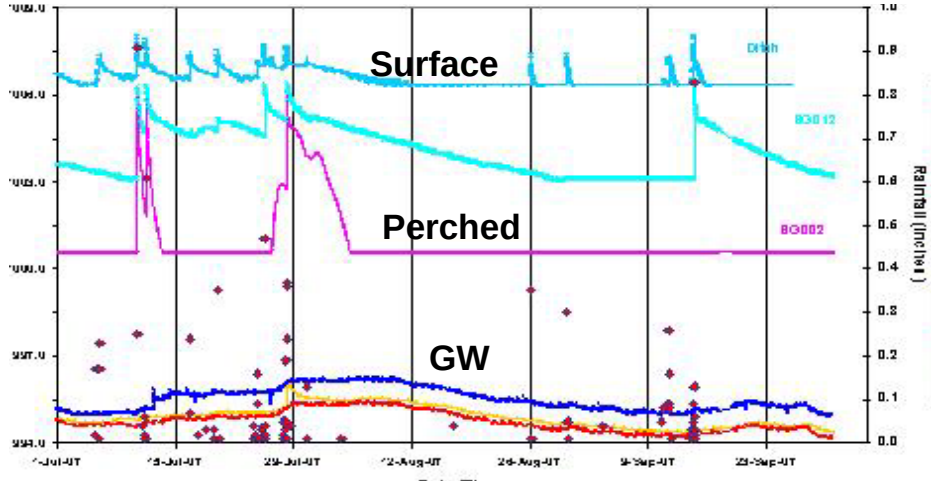
Coupling of traditional hydro-geo-micro measurements with novel real-time geophysics







Monitoring perched water table dynamics and interaction with GW as a function of storm events coupled with real time geophysics

Use of temperature profiles to quantify recharge

Long-term temporal / spatial resolution of recharge impact on contaminant dilution and plumes

TD ~52' BGS

1.1-10.1m (3.6-33.13'):15 Electrodes @0.6m spacing

10.4-14.9m (34,12-48.88') 15 Electrodes @0.3m spacing

Quantifying Natural Attenuation Processes: *Geochemical Impacts*

Oak Ridge Integrated Field Research Challenge

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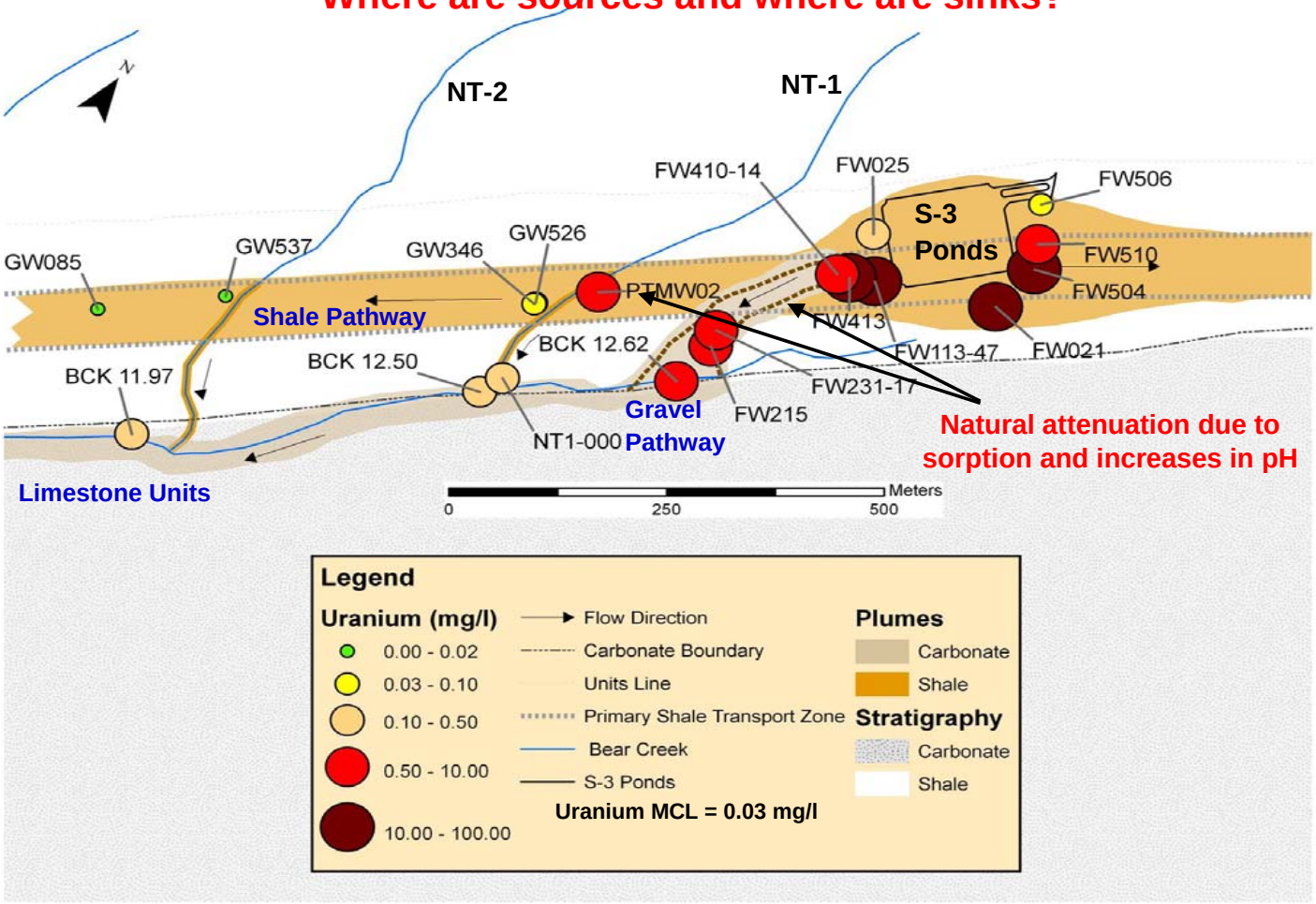
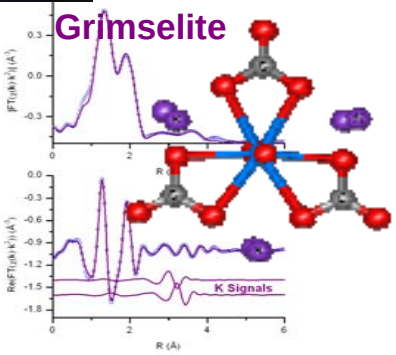
Coupling of traditional borehole assessment with novel real-time geophysics (watershed processes slide)

