SRNL™
SAVANNAH RIVER NATIONAL LABORATORY
Operated by Savannah River Nuclear Solutions, LLC
We Put Science To Work

CSGSS
CENTER FOR SUSTAINABLE GROUNDWATER & SOIL SOLUTIONS
Managed by Savannah River National Laboratory
PROTECT • PRESERVE • DEVELOP • SUSTAIN

Working with Nature in Remediating Groundwater Contaminated with Metals and Radionuclides

Miles Denham
Savannah River National Laboratory

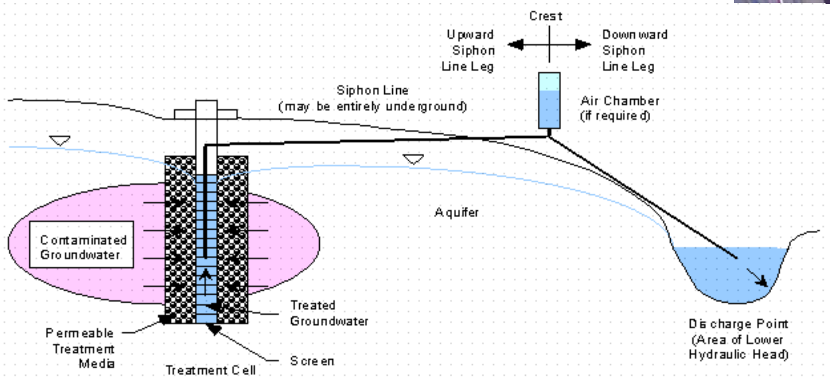
Thanks to: Karen Vangelas, Jay Noonkester, Maggie Millings

Natural Forces Harnessed to Remediate Organics

Solar -- Microblower™

Barometric Changes BaroBall™

Gravity -- GeoSiphon™



SRNL

Metals and Radionuclides Pose Different Challenges

Removal from system requires "muck-and-truck" or "pump-and-treat"

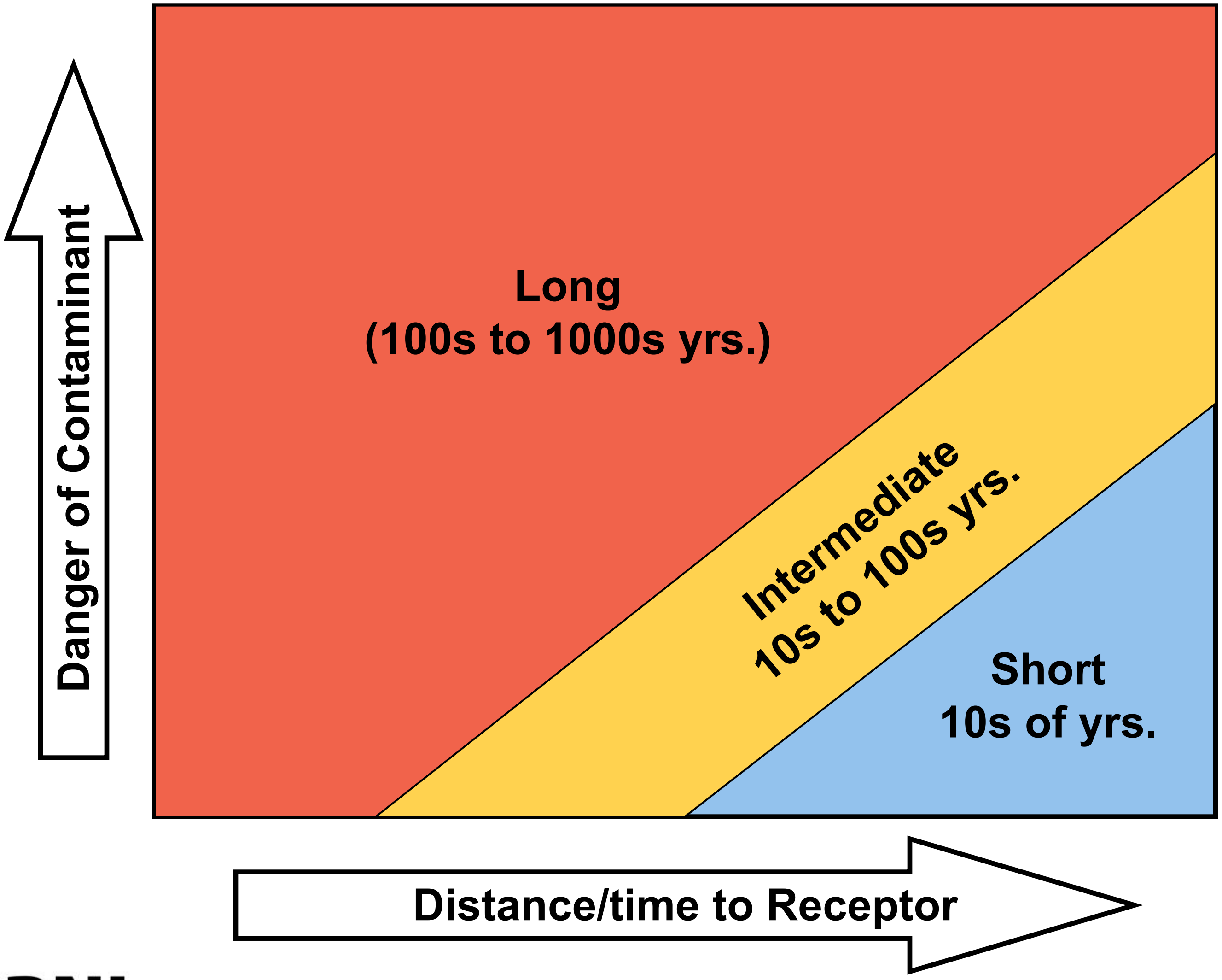
- **Both are inefficient and can create further environmental damage**
- **Exceptions are relatively short lived radionuclides and the rare sites where Hg and I-129 can be removed by vapor extraction**

Generally, most effective option is *in situ* immobilization by adsorption or precipitation

- **Both processes sensitive to geochemical conditions**
- **Must remain effective for long time periods**

