Transitioning from Active to Sustainable Remediation for Metals and Radionuclides

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Sustainable Remediation

- Remediation that meets performance objectives for the long-term benefit of the public while minimizing maintenance, cost, and collateral environmental damage.
  - Particularly difficult for metal and radionuclide contamination because of time-frames involved

What is Long-Term?

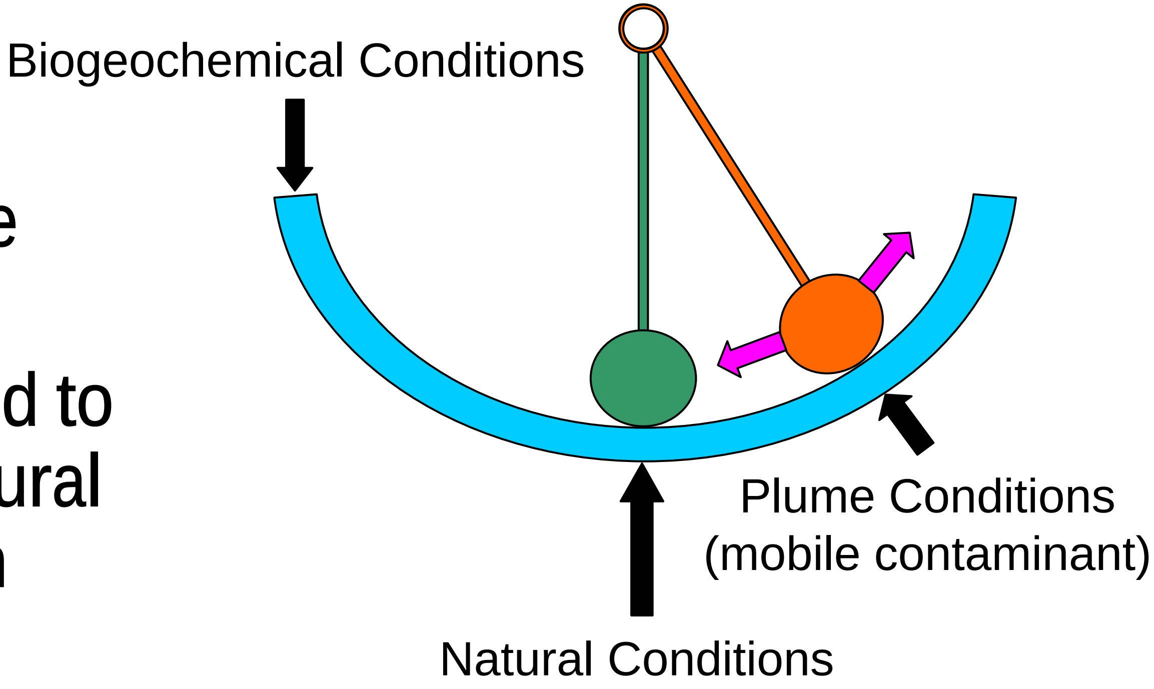
- Depends on site – perhaps centuries to millennia
- Examples:
  - SRS nuclear waste disposal facilities must be designed to meet performance objectives for 10,000 years
  - Yucca Mountain must meet safety requirements for 1,000,000 years
- Waste site owner must demonstrate high probability that remediation will meet agreed upon long-term requirements