Uranium Concentration Down the Valley

What are the kinetics and mechanisms of uptake and release?
Where are sources and where are sinks?



X-ray Absorption Spectroscopy defines U solid phase speciation

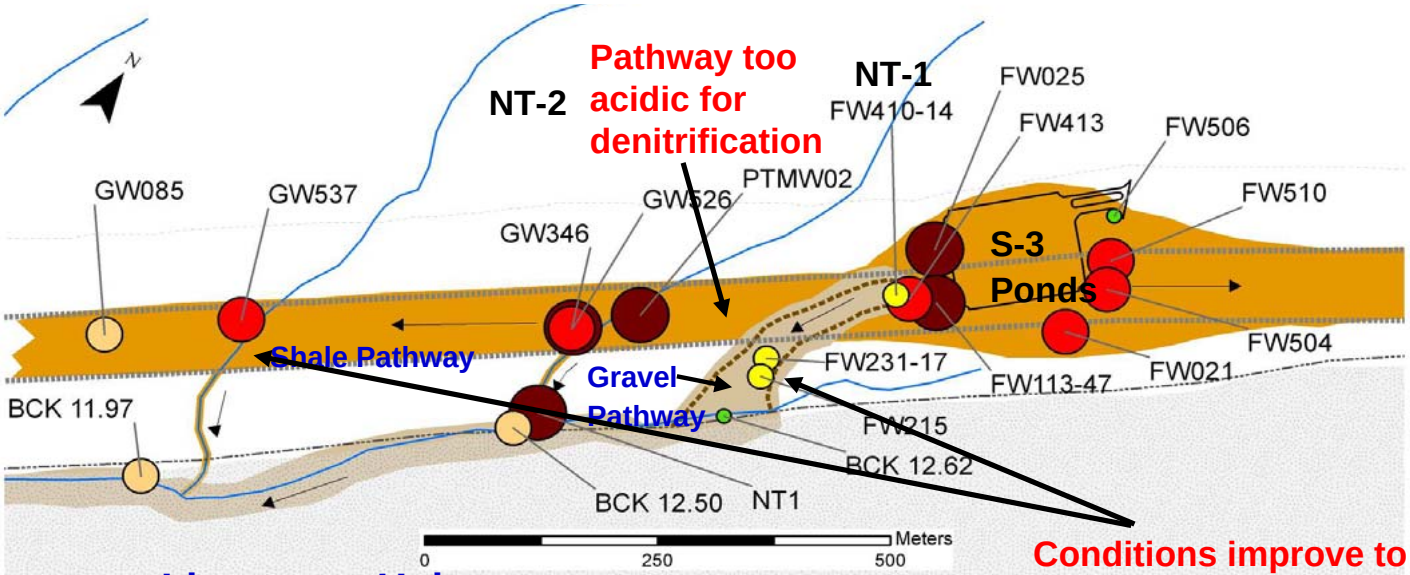


Quantify *in situ* contaminant behavior: *a better way to assess in situ kinetic reactions*