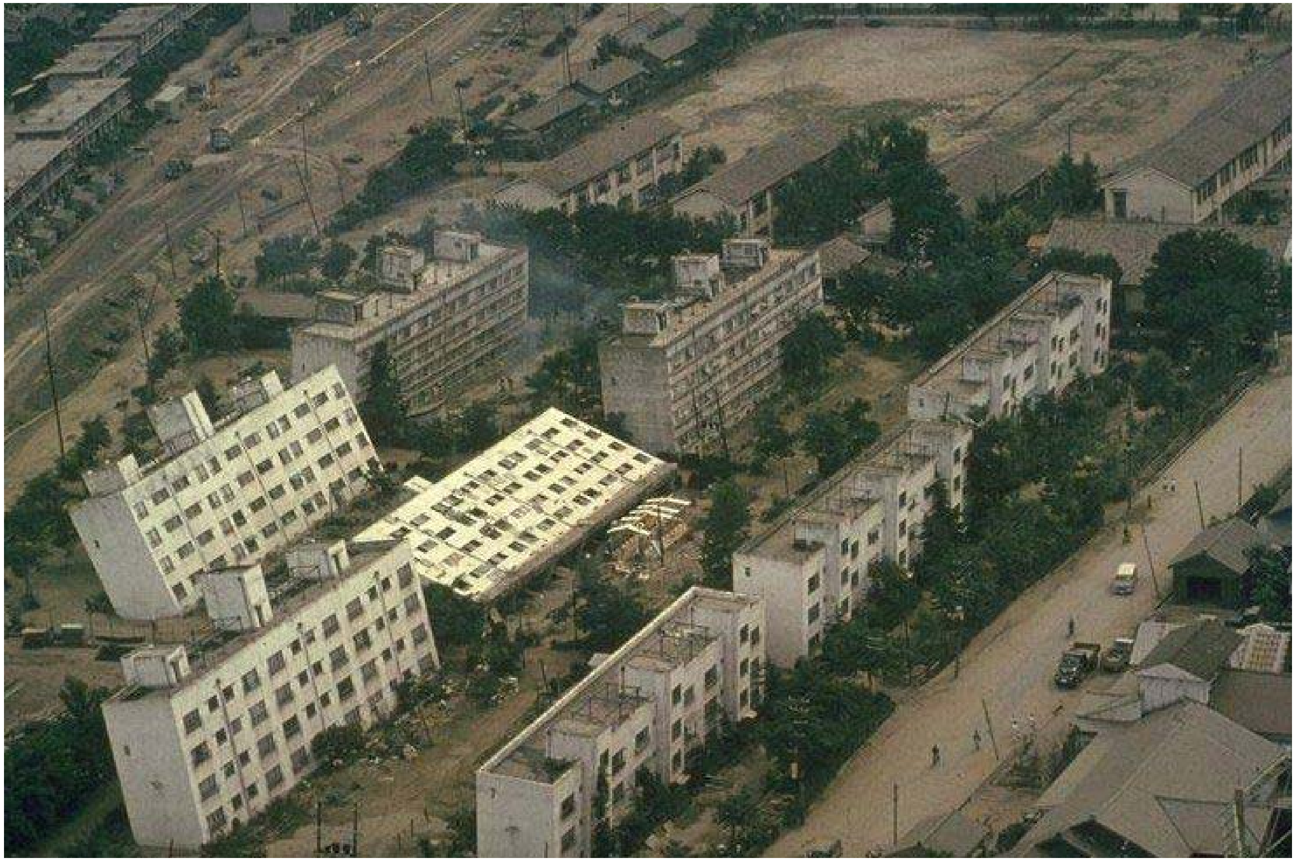
SRNL™

Sustainability Timeframes



SRNL

You Can't Fight Mother Nature



1964 Earthquake – Niigata, Japan

SRNL™

Working with Nature Requires a Holistic View

Must recognize all parts of the system and how they interact

- **Nature of the contaminant source**
- **Hydrology**
- **Geochemistry**
- **Microbiology**

Must understand how they change with time

- **Evolution of the waste site**

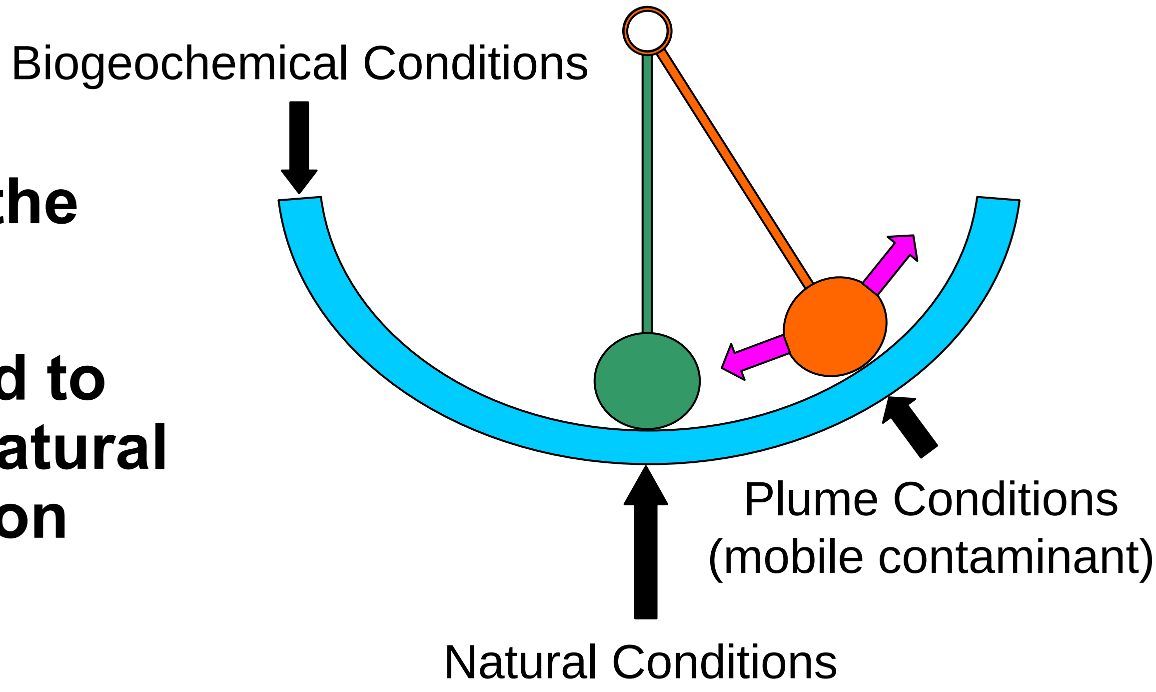
SRNL™

Waste Site Evolution

Contaminant plume is a perturbation of natural conditions

Remediation is a perturbation of the perturbation

Waste site will tend to evolve toward natural pre-contamination conditions



SRNL

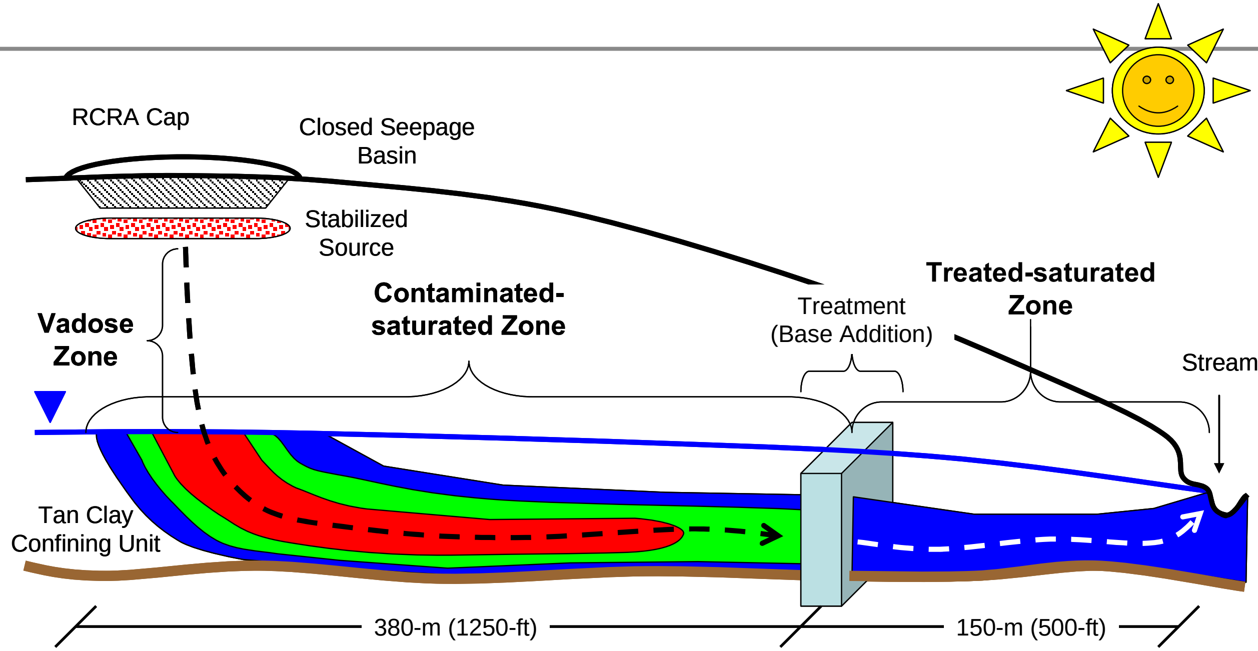
Working with Nature

1. **Develop conceptual model that includes geochemical evolution of site**
2. **Use EPA guidance on MNA of metals and rads and ITRC team Tech Reg guidance (coming soon)**
3. **If you reach the "Evaluate enhancement options" box on the ITRC Framework Diagram**

