



# Advances in Long-term Monitoring Technologies for Supporting Bioremediation

Haruko Wainwright, Ken Williams

06/05



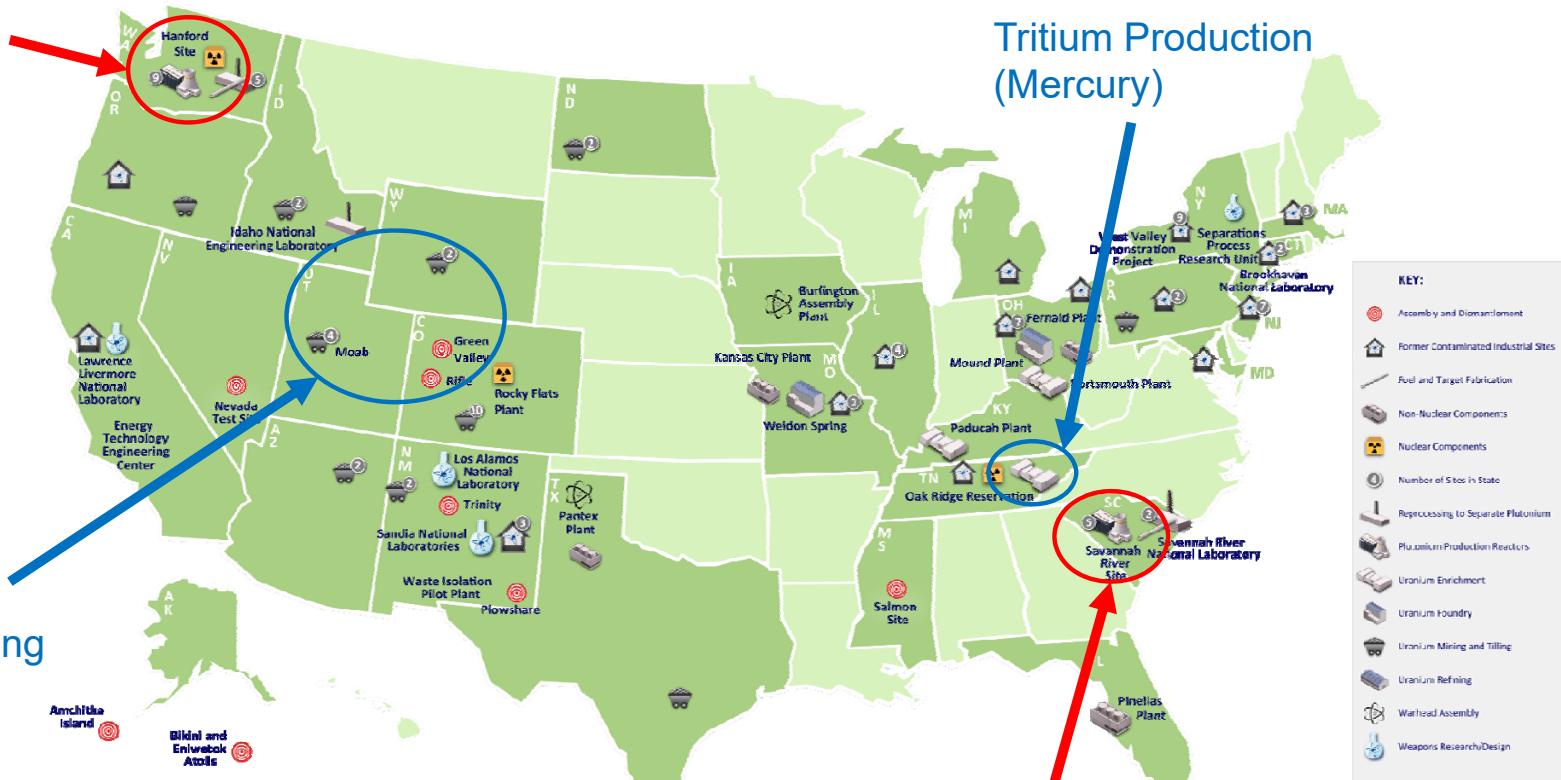
# Nuclear Weapon Production Sites



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# Nuclear Weapon Production Sites

Plutonium  
Production



Tritium Production  
(Mercury)

Plutonium  
Production



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# Key/Priority Elements of Existing Contamination

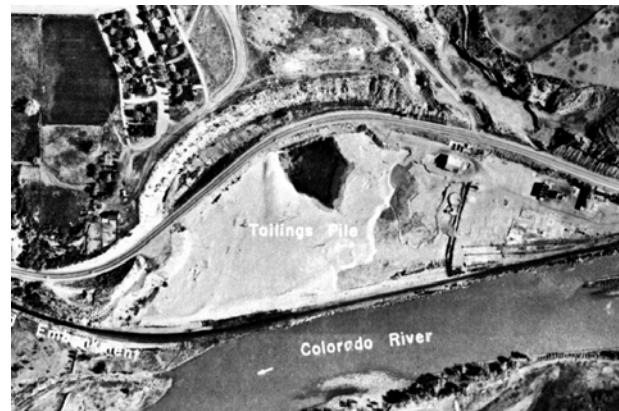


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# Low-level Radioactive Waste Disposal



Hanford 300 Area



Rifle, CO



Savannah River Site F-Area



# DOE's Rifle Site

- U.S. Department of Energy (DOE)
- Uranium/vanadium ore processing facility: 1924-1958
- 761,000 tons of ore processed
- 2,000 tons of U concentrate

Rifle, CO: Mill site in ca. 1957



- Tailings were consolidated & stabilized late 1950's & 60's
- Site stabilized; surface cover added and closure in 1996
- **GCAP:** *Natural flushing* of residual groundwater U(VI) ca.  $0.5\text{--}1.5\mu\text{M}$

# Prime Site for U Bioremediation

APPLIED AND ENVIRONMENTAL MICROBIOLOGY, Oct. 2005, p. 6308–6318  
0099-2240/05/\$08.00+0 doi:10.1128/AEM.71.10.6308–6318.2005  
Copyright © 2005, American Society for Microbiology. All Rights Reserved.

Vol. 71, No. 10

*Geomicrobiology Journal*, 28:519–539, 2011  
Copyright © Taylor & Francis Group, LLC  
ISSN: 0149-0451 print / 1521-0529 online  
DOI: 10.1080/01490451.2010.520074

## Microbiological and Geochemical Heterogeneity in an In Situ Uranium Bioremediation Field Site

Helen A. Vrionis,<sup>1\*</sup> Robert T. Anderson,<sup>1</sup> Irene Ortiz-Bernad,<sup>1</sup> Kathleen R. O'Neill,<sup>1</sup> Charles T. Resch,<sup>2</sup> Aaron D. Peacock,<sup>3</sup> Richard Dayvault,<sup>4</sup> David C. White,<sup>3</sup> Philip E. Long,<sup>2</sup> and Derek R. Lovley<sup>1</sup>

Department of Microbiology, University of Massachusetts, Amherst, Massachusetts 01003<sup>1</sup>; Pacific Northwest National Laboratory, Richland, Washington 99352<sup>2</sup>; S. M. Stoller Corporation, Lafayette, Colorado 80026<sup>4</sup>; and Center for Biomarker Analysis, University of Tennessee, Knoxville, Tennessee 37932<sup>3</sup>

## Acetate Availability and its Influence on Sustainable Bioremediation of Uranium-Contaminated Groundwater

Kenneth H. Williams,<sup>1</sup> Philip E. Long,<sup>2</sup> James A. Davis,<sup>3</sup> Michael J. Wilkins,<sup>2</sup> A. Lucie N'Guessan,<sup>2</sup> Carl I. Steefel,<sup>1</sup> Li Yang,<sup>1</sup> Darrell Newcomer,<sup>2</sup> Frank A. Spane,<sup>2</sup> Lee J. Kerkhof,<sup>4</sup> Lora McGuinness,<sup>4</sup> Richard Dayvault,<sup>5</sup> and Derek R. Lovley<sup>6</sup>

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<sup>5</sup>S.M. Stoller Corporation, Grand Junction, Colorado, USA

<sup>6</sup>Department of Microbiology, University of Massachusetts, Amherst, Massachusetts, USA

## Mineral Transformation and Biomass Accumulation Associated With Uranium Bioremediation at Rifle, Colorado

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KENNETH H. WILLIAMS,<sup>†</sup>  
MICHAEL J. WILKINS,<sup>‡</sup> AND  
SUSAN S. HUBBARD<sup>†</sup>

Earth Sciences Division, Lawrence Berkeley National Laboratory, 1 Cyclotron Road, MS 90-1116, Berkeley, California 94720, and Department of Earth and Planetary Science, University of California, Berkeley, California 94720

Received January 4, 2009. Revised manuscript received May 21, 2009. Accepted May 27, 2009.