Quantifying Natural Attenuation Processes: *Microbial Impacts*

Coupling of traditional borehole assessment with novel real-time geophysics (watershed processes slide)

Nitrate Concentration Down the Valley



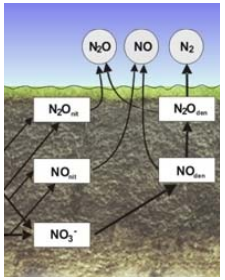
Conditions improve to support microbial activity (e.g. pH ~ 7). Indication of natural attenuation via denitrification

Dissolved gases: CO_2, N_2, O_2, N_2O, CH_4, H_2, and CO.

Stable isotopes: $^{15}N/^{14}N$ in nitrate, N_2O, and N_2; $^{18}O/^{16}O$ in nitrate; $^{2}H/^{1}H$ in dissolved H_2 gas; and $^{13}C/^{12}C$ in acetate, citrate, natural DOC, CO_2, and CH_4

qPCR that allow for quantitative estimates of population numbers for important metal reducing FRC taxa (*Geobacter, Anaeromyxobacter* & others) and functional genes involved in processes of interest (dsrA, gltA and nirK/S)

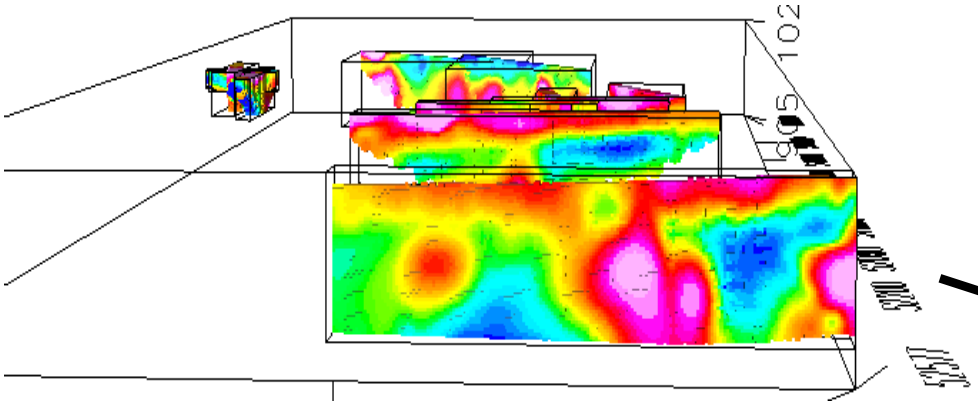
A new ***tRFLP*** methods for rapid screening of overall community diversity and activity