Key Question in Evaluating Sustainability

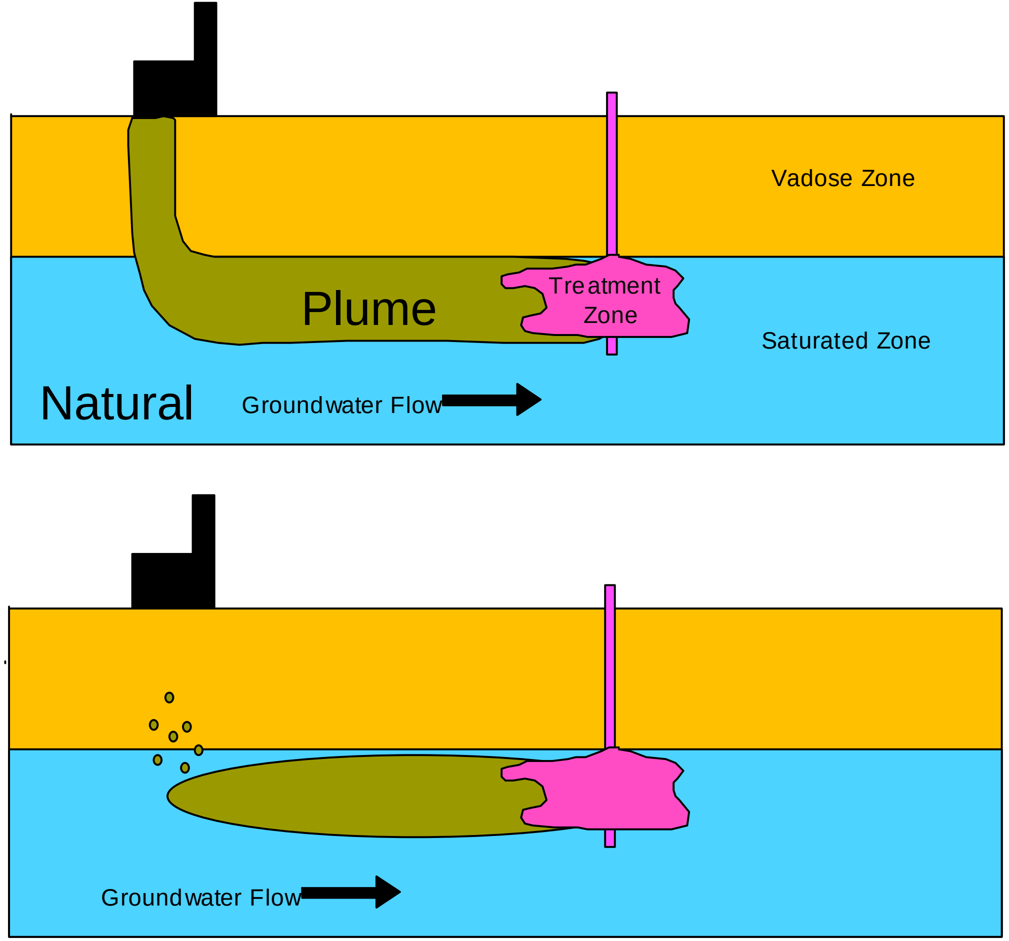
- How long must active remediation proceed before natural processes can be relied upon to meet long-term remediation goals?
  - How long does it take for a site to return to near-natural conditions after active remediation ceases?
    - Overall biogeochemical evolution of a waste site
    - Focus of new work at SRNL
  - How does this affect contaminant flux to compliance point?
    - Contaminant chemistry/specific attenuation mechanisms
    - Focus of much past and current research

Natural Tendencies

- 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



Post In Situ Treatment Considerations

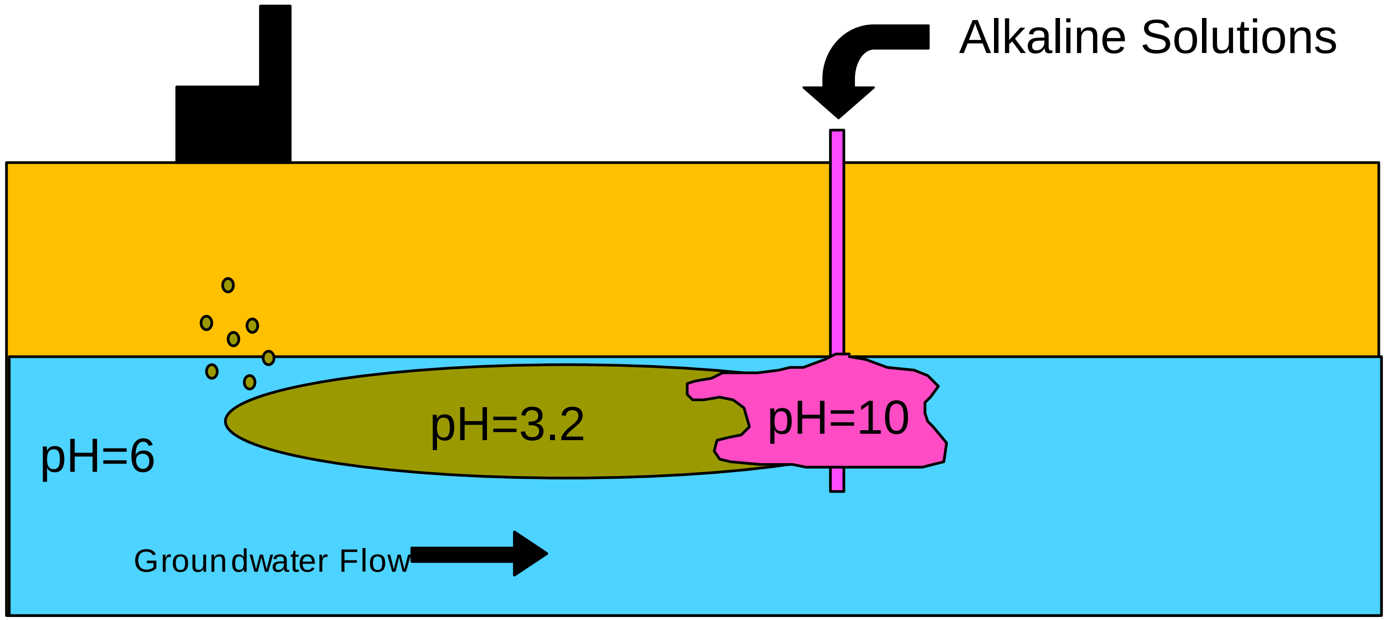


Flux of contaminant out of treatment zone must remain at acceptable levels for the long-term