## Geophysical Imaging of Stimulated Microbial Biomineralization

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DIMITRIOS NTARLAGIANNIS,<sup>§</sup>  
LEE D. SLATER,<sup>§</sup> ALICE DOHNALKOVA,<sup>||</sup>  
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Current Opinion in Biotechnology

Volume 24, Issue 3, June 2013, Pages 489-497



Bioremediation of uranium-contaminated groundwater: a systems approach to subsurface biogeochemistry

Kenneth H Williams<sup>1</sup> , John R Bargar<sup>2</sup>, Jonathan R Lloyd<sup>3</sup>, Derek R Lovley<sup>4</sup>

# Fundamental Understanding of Microbiology

LETTER

doi:10.1038/nature14486

## Unusual biology across a group comprising more than 15% of domain Bacteria

Christopher T. Brown<sup>1</sup>, Laura A. Hug<sup>2</sup>, Brian C. Thomas<sup>2</sup>, Itai Sharon<sup>2</sup>, Cindy J. Castelle<sup>2</sup>, Andrea Singh<sup>2</sup>, Michael J. Wilkins<sup>3,4</sup>, Kelly C. Wrighton<sup>4</sup>, Kenneth H. Williams<sup>5</sup> & Jillian F. Banfield<sup>2,5,6</sup>

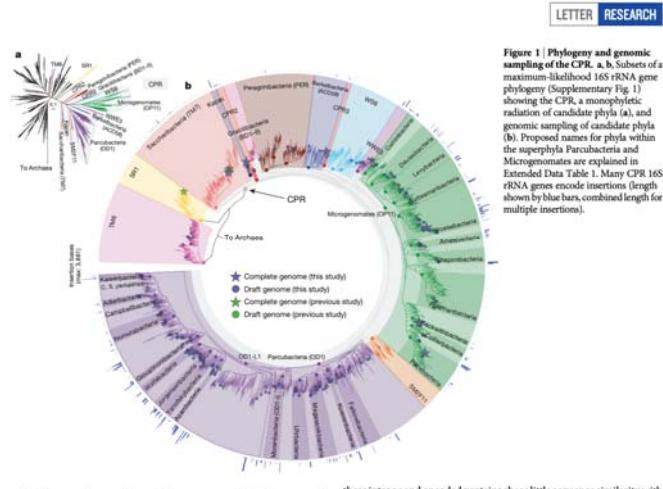
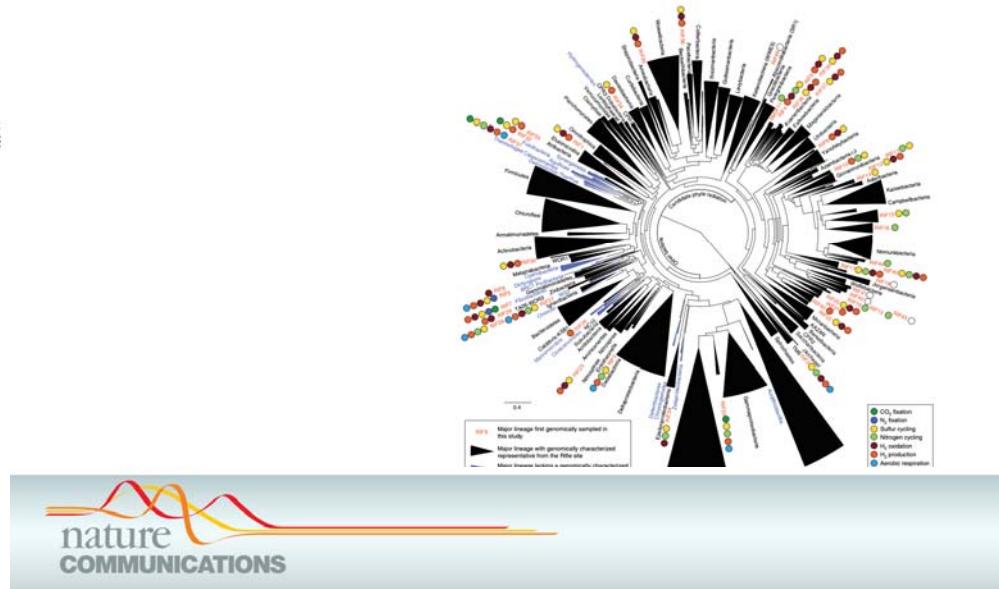


Figure 1 | Phylogeny and genomic sampling of the CPR. **a**, **b**, Subsets of a maximum-likelihood 16S rRNA gene phylogeny (Supplementary Fig. 1) showing the CPR, a monophyletic radiation of candidate phyla (**a**), and genomic sampling of candidate phyla (**b**). Prokaryotes for which within the monophyletic Parcubacteria and Microgrommatidae are explained in Extended Data Table 1. Many CPR 16S rRNA genes encode insertions (length shown by blue bars, combined length for multiple insertions).



## ARTICLE

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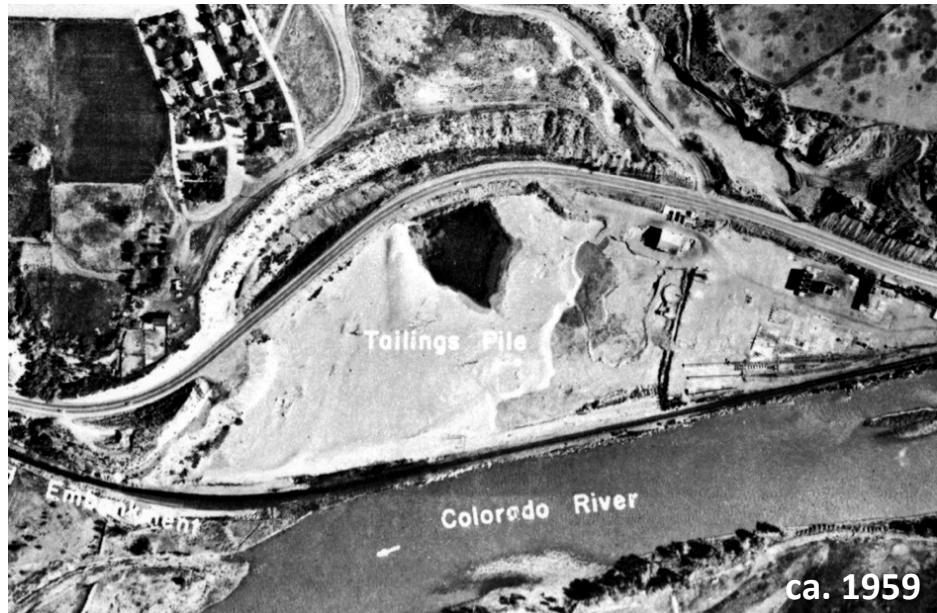
OPEN

## Thousands of microbial genomes shed light on interconnected biogeochemical processes in an aquifer system

Karthik Anantharaman<sup>1</sup>, Christopher T. Brown<sup>2</sup>, Laura A. Hug<sup>1</sup>, Itai Sharon<sup>1</sup>, Cindy J. Castelle<sup>1</sup>, Alexander J. Probst<sup>1</sup>, Brian C. Thomas<sup>1</sup>, Andrea Singh<sup>1</sup>, Michael J. Wilkins<sup>3</sup>, Ulas Karaoz<sup>4</sup>, Eoin L. Brodie<sup>4</sup>, Kenneth H. Williams<sup>4</sup>, Susan S. Hubbard<sup>4</sup> & Jillian F. Banfield<sup>1,4</sup>



# Milling History: 1950



**Union Carbide  
Corporation**



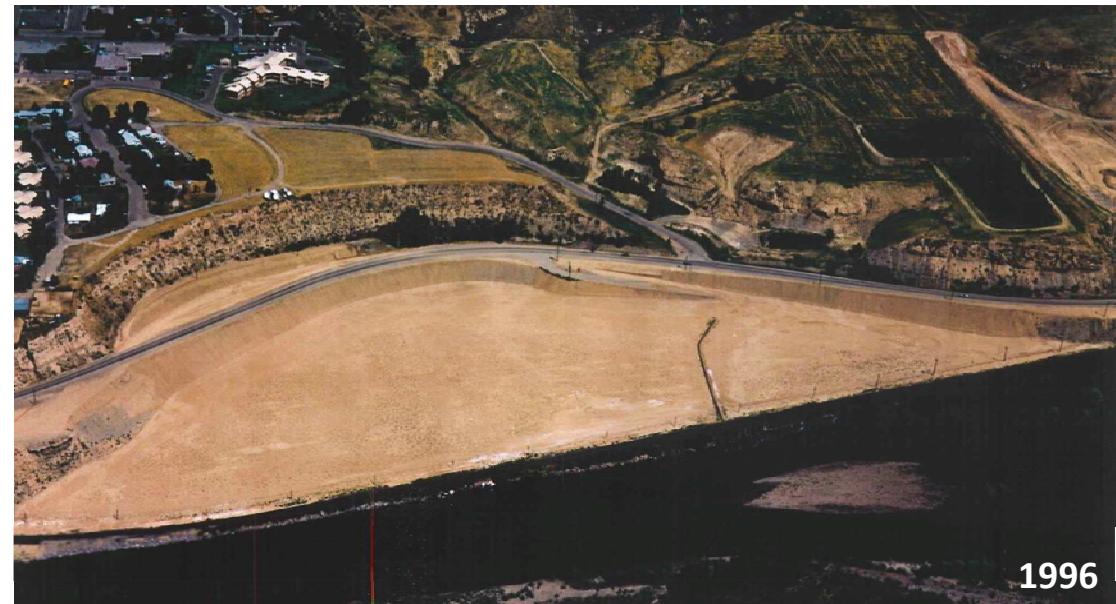
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# Milling History: 1960 - 1996



1967

**Tailings consolidated and stabilized by 1967;  
new ores processed at New Rifle site after 1958**

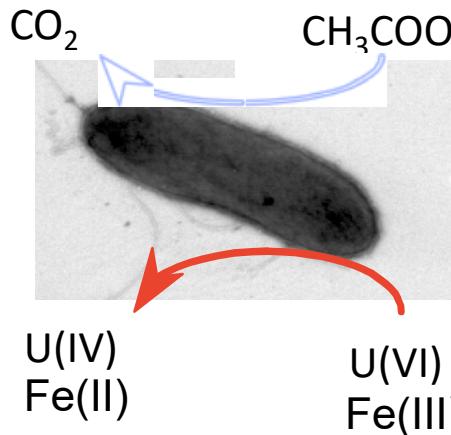
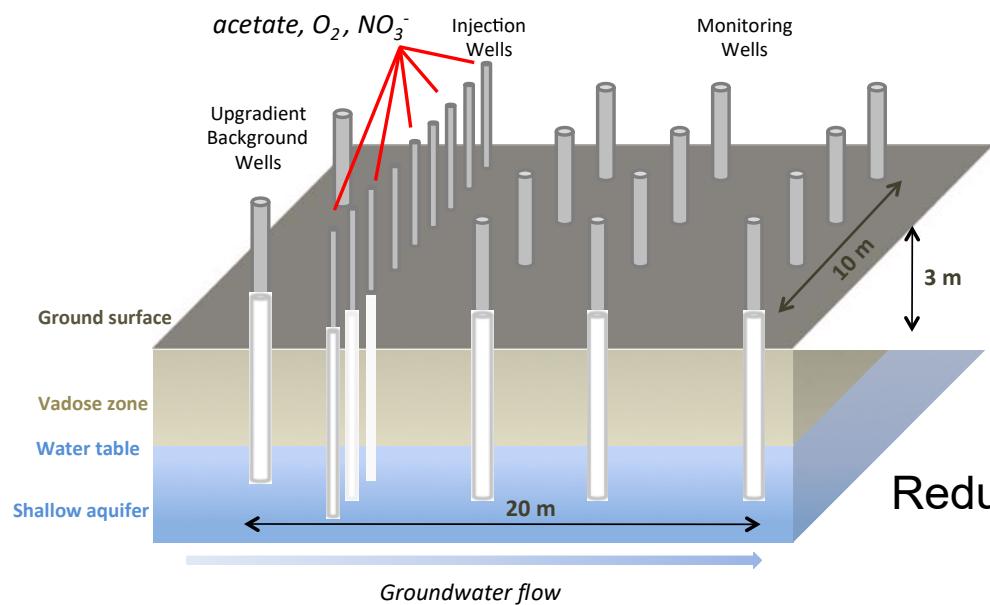