- **Evaluate whether attenuation of contaminants would be significantly greater at natural conditions of the aquifer**
- **If so, are there ways to accelerate evolution of waste site?**
- **If not, are there enhancements that are consistent with the natural evolution of the waste site?**
  - Enhancements that minimally alter the evolution and are effective throughout the evolution

SRNL™

F-Area Seepage Basins Plume



- F-Area was a processing facility that extracted Pu from irradiated depleted uranium targets
- Low-level radioactive process solutions were discharged to a series of seepage basins
- Groundwater now acidic (pH~3.2), contains Sr-90, uranium isotopes, I-129, Tc-99, tritium, nitrate

SRNL™

Original Treatment

Pump-Treat-Reinject

- **Designed to capture tritium in a loop until it decayed**
- **Reinjection required removal of all metals and radionuclides to below standards**
- **Water treatment system**
  - Chemical precipitation, reverse osmosis, ion exchange
- **Cost $1 million per month to operate**
- **Created 5000 ft^3 per year of radioactive solid waste**
  - High I-129
- **High environmental and monetary costs**

SRNL™

Development of New In Situ Treatment

Treat only the contaminants of regulatory concern

- **Sr-90, I-129, uranium isotopes, tritium**

Natural evolution of waste site will be toward higher pH (natural pH~5.5)

Acceleration of evolution possible by injection of base

- **Will cause stronger attenuation of Sr-90 and U**

Result: Hybrid funnel-and-gate system – base solutions injected in gates to neutralize acid

- **High alkalinity (buffering capacity) fluid with pH≤10 needed**
  - Sodium hydroxide-sodium carbonate solution

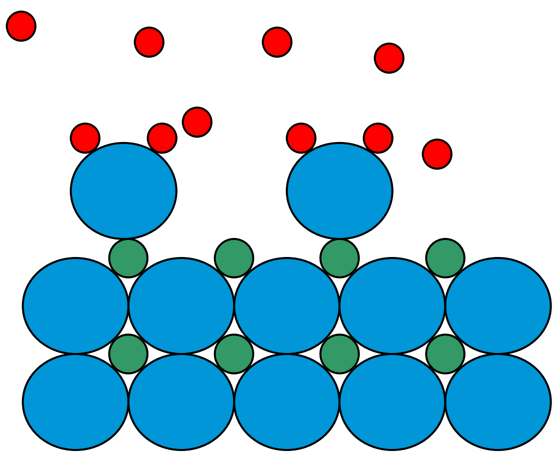
SRNL™

Why High Alkalinity Base?

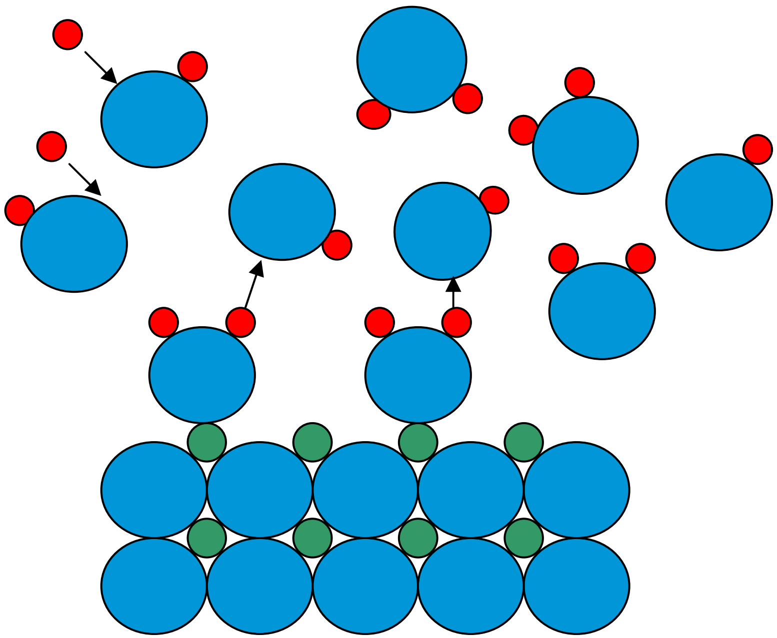
Neutralizing acid in pore water and on mineral surfaces