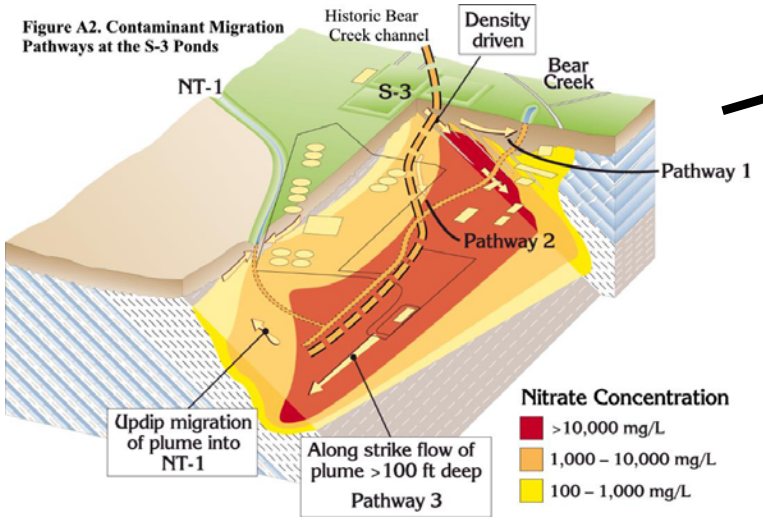


Watershed scale modeling

Multiprocess and Multiscale Numerical Modeling and Data Analysis

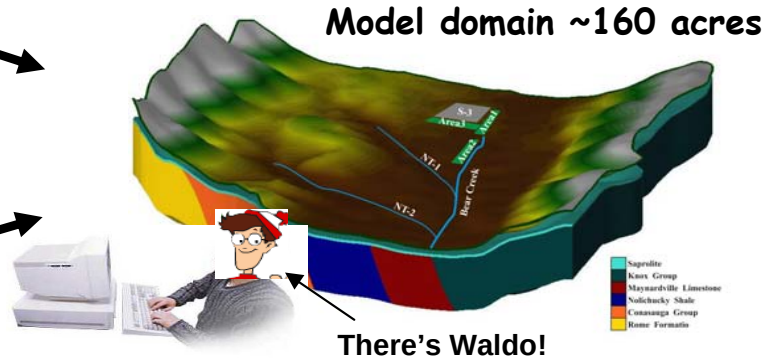
Coupling geophysical plume-scale flow architecture and recharge processes with traditional borehole hydrology, geochemistry, and microbiology for calibration and validation of the site wide numerical model





Multiscale, Multiprocess Site Wide Modeling

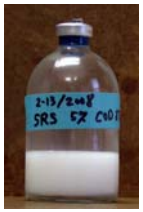
(enhanced predictive capability)



High-performance computing

Targeted Immobilization Strategies

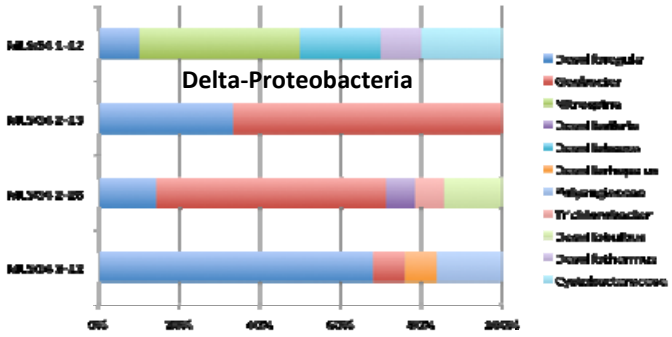
Slow Release Substrate for Long-Term Sustained U(VI) Reduction



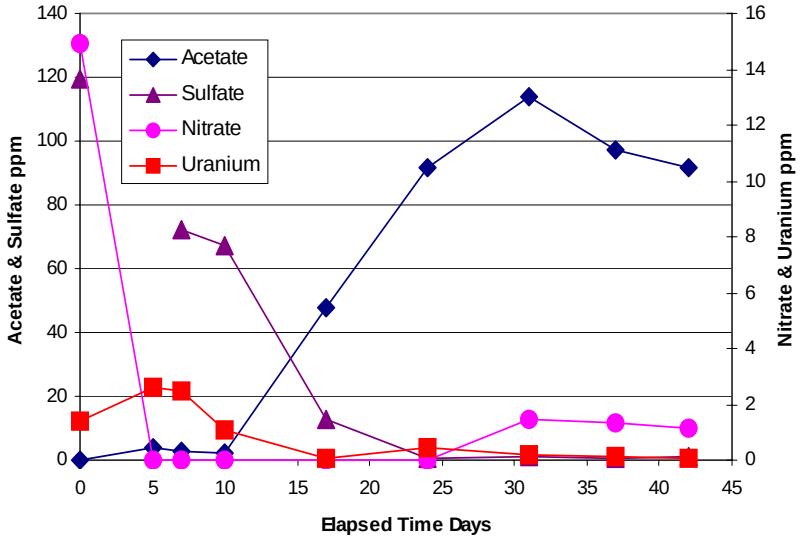
Linking geophysics, hydrology, geochemistry, microbiology, spectroscopy, and numerical modeling at the field scale

Quantifying coupled hydrological, geochemical, and microbial processes

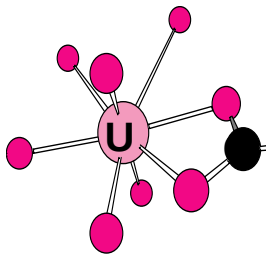
Sustained U(VI) reduction using Slow Release Substrate



DP13 (downgradient)



Geophysical monitoring coupled with spatially intensive borehole macroscopic measurements



Contaminant speciation with interfacial surface spectroscopy

Dissolved gases

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Targeted Immobilization Strategies