1996

**Site ‘closure’ and  
remediation  
finished in 1996.**

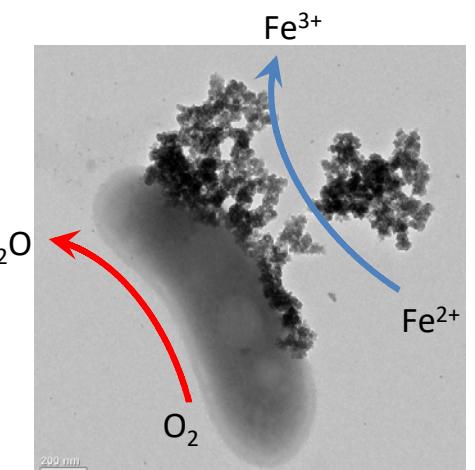


# Bioremediation Experiments



Reductive vs. oxidative pathways

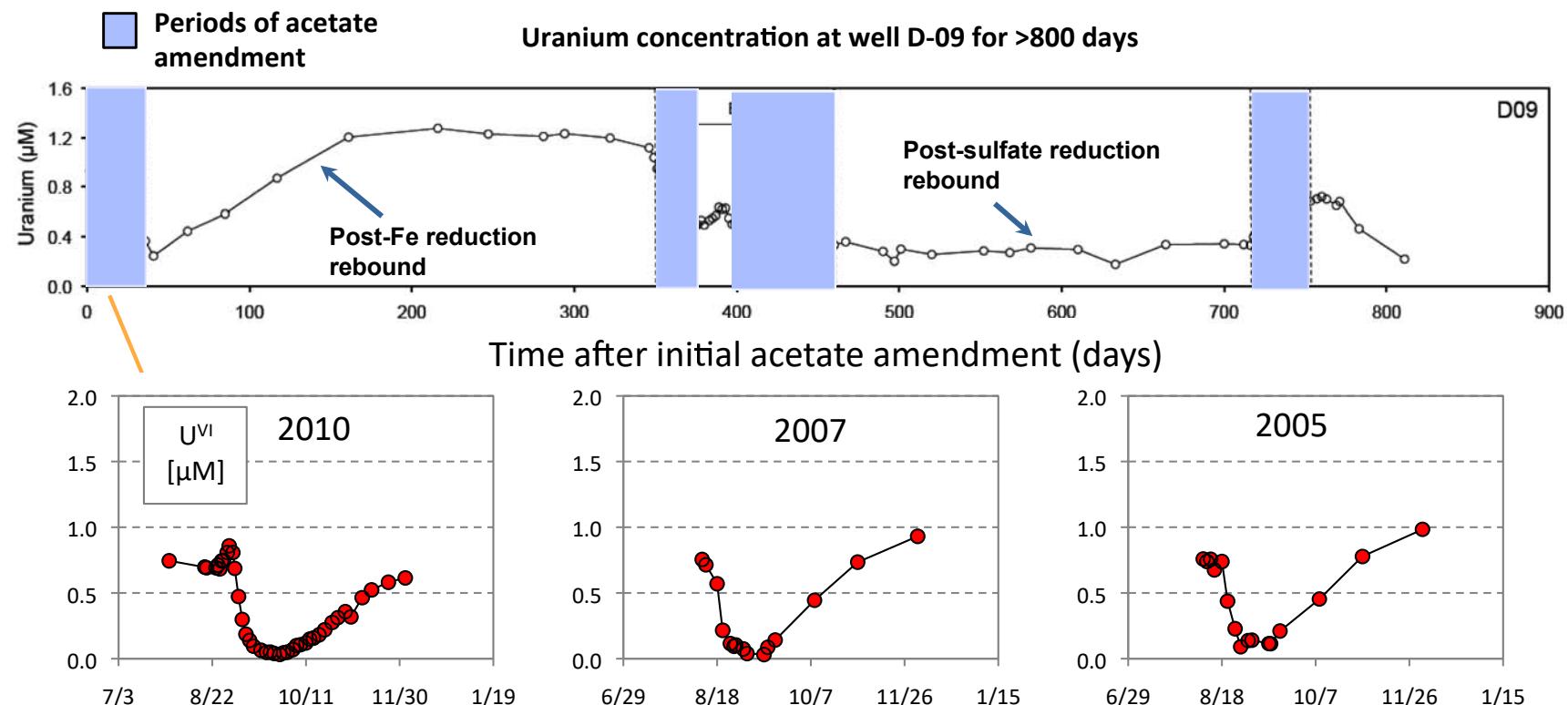
- **Amplify** pathways of interest (microbial, geochemical, mineralogical)
- Enable **high resolution sampling**
- Experimentally **tractable timelines**



Courtesy: Clara Cha

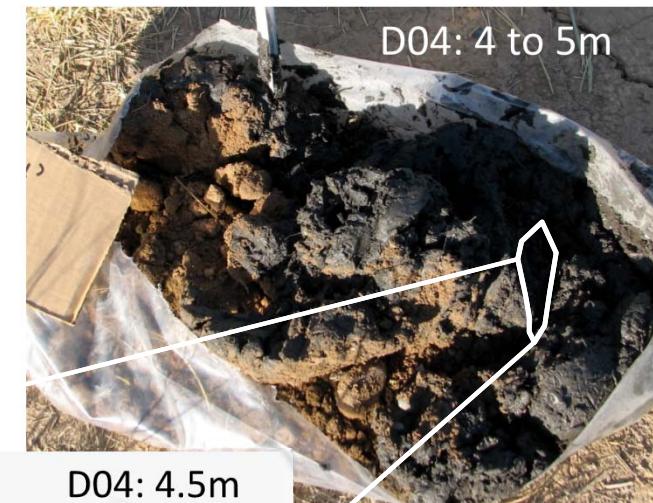
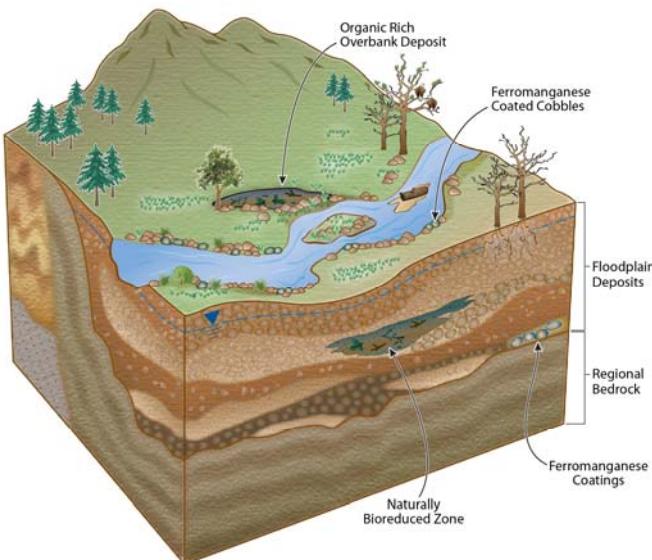


# Bioremediation: Acetate Injection



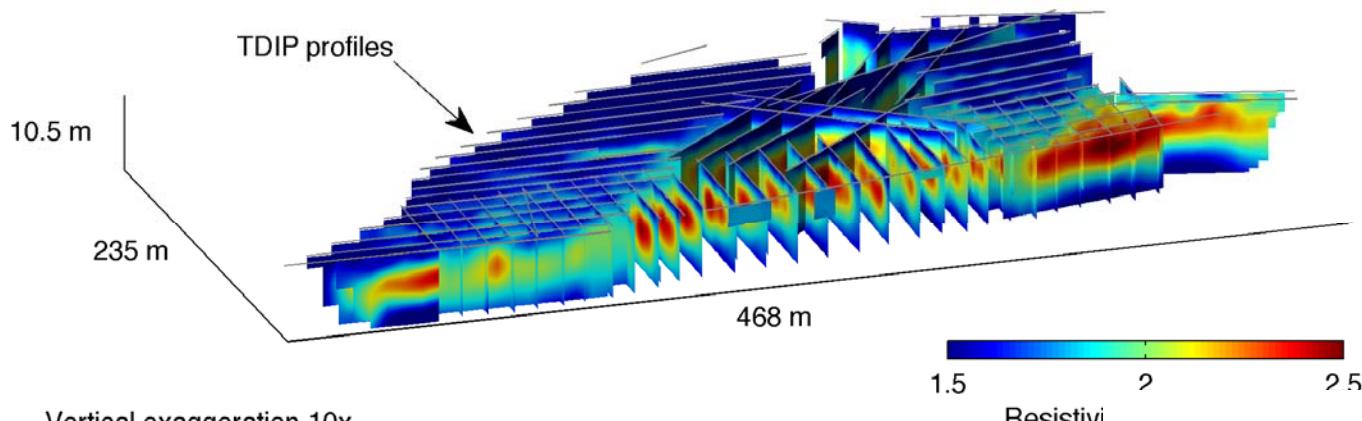
# Naturally Reduced Zones

- Drilling delineated regions of refractory organic carbon accumulation (e.g. twigs, roots, cones, etc.)
- Visibly reduced sediments (e.g. FeS)
- Elevated uranium levels *likely* due to same processes catalyzed by acetate injection