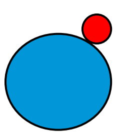
Acid System

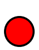
Mineral Surface



Injection of Base



 Hydroxyl ion

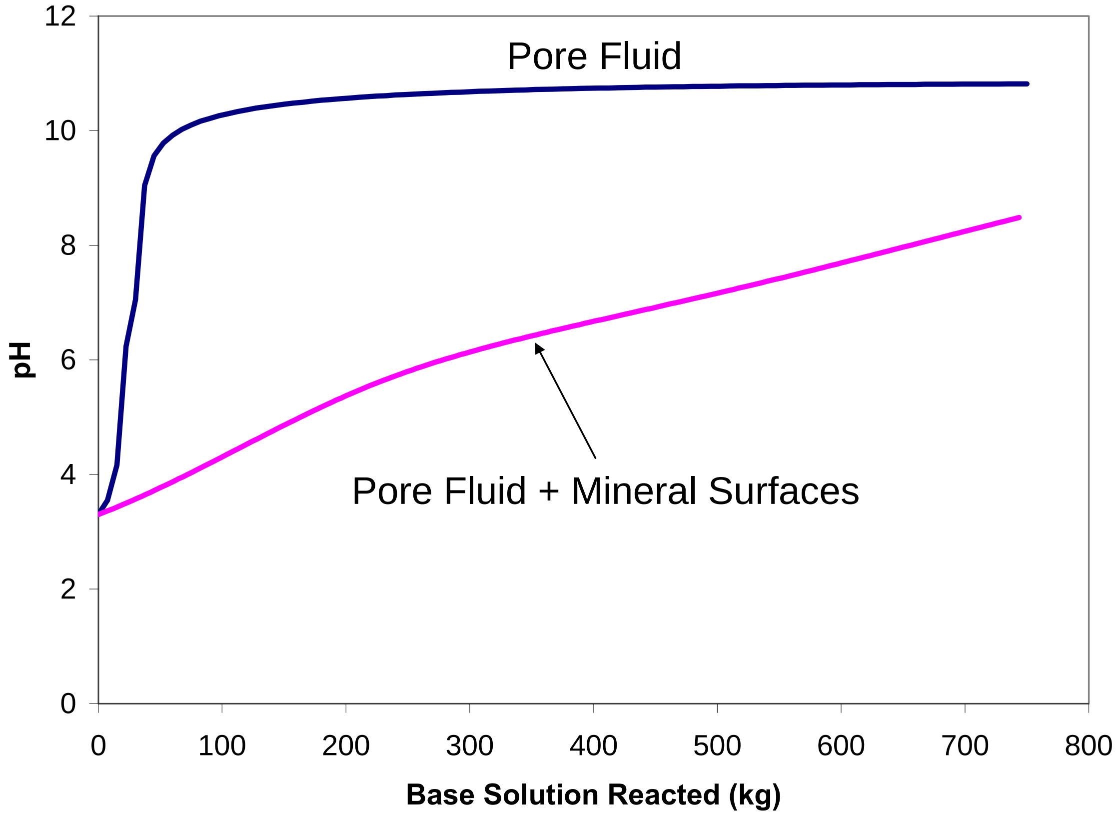
 Hydrogen ion

$$S\text{-}OH_2 + OH^- = S\text{-}OH^- + H_2O$$

$$S\text{-}OH^- + OH^- = S\text{-}O^{-2}$$

SRNL

Effect of Mineral Surface Acidity

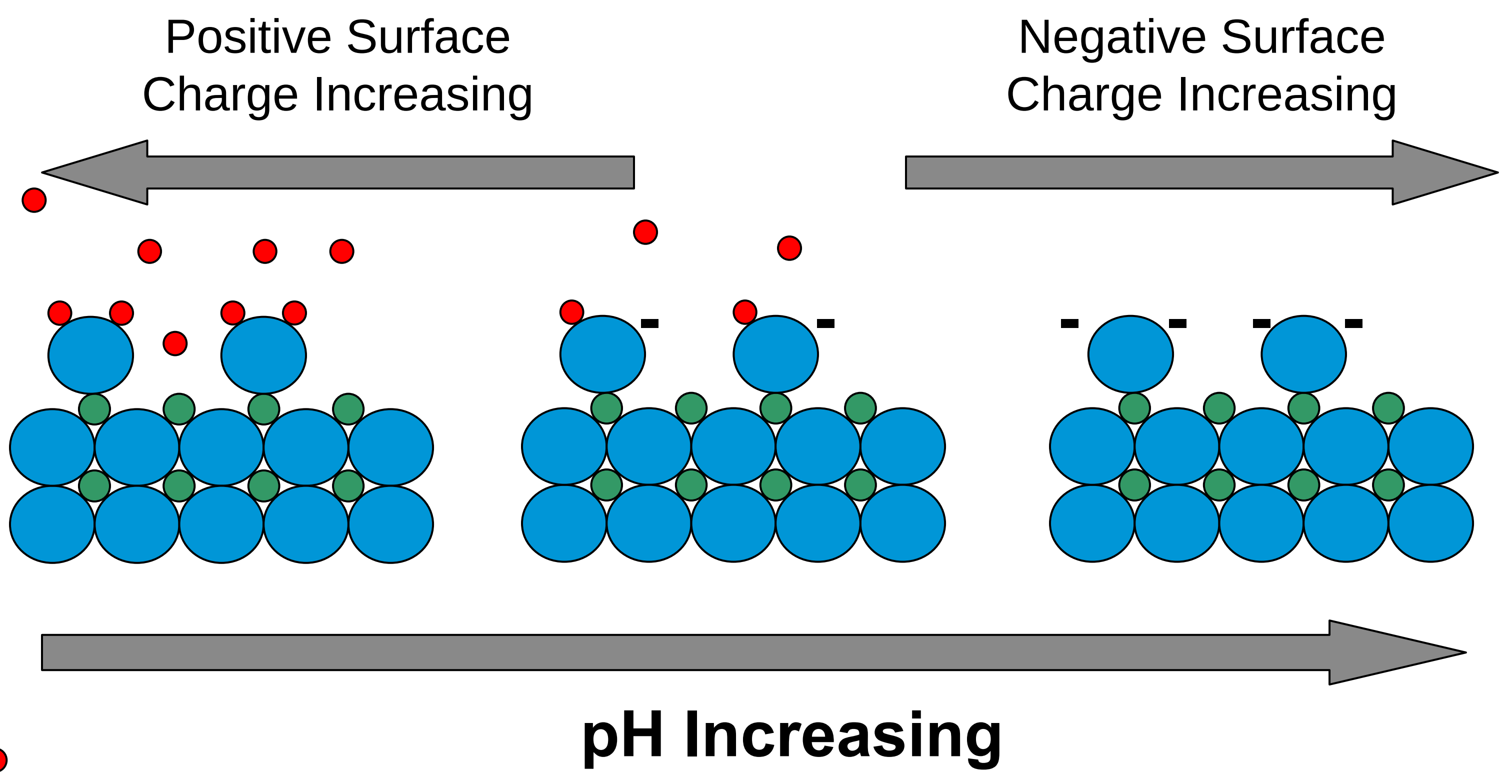


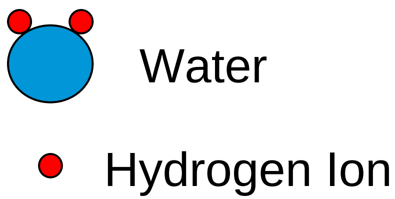
Simulation Parameters

- 1 m^3 sediment
- 25% porosity
- Surface active minerals
  - Kaolinite
  - Goethite