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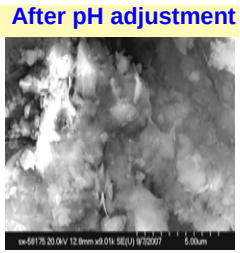
Subsurface pH adjustment for the sustained co-precipitation of U(VI) and Tc (talking advantage of high aluminum concentrations on solid and in solution)

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

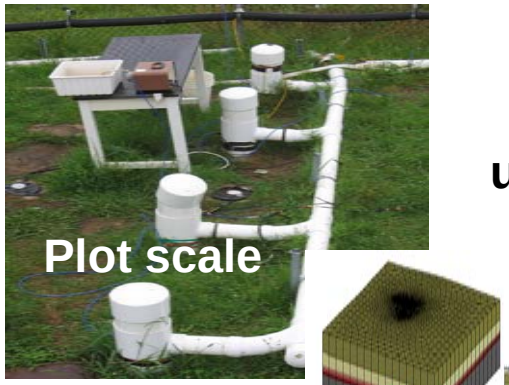
Polymers preferential sorb to the solid phase.

Serve as a template for the sorption of more Al. Thus the piling on effect which seals the U and Tc within precipitated phase

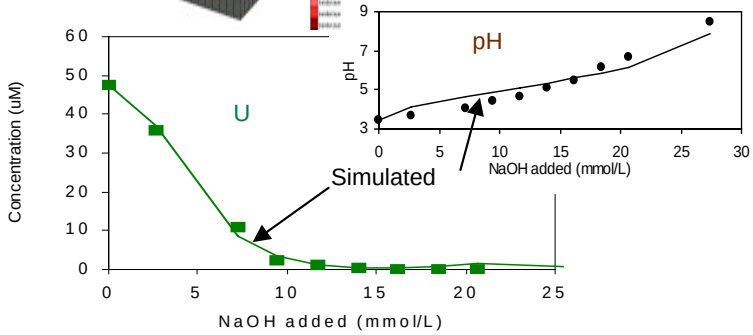
Large cores with coupled molecular analyses to define rates and mechanisms of U and Tc sequestration

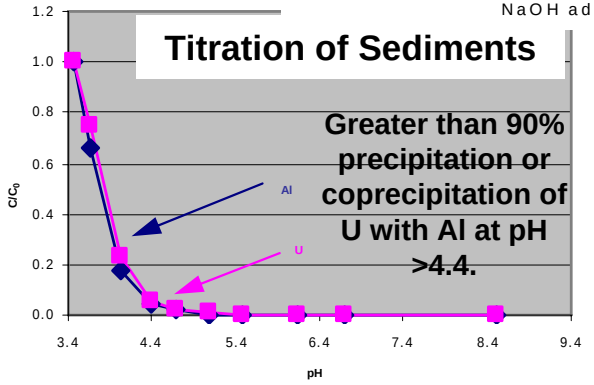


SEM of Al-U coprecipitates on ORIFC sediment (formation of sparingly soluble species)



Experimental upscaling to field





Multiscale coupled processes modeling

Data Management, Information Sources, and Technology Transfer

Characterization and other data of wide appeal are being loaded into a central database

- **Data is searchable and accessible via a Web interface.**
- **Map-based queries are available.**

Data resulting from investigator experiments will be documented using a Web-based metadata entry tool

- **The metadata will contain hyperlinks to the associated data.**
- **Each investigator will create their own data sets in consultation with IFRC-ORFRC data manager.**

A publicly accessible ORIFRC website (http://www.esd.ornl.gov/orifrc)

Dozens and dozens and dozens etc. of manuscripts and scientific presentations. Site-wide conceptual and numerical models continue to be improved. ORIFC samples have been sent to many independent ERSD researchers.

The project Field Research Manager has been active participants in the DOE Oak Ridge Reservation Closure Project Team in order to ensure remediation planning needs are addressed and technical insights are transferred into DOE remediation efforts.

Oak Ridge IFRC Plenary Session

Complete

1) "Overview of the Oak Ridge Integrated Field Research Challenge Project" 20 min (Speaker – Philip Jardine ORNL)

2) "Geophysical Quantification of Plume-Scale Flow Architecture and Recharge Processes" 25 min (Speaker – Susan Hubbard LBNL)

3) "Bioreduction of Uranium with Slow Release Substrates" 25 min (Speaker – Watson ORNL)

4) "Microbial Community Response Parallels Uranium Immobilization and Remobilization during *In Situ* Field Manipulation" 25 min (Speaker - Joel Kostka FSU)

5) "Sequestering Uranium and Technetium through Co-precipitation with Aluminum in ORIFRC Acidic Sediments" – 25 min Speaker – Baohua Gu ORNL)