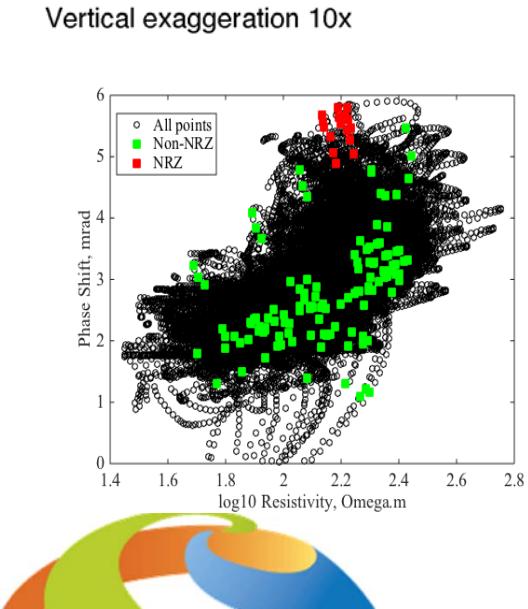


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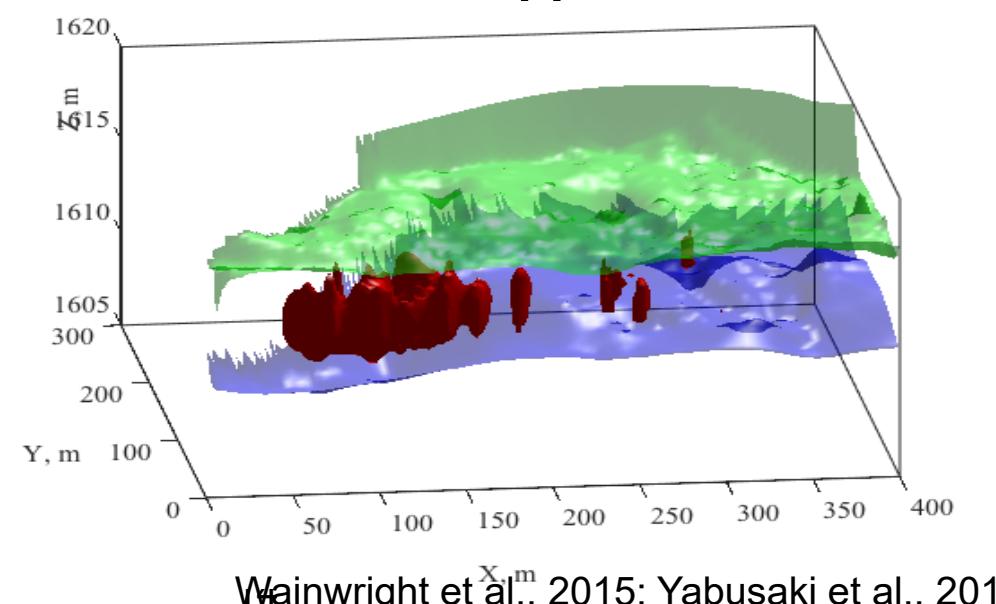
# 3D Mapping of NRZ



Mapped NRZs

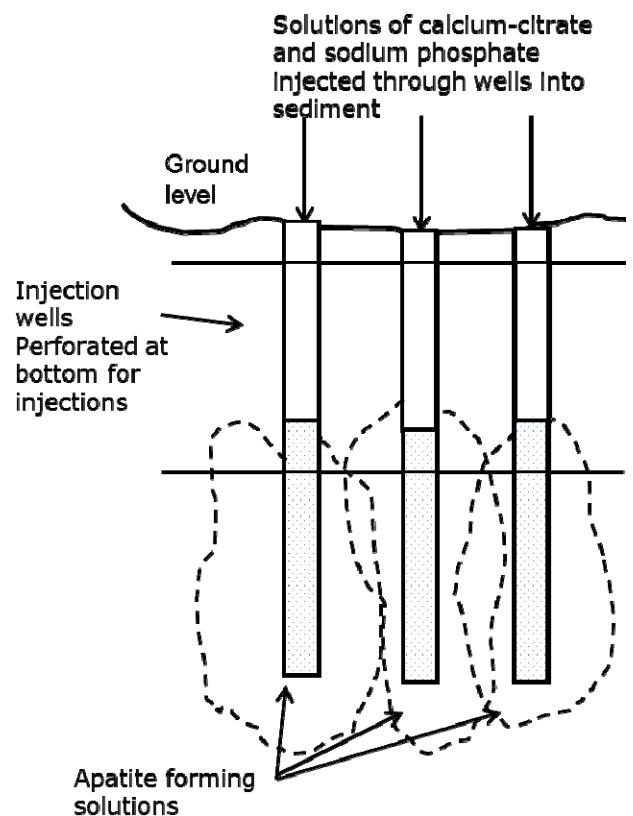
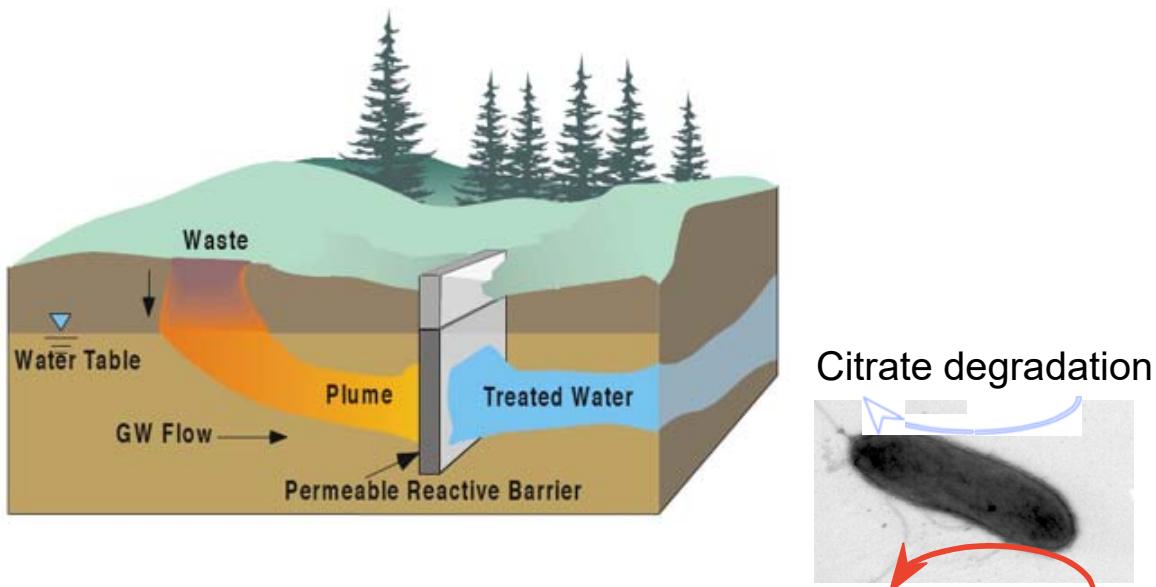


- Induced polarization method
- Phase shift
- Resistivity



# More Effective Alternative?

Apatite-based “Chemically Induced” Permeable Reactive Barrier (PRB) Technology



Slow release:  $\text{calcium}-(\text{citrate})_2 + \text{sodium phosphate} \rightarrow \text{apatite}$

# Hydroxyapatite as a PRB Material

- 3-dimensional lattice of calcium phosphate,  $\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2$
- Very stable; extremely low solubility.
- Can sequester a wide variety of radionuclides, heavy metals and other contaminants through substitution into the structure or sorption onto the surface as metal phosphate compounds.
- Immobilization and sequestration of uranium as an oxidized ( $\text{U}^{6+}$ ) form → less prone to (re)oxidative dissolution
- **Can be formed in situ by solution injection in the subsurface (No need to trench)**