SRNL

Adsorption and Surface Charge

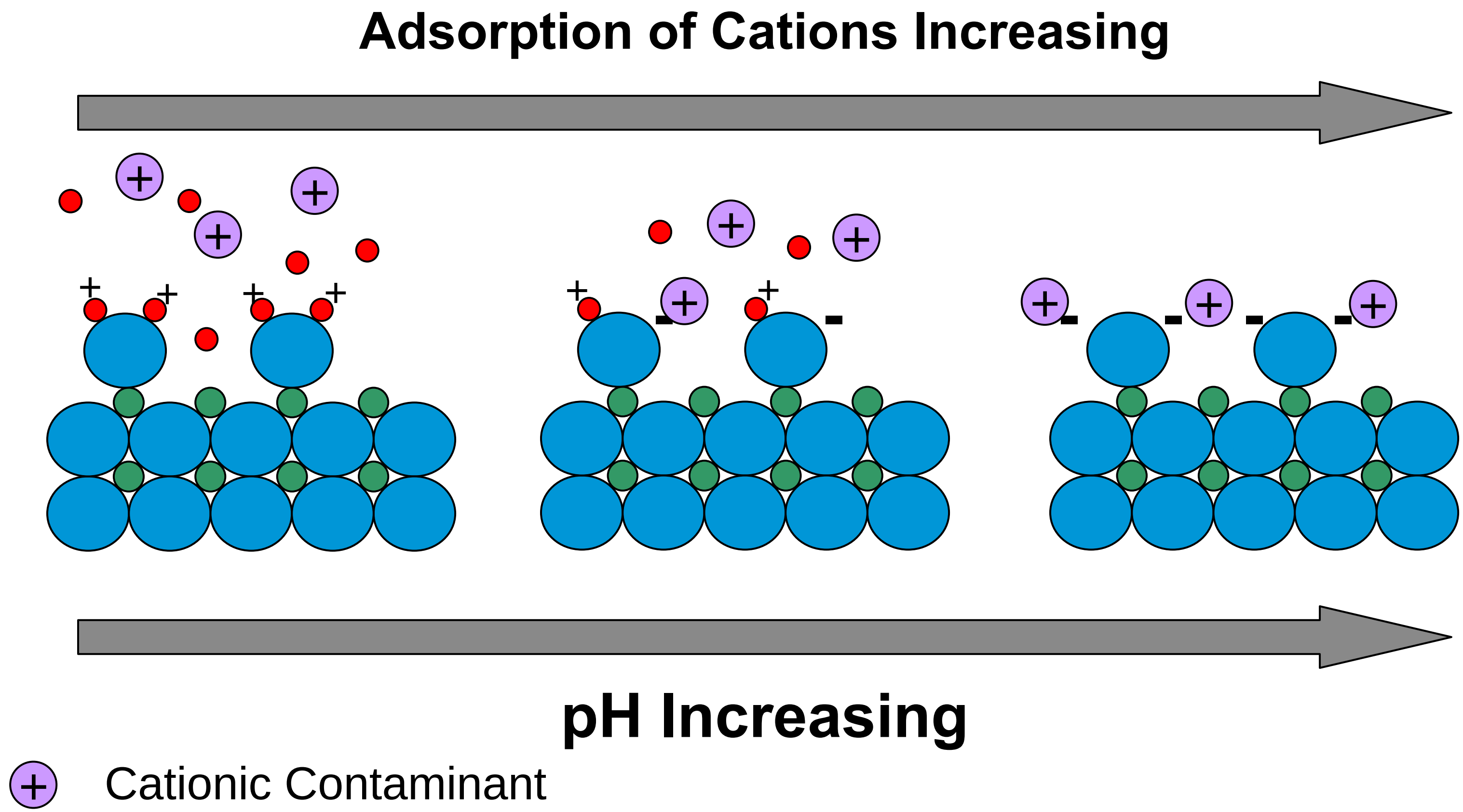




$$S\text{-}O^{-2} + H^{+} = S\text{-}OH^{-}$$

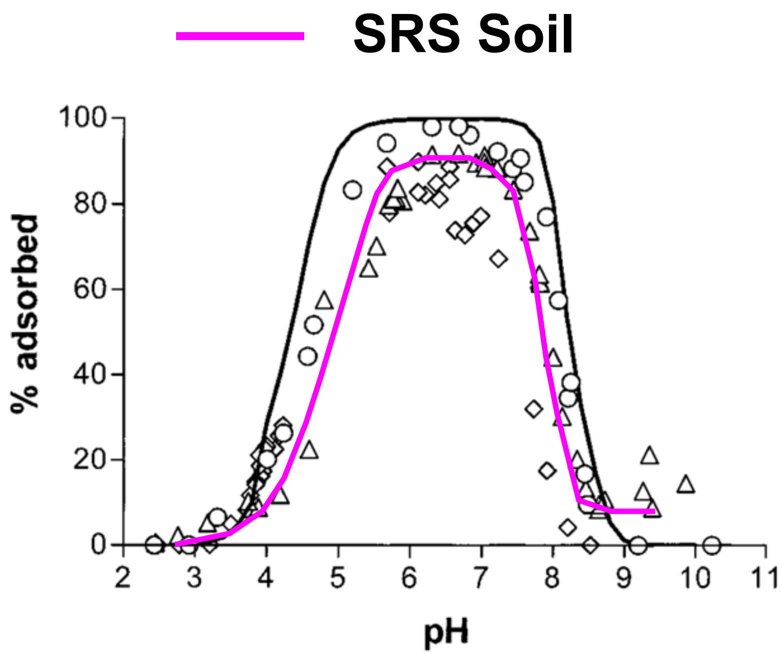
$$S\text{-}OH^{-} + H^{+} = S\text{-}OH_{2}$$

Adsorption of Cationic Contaminants



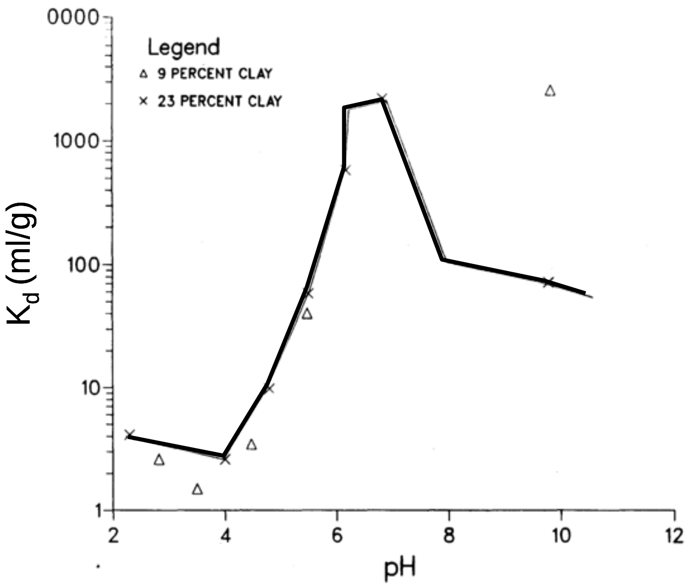
SRNL

Adsorption of Sr-90 and U on SRS Soils



(Barnett et al., 2002)

Uranium Adsorption



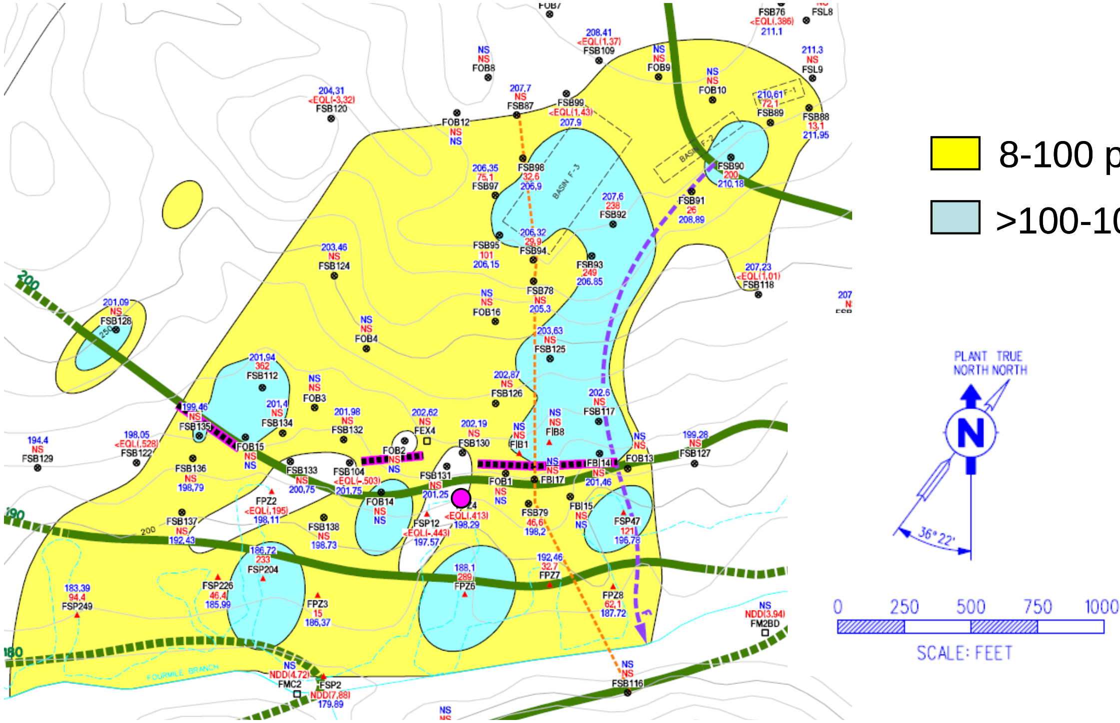
Hoeffner, (1984)

