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
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

You are here

ORIFC Site

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.*
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 DOE 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, NO₃)

- **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
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**

(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)*

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

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


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

Looking Eastward, toward S-3 Ponds

Electrical resistivity profiles down the watershed (contaminant concentration)

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.
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.

Coupling of traditional hydro-geo-micro measurements with novel real-time geophysics.
Quantifying Natural Attenuation Processes: Geochemical Impacts

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?

Natural attenuation due to sorption and increases in pH

Legend

<table>
<thead>
<tr>
<th>Uranium (mg/l)</th>
<th>Flow Direction</th>
<th>Plumes</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.00 - 0.02</td>
<td>Carbonate Boundary</td>
<td>Carbonate</td>
</tr>
<tr>
<td>0.03 - 0.10</td>
<td>Units Line</td>
<td>Shale</td>
</tr>
<tr>
<td>0.10 - 0.50</td>
<td>Primary Shale Transport Zone</td>
<td>Stratigraphy</td>
</tr>
<tr>
<td>0.50 - 10.00</td>
<td>Bear Creek</td>
<td>Carbonate</td>
</tr>
<tr>
<td>10.00 - 100.00</td>
<td>S-3 Ponds</td>
<td>Shale</td>
</tr>
</tbody>
</table>

Uranium MCL = 0.03 mg/l

Solid phase U >12,000 ppm gravel pathway

Grimselite

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

**Dissolved gases:** CO₂, N₂, O₂, N₂O, CH₄, H₂, and CO.

**Stable isotopes:** ¹⁵N/¹⁴N in nitrate, N₂O, and N₂; ¹⁸O/¹⁶O in nitrate; ²H/¹H in dissolved H₂ gas; and ¹³C/¹²C in acetate, citrate, natural DOC, CO₂, and CH₄

**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)

Model domain ~160 acres

There’s Waldo!

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

**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

Quantifying coupled hydrological, geochemical, and microbial processes

**Delta-Proteobacteria**

**PELCAPS**

Dissolved gases
Subsurface pH adjustment for the sustained co-precipitation of U(VI) and Tc (taking 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
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.
Here is what’s in store for you!

Oak Ridge IFRC Plenary Session

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)