Sandia  
National  
Laboratories

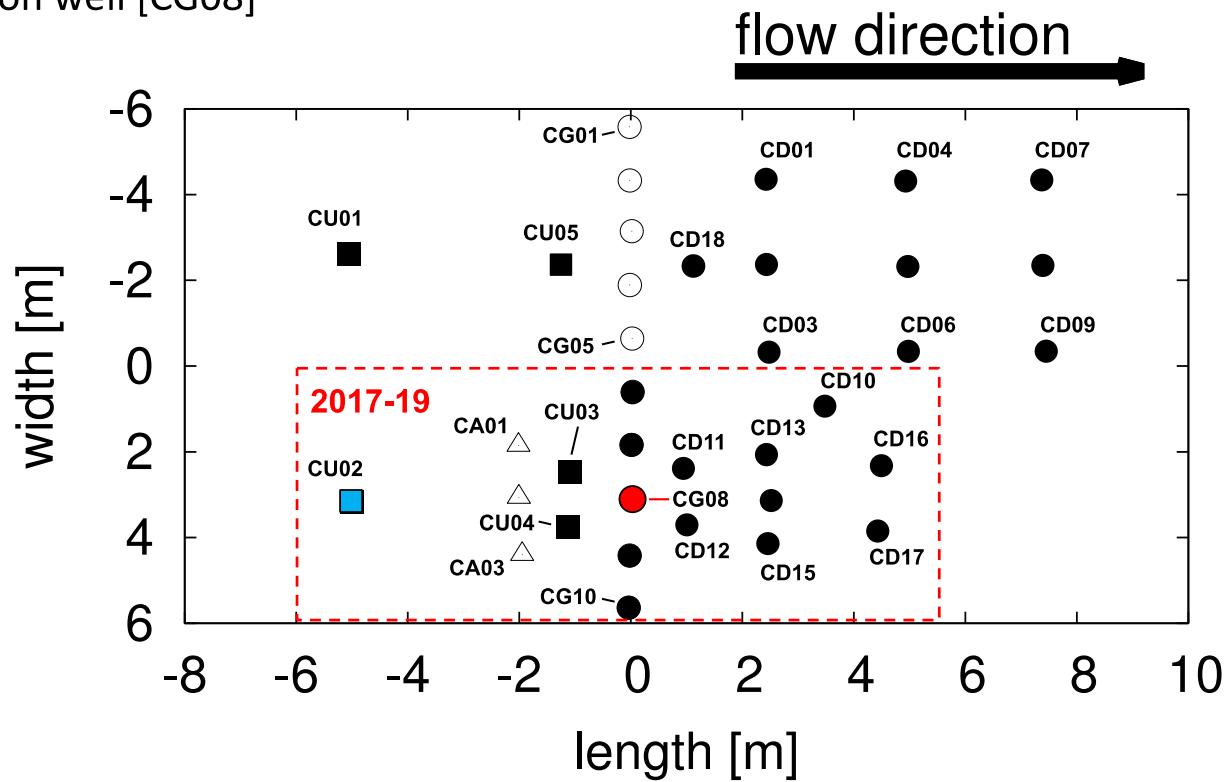


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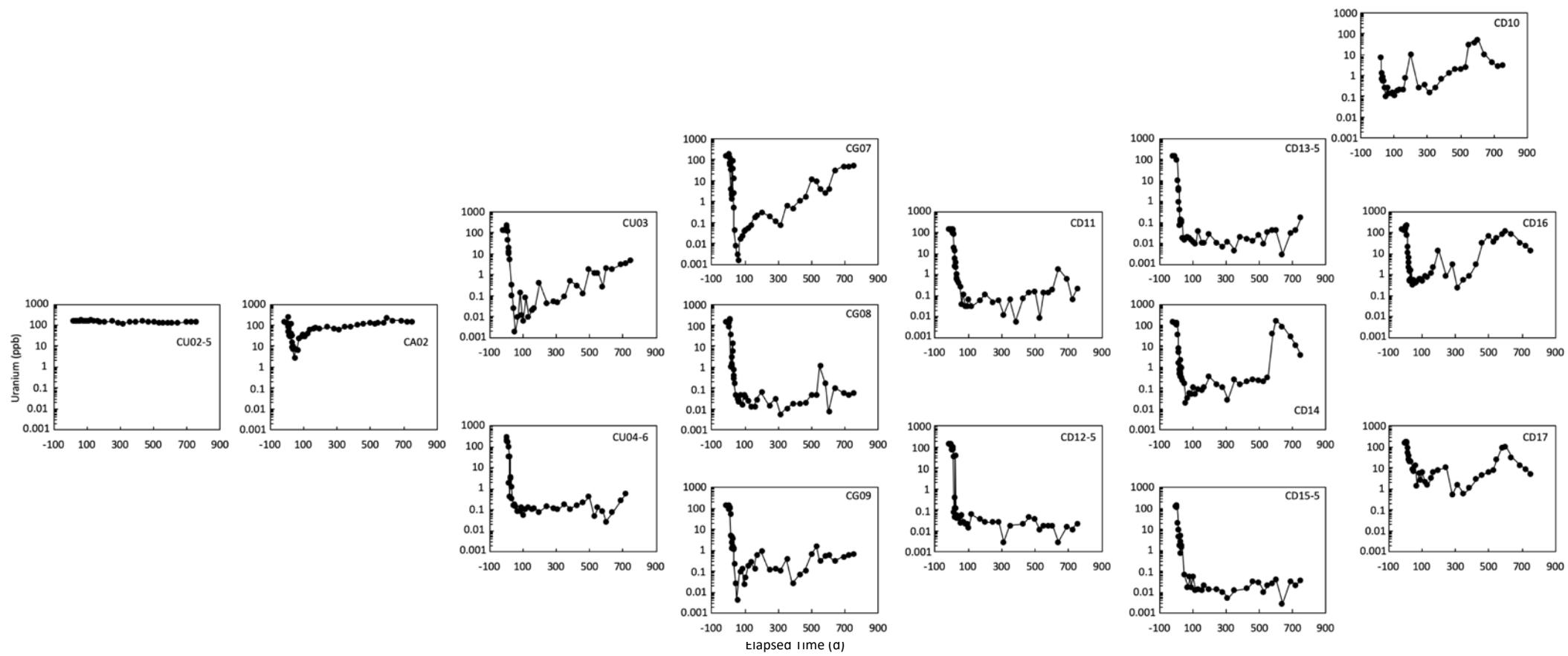
# Experiment Design

■ Upgradient control well [CU02]

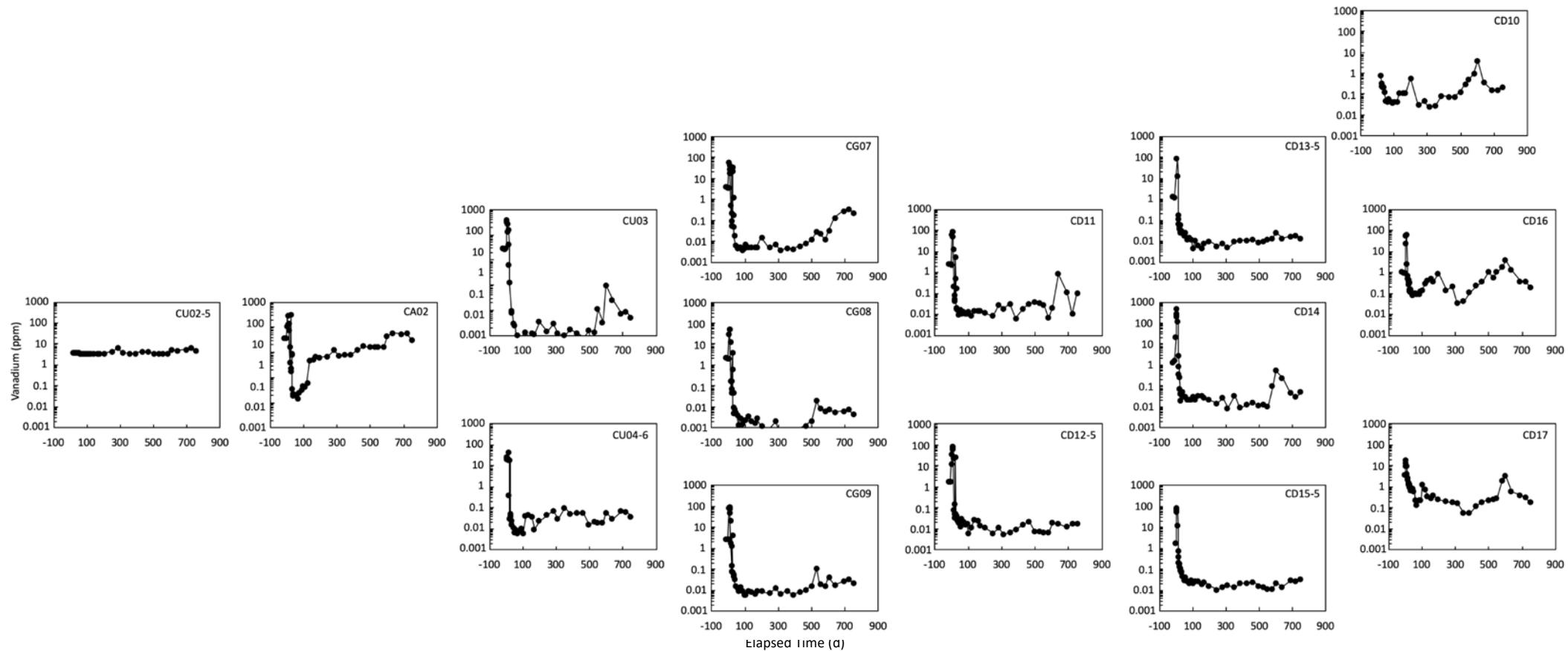
● Injection well [CG08]



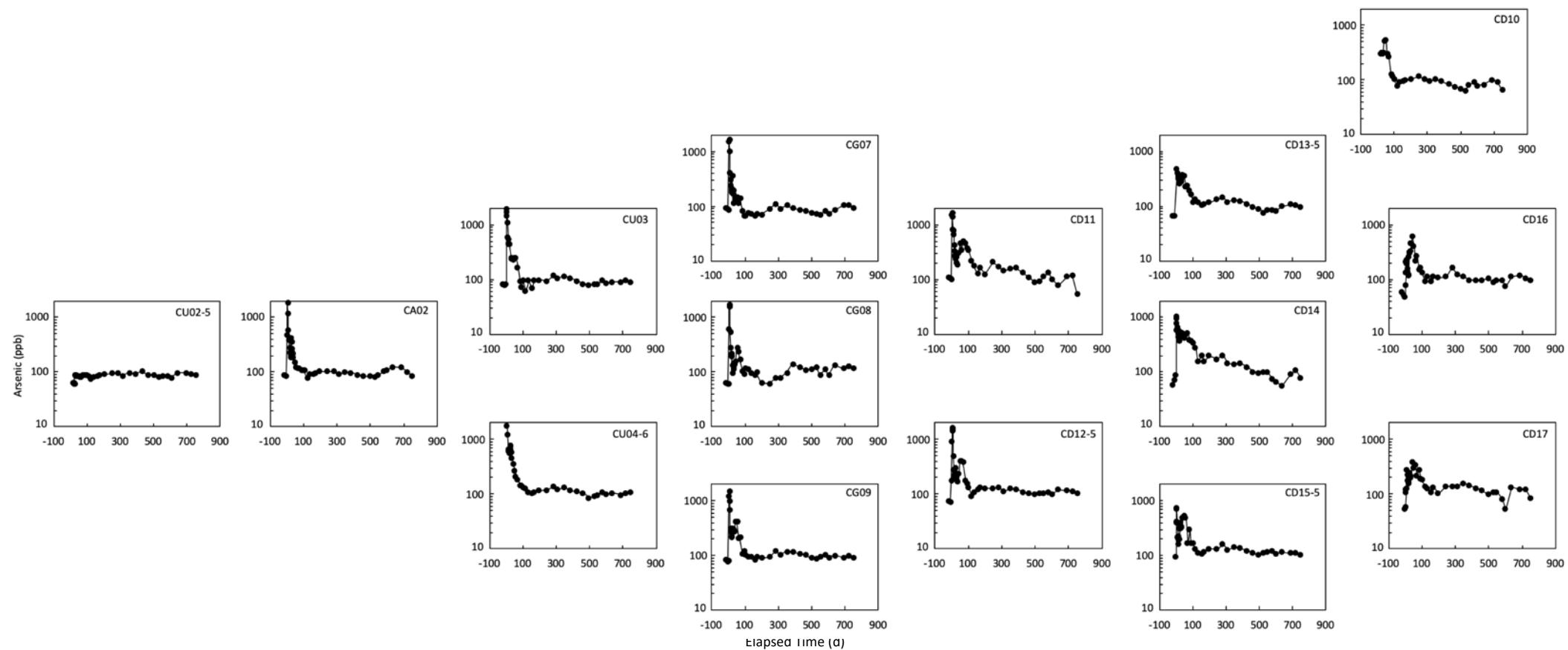
# Uranium (ppb)