Sr-90 Adsorption on SRS Soil

SRNL

Hybrid Funnel-and-Gate / Base Injection System

Sr-90 Plume



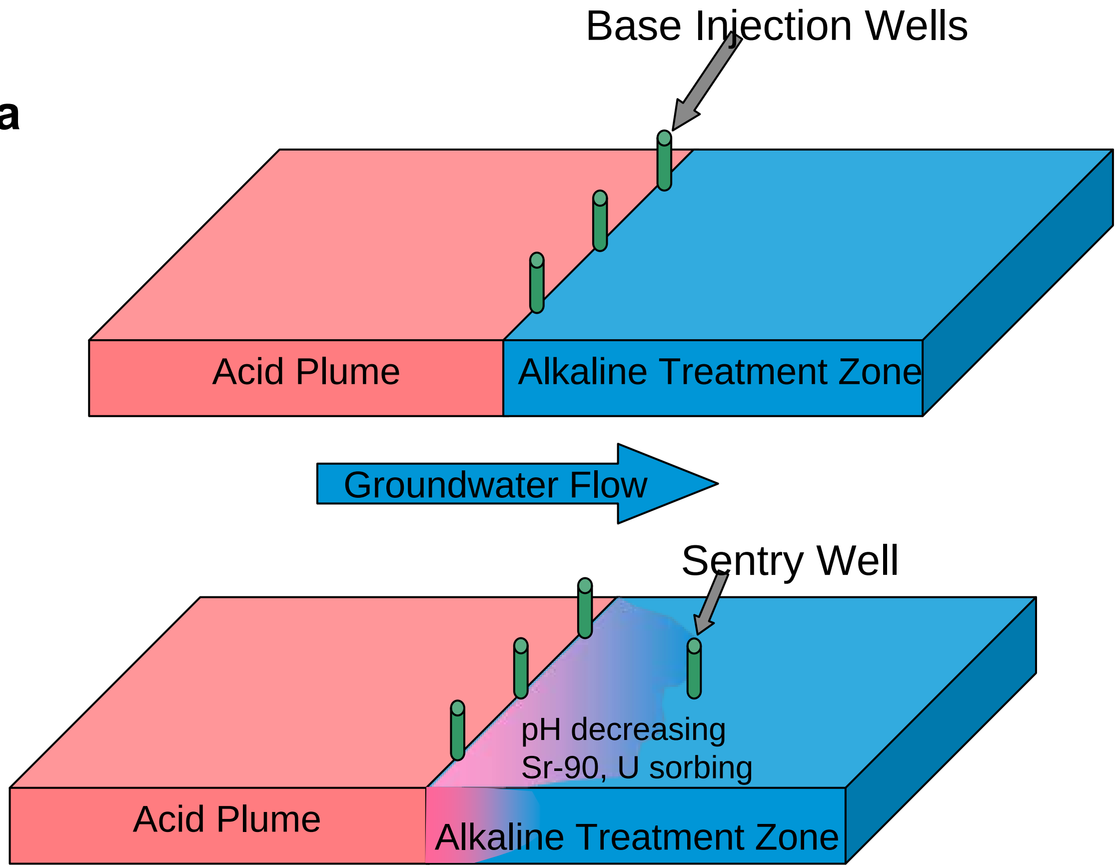
8-100 pCi/L

>100-1000 pCi/L

SRNL

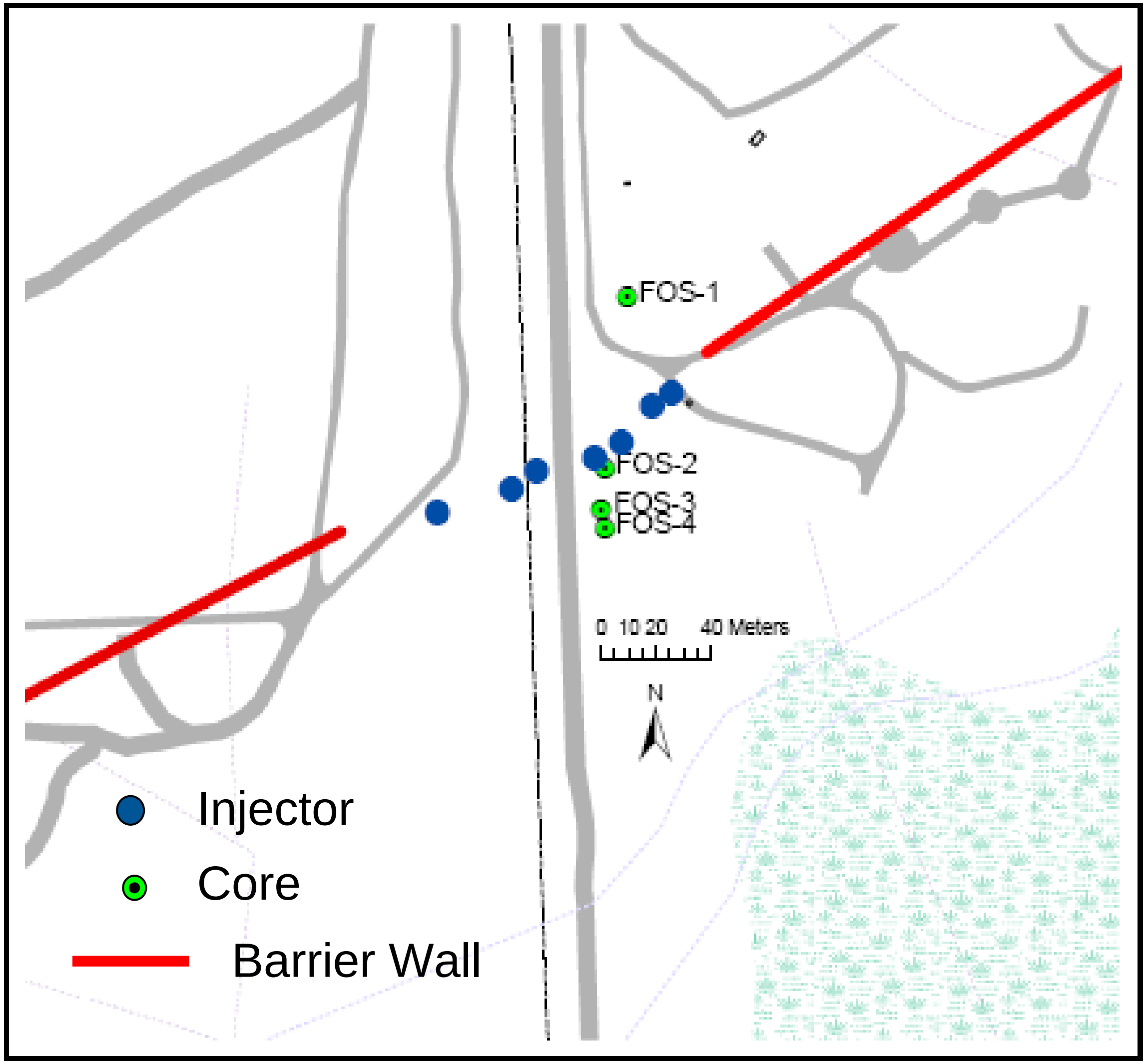
Base Injection to Raise pH

- **Periodic injections of alkaline solution forms a treatment zone**
- **Between injection periods, pH of acidic water migrating into treatment zone is neutralized**
- **Sr-90 and U sorb to aquifer minerals**
- **Injection resumes when sentry well pH is 5.5**