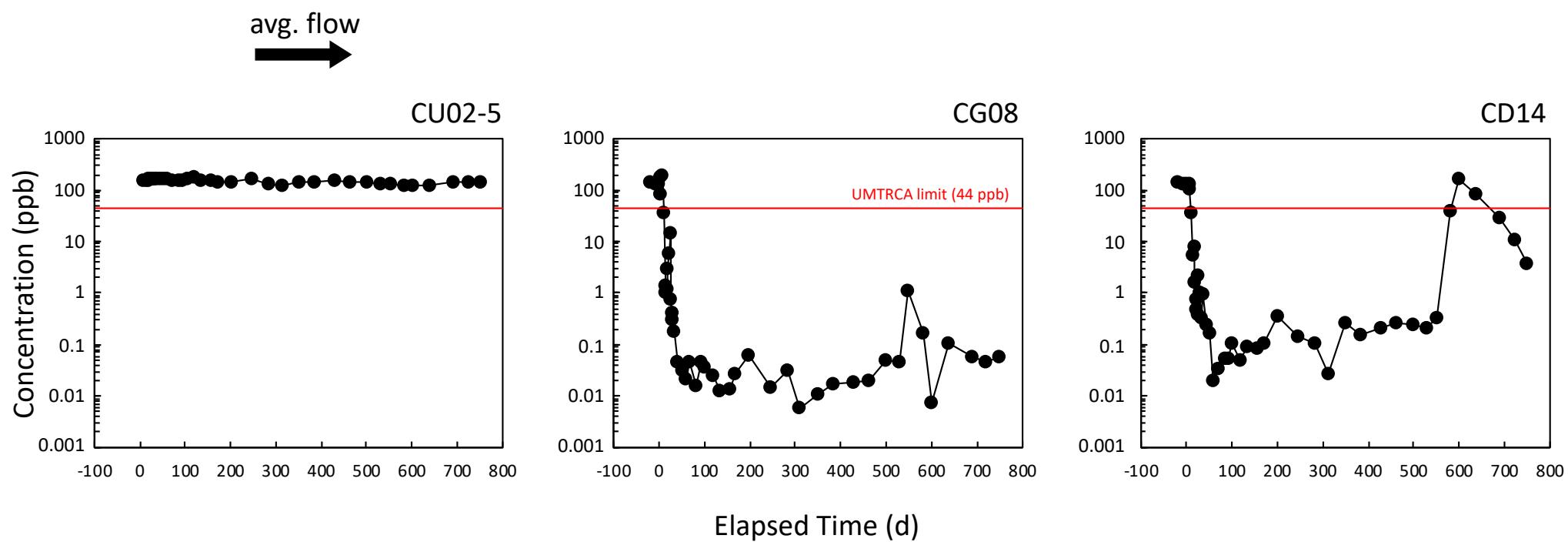
# Vanadium (ppm)



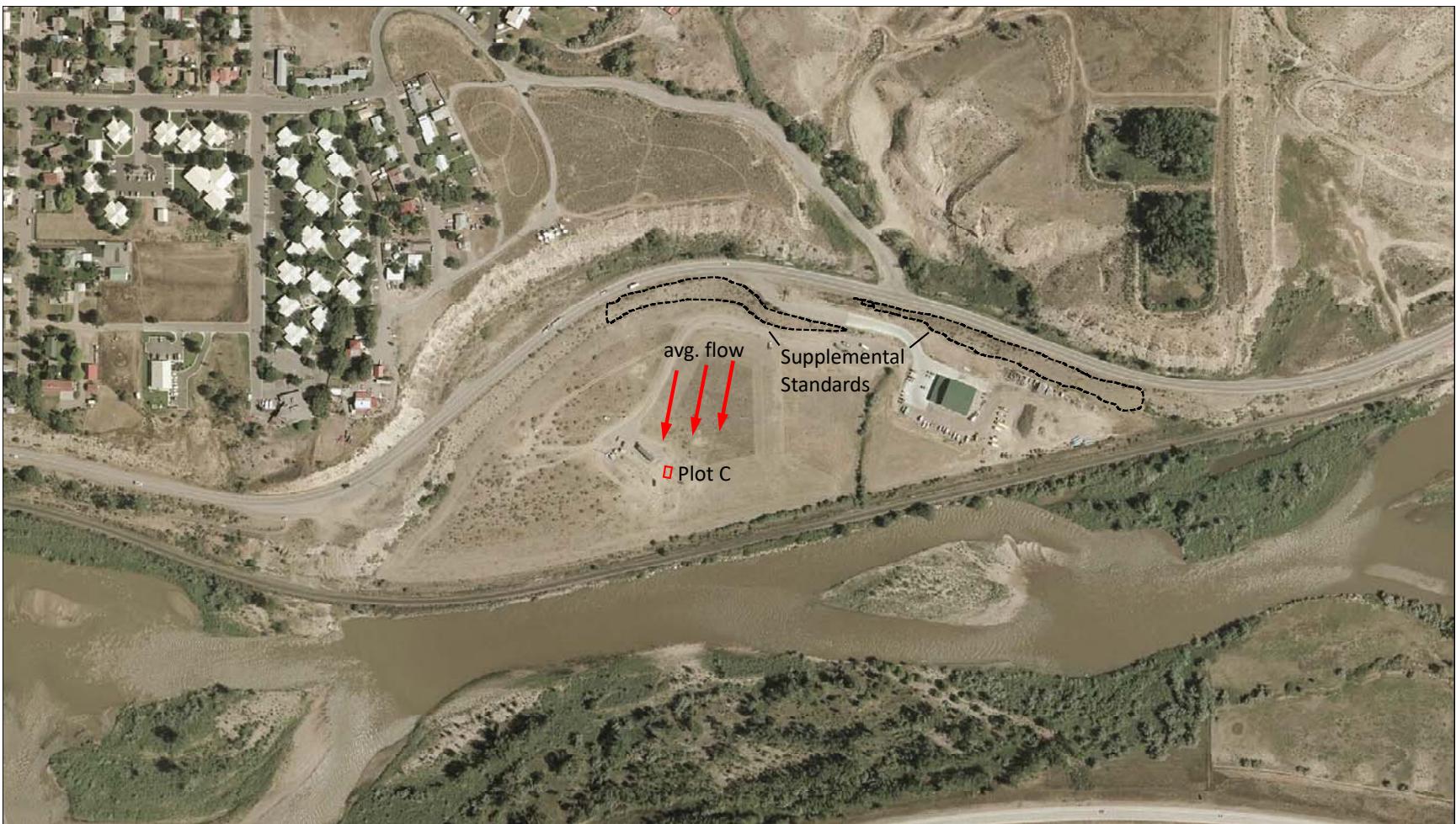
# Arsenic (ppb)



# Uranium (ppb)

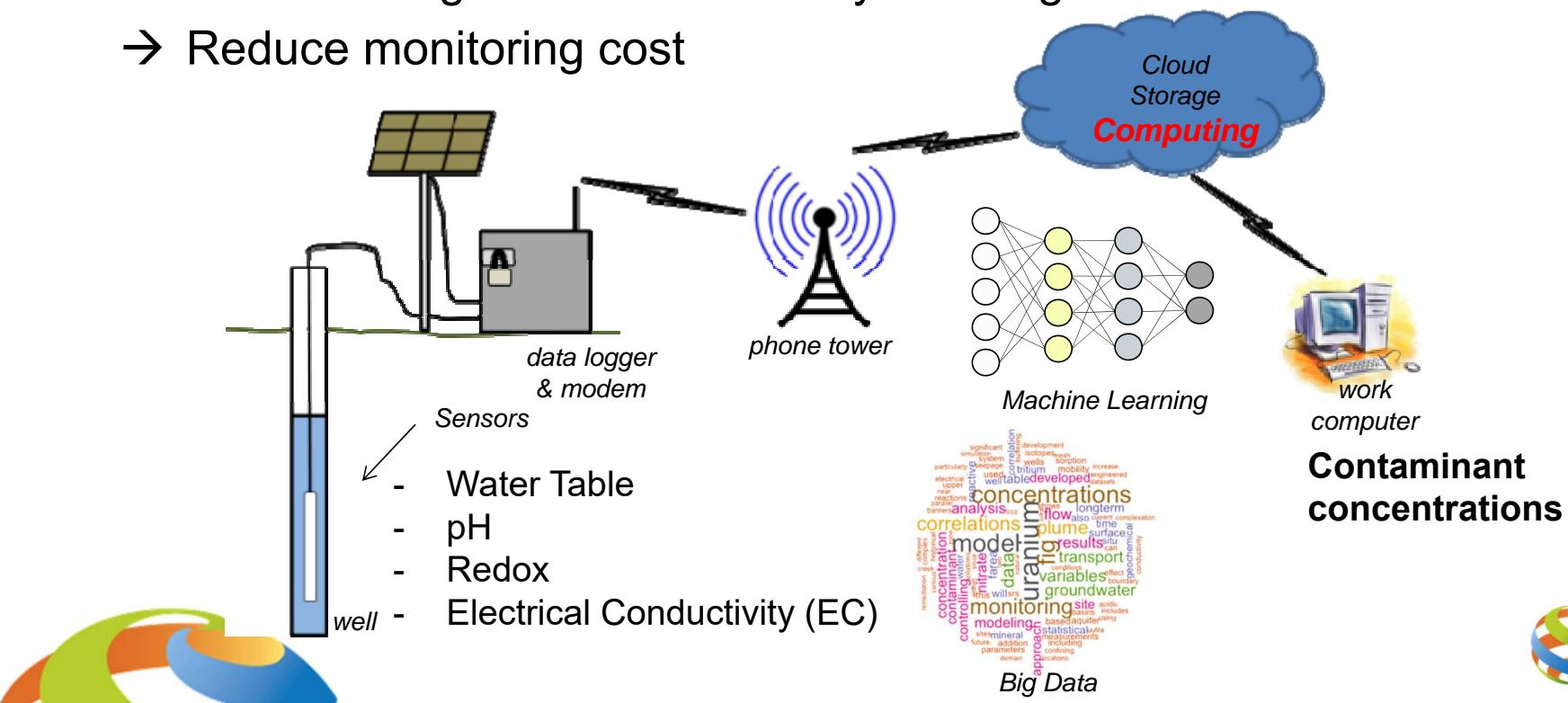


# Residual Contaminants: Groundwater Flow Direction



# New Paradigm of Groundwater Monitoring

- **Low-cost in situ sensors, wireless network, cloud computing**
  - Autonomous continuous monitoring
  - Detect changes real-time = Early Warning
  - Reduce monitoring cost



# Importance of Long-Term Monitoring

The Denver Post

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OPINION   OPINION COLUMNISTS

Activists ignore the science that says Rocky Flats National Wildlife Refuge is safe

By VINCENT CARROLL | The Denver Post  
PUBLISHED: June 16, 2017 at 12:00 pm | UPDATED: June 16, 2017 at 2:36 pm

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14  14



Andy Cross, Denver Post file

Good example: Monitoring data proves that the site is safe to dismiss false claims

- Ensure public safety
- Prepare for liability issues
- Tackle fake news

**Beneficial for both residents and site operators**



# Climate Change Impact: What to expect?



Climate change impact on residual contaminants under sustainable remediation



Arianna Libera<sup>a,\*</sup>, Felipe P.J. de Barros<sup>a</sup>, Boris Faybishenko<sup>b</sup>, Carol Eddy-Dilek<sup>c</sup>, Miles Denham<sup>d</sup>, Konstantin Lipnikov<sup>e</sup>, David Moulton<sup>e</sup>, Barbara Maco<sup>f</sup>, Haruko Wainwright<sup>b</sup>

<sup>a</sup> Sonny Astani Dept. of Civil and Environmental Engineering, University of Southern California, Los Angeles, California, USA

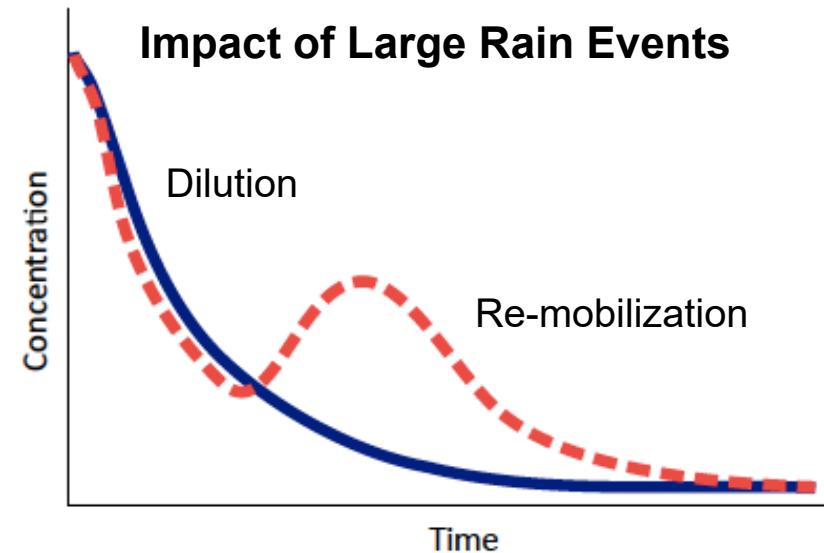
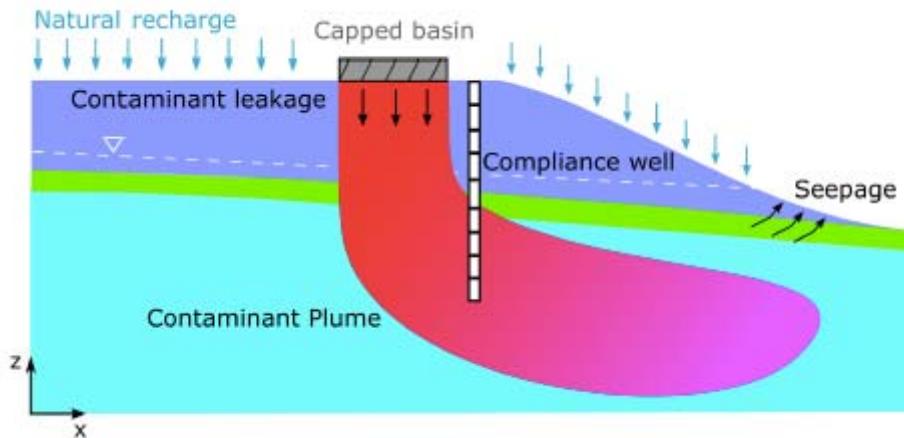
<sup>b</sup> Lawrence Berkeley National Laboratory, Berkeley, CA, USA

<sup>c</sup> Savannah River National Laboratory, Aiken, SC, USA

<sup>d</sup> Panoramic Environmental Consulting, LLC, Aiken, SC, USA

<sup>e</sup> Los Alamos National Laboratory, Los Alamos, NM, USA

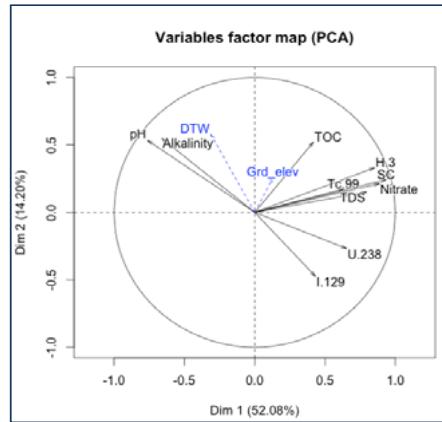
<sup>f</sup> Wactor & Wick LLP Environmental Lawyers, Oakland, CA, USA



## Importance of

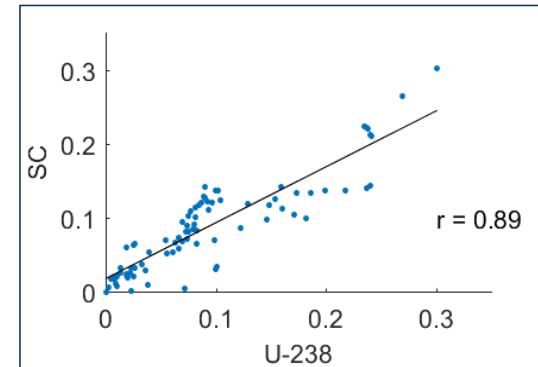
- **Surface capping: Limit infiltration through the source zone**
- **Source-zone monitoring to detect re-mobilization**

# Data Analytics Workflow

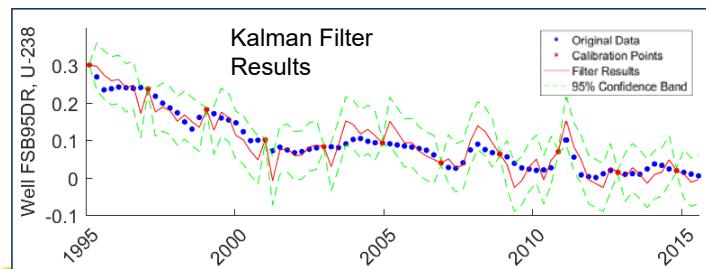


Exploratory Data Analysis

Quantification of Correlations

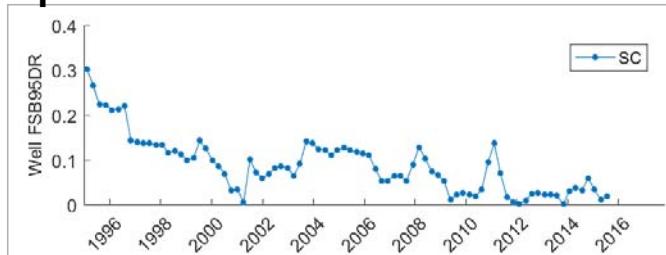


Contaminant Concentration Estimation  
Machine Learning

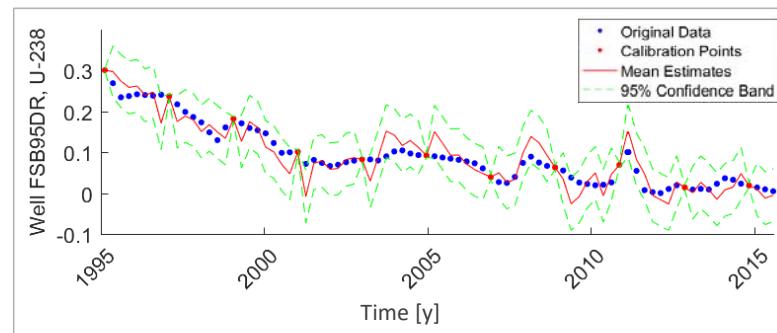
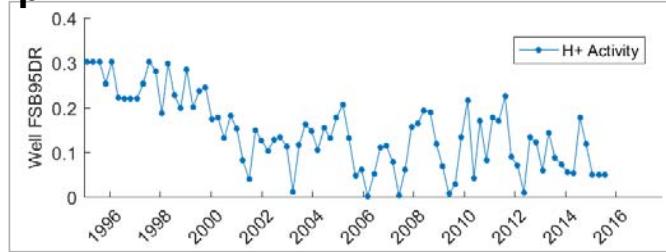


# Kalman Filter: Application at Savannah River Siste

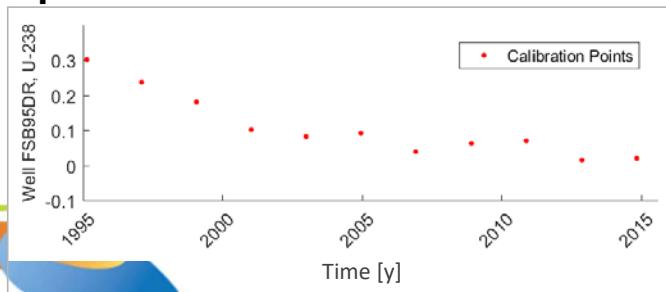
## Specific Conductance



## pH



## Sparse Direct Measurements



- Confidence interval captures validation points
- Mean estimate captures natural fluctuation

Schmidt et al, 2019 EST

# Big Interest in ML x Environment



Environ. Sci. Technol. Environ. Sci. Technol. Lett.