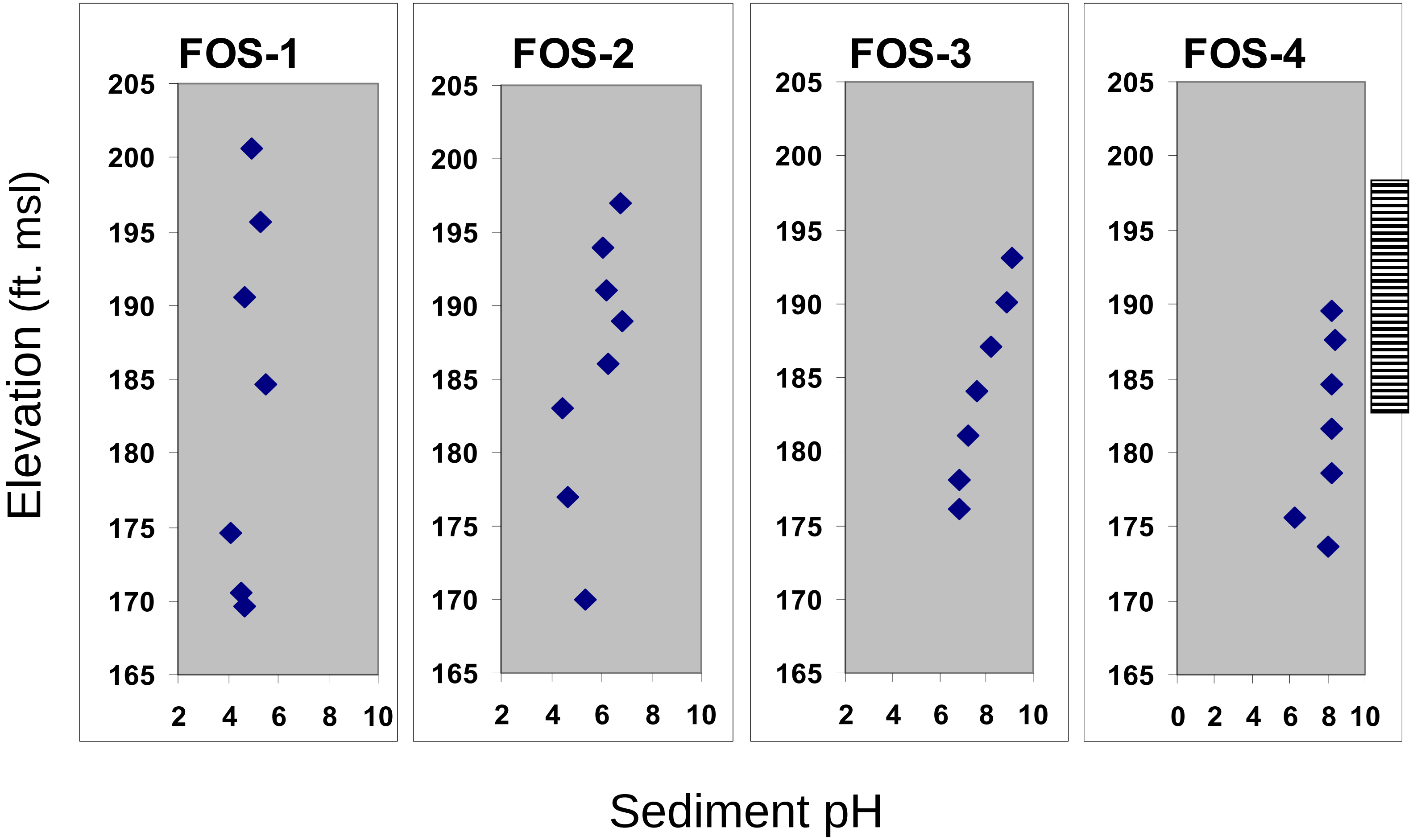
SRNL™

F-Area Base Injection Map



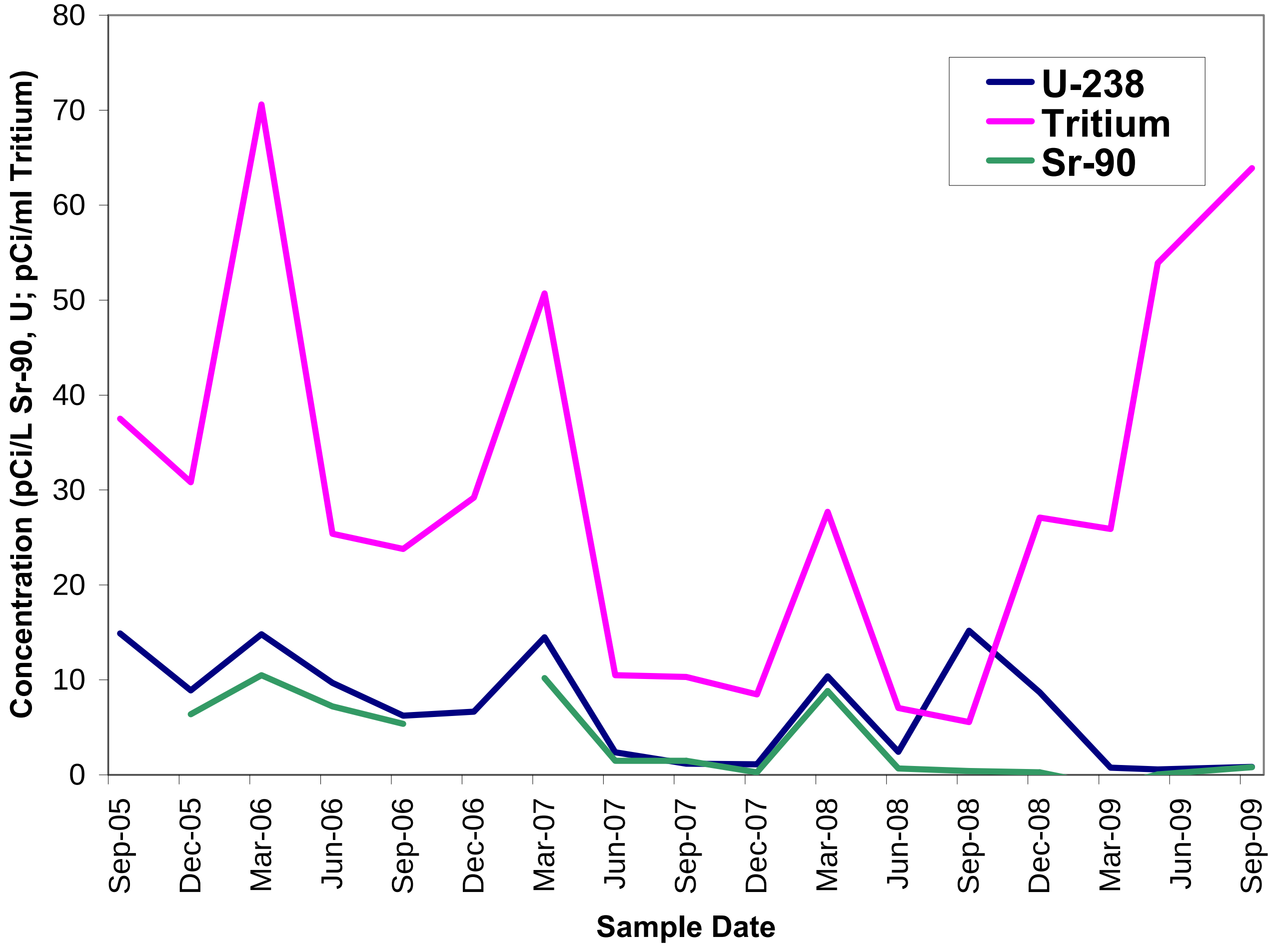
SRNL

pH of Aquifer Sediments



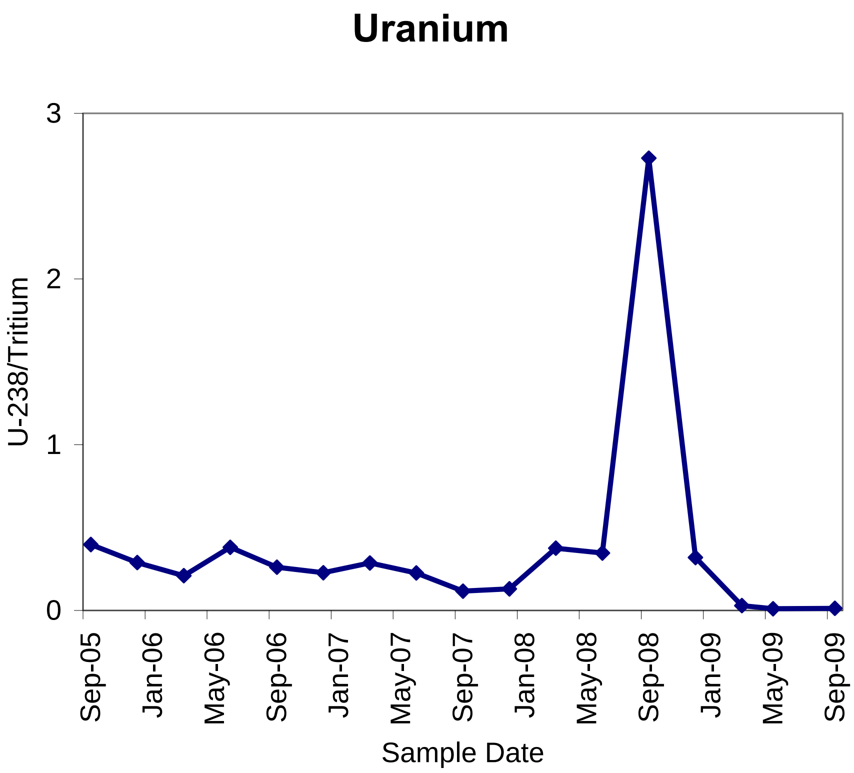
SRNL

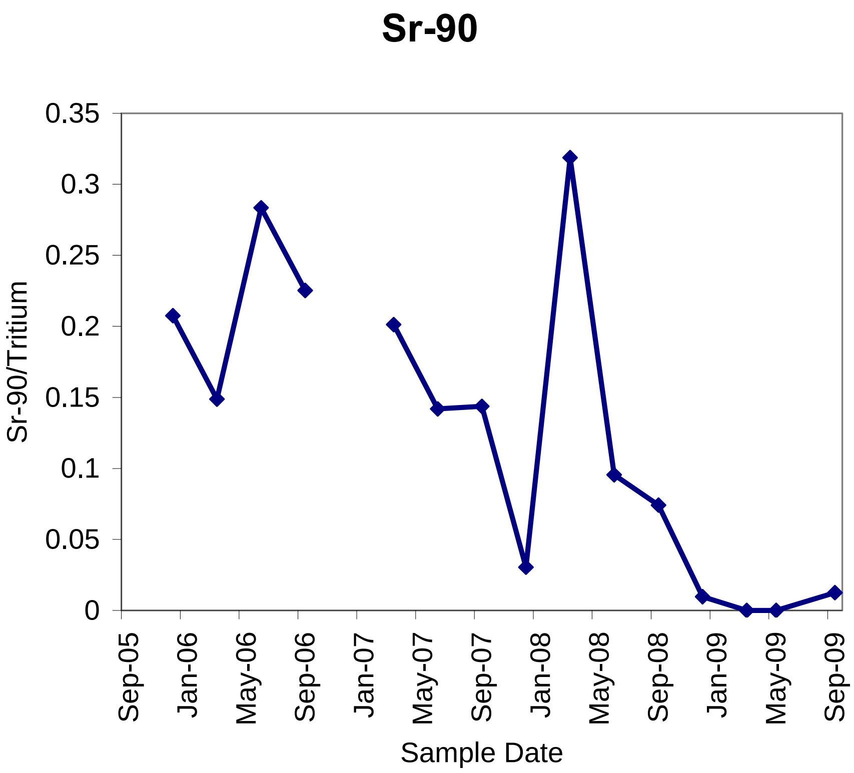
Overall Effect of System



SRNL™

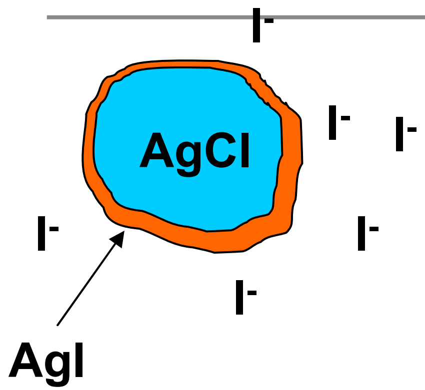
Effect of Base on Adsorption





SRNL

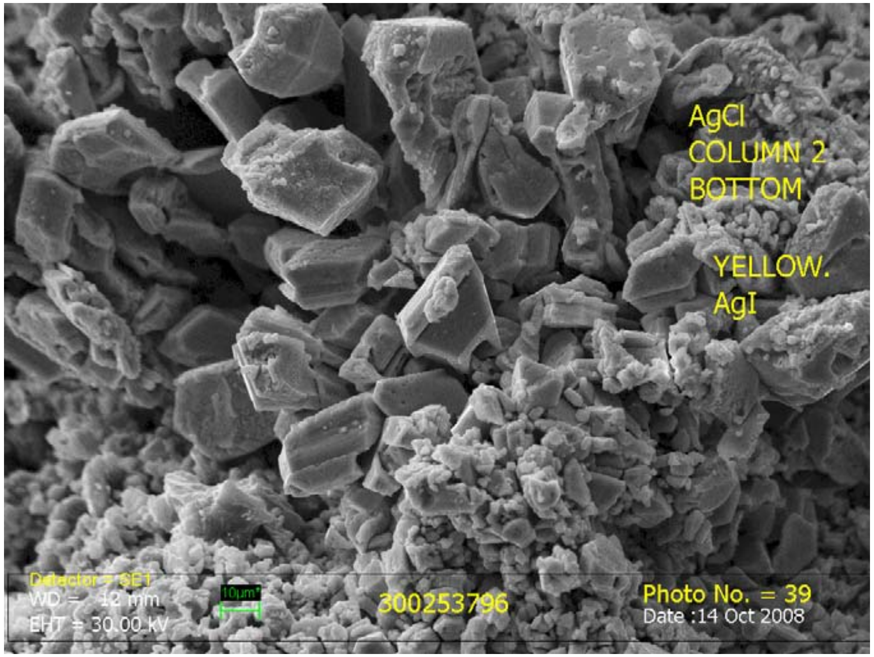
Working on Injectable Amendment for I-129



$AgCl = Ag^+ + I^-$ log K = -9.7

$AgI = Ag^+ + I^-$ log K = -16.1

- **Column studies show 0.05 wt.% treats >50 pore volumes of 8 mg/L I^-**
  - **Groundwater contains <5 ug/L I^-**
- **Field test is ongoing**



SRNL

Summary

Working with nature to remediate metals and radionuclides is generally more efficient and effective than working against nature

Conceptual model must include understanding of all parts of the system and their interactions

Conceptual model must consider geochemical evolution of site

These concepts used at SRS to replace a costly pump-and-treat system with a system that works with nature

- It is inexpensive, effective, and sustainable