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Article

## In Situ Monitoring of Groundwater Contamination Using the Kalman Filter

Franziska Schmidt<sup>†</sup> , Haruko M. Wainwright<sup>\*‡</sup>, Boris Faybishenko<sup>§</sup>, Miles Denham<sup>¶</sup>, and Carol Eddy-Dilek<sup>⊥</sup>

<sup>†</sup> Department of Nuclear Engineering, University of California Berkeley, Etcheverry Hall, 2521 Hearst Avenue, Berkeley, California 94709, United States

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<sup>¶</sup> Panoramic Environmental Consulting, LLC, P.O. Box 906, Aiken, South Carolina 29802,

<sup>⊥</sup> Savannah River National Laboratory, Savannah River Site, Aiken, South Carolina 2980

Environ. Sci. Technol., 2018, 52 (13), pp 7418–7425

DOI: 10.1021/acs.est.8b00017

Publication Date (Web): June 22, 2018

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Cite this: Environ. Sci.

7425

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Efficiency & Environment

## Scientists develop new method to track groundwater pollutants in real-time

It is expected to reduce the frequency of manual groundwater sampling and lab analysis and therefore cut the monitoring cost

The University Network

## New Algorithm Provides Real-Time Monitoring Of Groundwater Pollutants

Sam Benzeria 8 Months Ago



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PUBLIC RELEASE: 13-AUG-2018

## Algorithm provides early warning system for tracking groundwater contamination

Berkeley Lab researchers devise system to monitor contaminant plumes

DOE/LAWRENCE BERKELEY NATIONAL LABORATORY

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GCN

The Technology that Drives Government IT

AI & Automation Cybersecurity Cloud & Infrastructure Data & Analytics Smart Cities & IoT Emergency

## Machine learning improves contamination monitoring

BY MATT LEONARD | AUG 14, 2018

Because groundwater is [susceptible to pollution](#) from automotive fuel, fertilizer or naturally occurring substances like iron, the Environmental Protection Agency and its state-level counterparts conduct annual or quarterly sampling and analysis.

# Advanced Long-term Monitoring Systems (ALTEMIS)



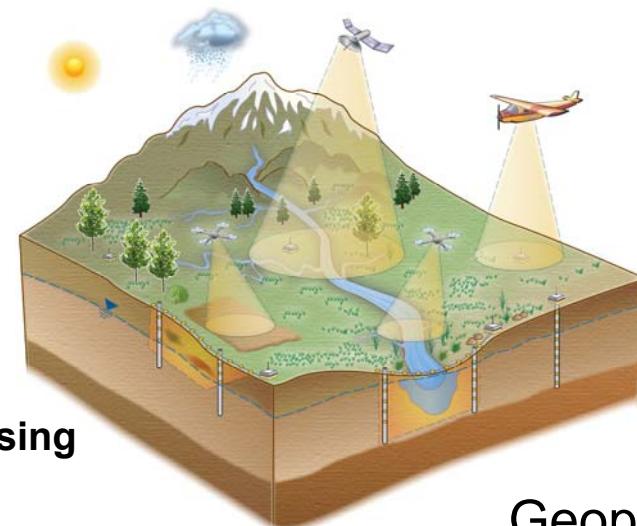
ML/AI

Sensing

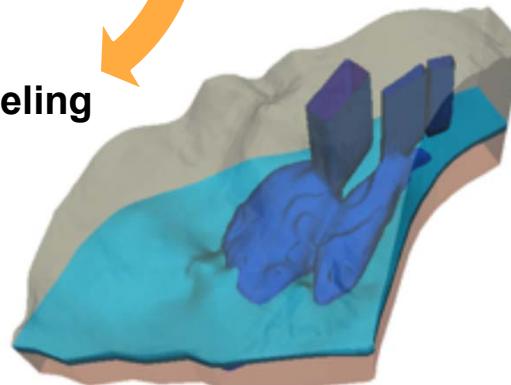
Modeling

R/Python packages

- Historical data analysis
- Well placement optimization



Geophysics  
Fiber optics



Remote sensing  
- Wetland  
- Surface Barrier



# Summary

- **Bioremediation of Uranium or Heavy Metal in general**
  - Control redox conditions to reduce solubility: Microbial stimulation
    - Problem with rebound/remobilization
  - Naturally reduced zone: organic rich layer
    - Persistent plume
  - Permeable reactive barrier: Create stable co-precipitates
    - Successful immobilization for 2+ years
- **Monitoring for Bioremedaiton**
  - Critical to understand the variability of bioremediation effects
    - Groundwater flow direction changes, multiple sources
  - Stability of immobilized contaminants need to be monitored for an extended time
  - New technologies (AI/Sensing/Modeling capabilities) have a great potential to improve the risk quantification at contaminated sites

# Thank You!

## Contact

Haruko Wainwright  
[HMWainwright@LBL.gov](mailto:HMWainwright@LBL.gov)

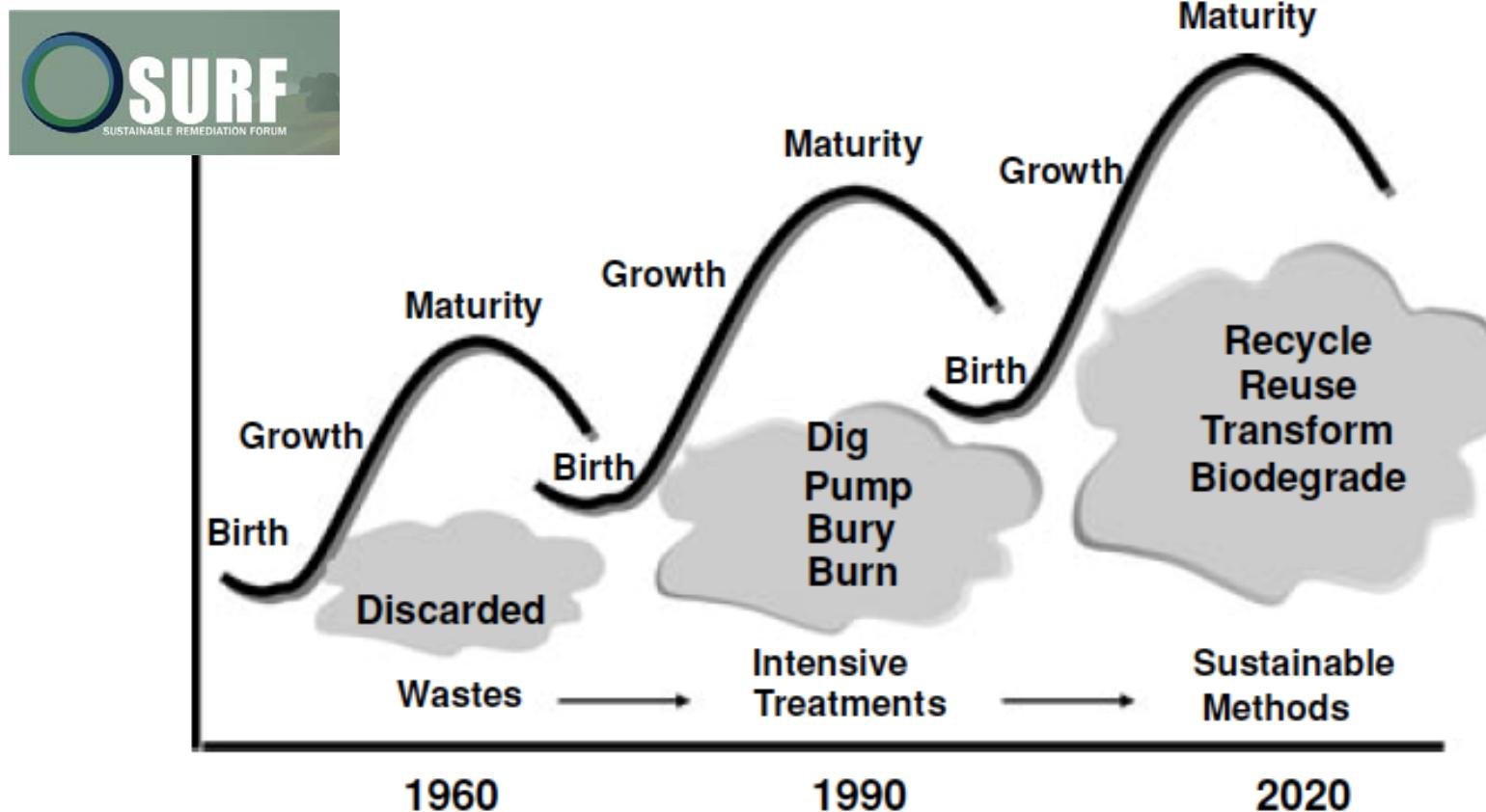
Ken Williams  
[KHWilliams@lbl.gov](mailto:KHWilliams@lbl.gov)

## Acknowledgment

DOE Office of Science  
DOE Office of Environmental Management



# Environmental Remediation: Evolution



Sustainable Remediation Forum (SURF), "Integrating sustainable principles, practices, and metrics into remediation projects", Remediation Journal, 19(3), pp 5 - 114, editors P. Hadley and D. Ellis, Summer 2009

# Sustainable Remediation: Net Environmental Impact

- Reuse/recycle
- Reduce energy, water use and waste
- Passive remediation
- Monitored natural attenuation
- Longer institutional control with alternative/attractive end-use



Former Reilly Tar & Chemical Corporation Plant



Rocky Flats National Wildlife Refuge