Controls on Biogeochemical Evolution of Waste Site

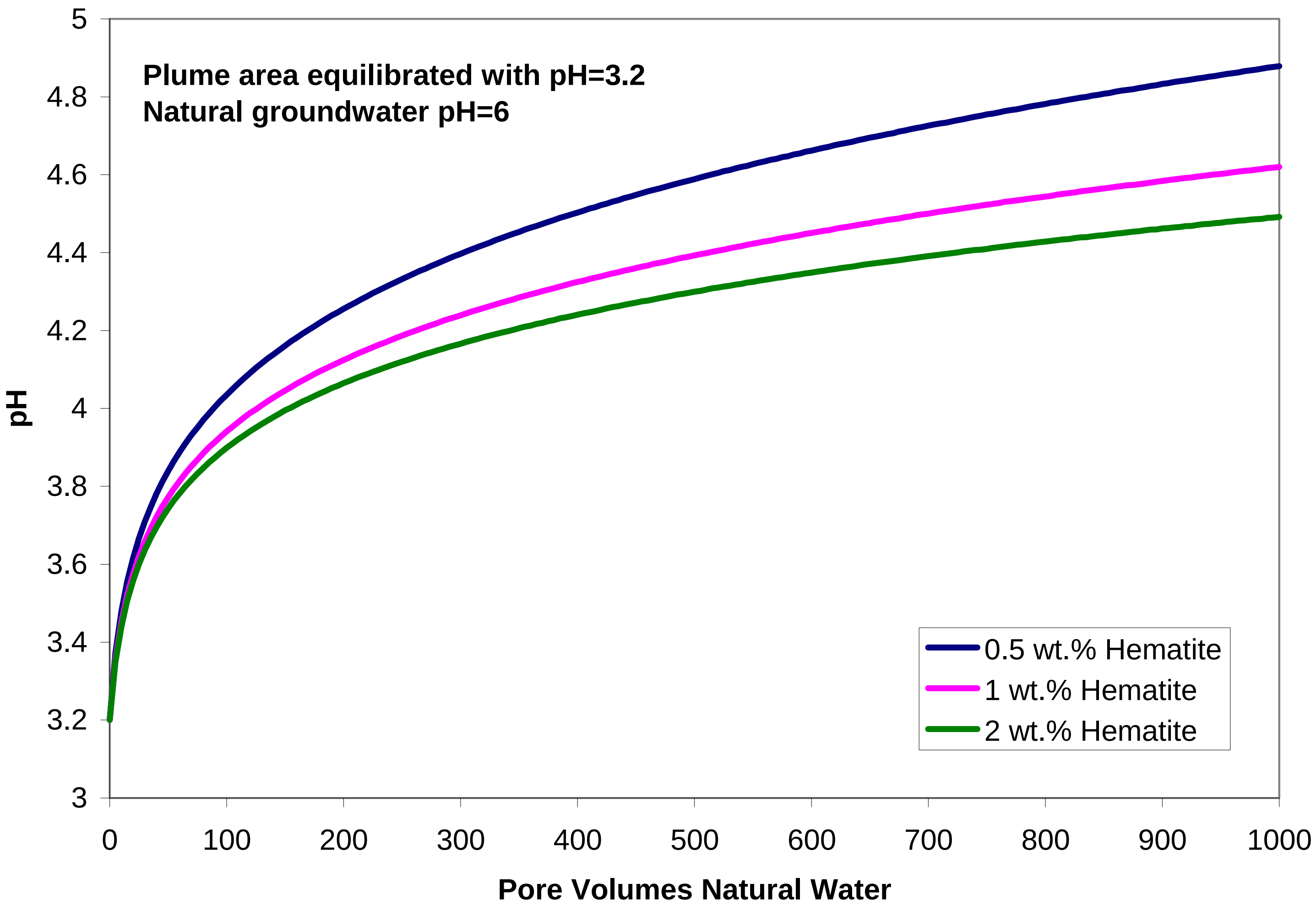
- Hydrogeology
- Biogeochemical conditions of plume
- Biogeochemical conditions of treatment zone
  - Including duration of active treatment
- Biogeochemical conditions of uncontaminated, untreated groundwater (natural groundwater)
- Mineralogy of natural aquifer, plume zone, and treatment zone
- Distribution of reactive minerals

Acid Plume/Alkaline Treatment

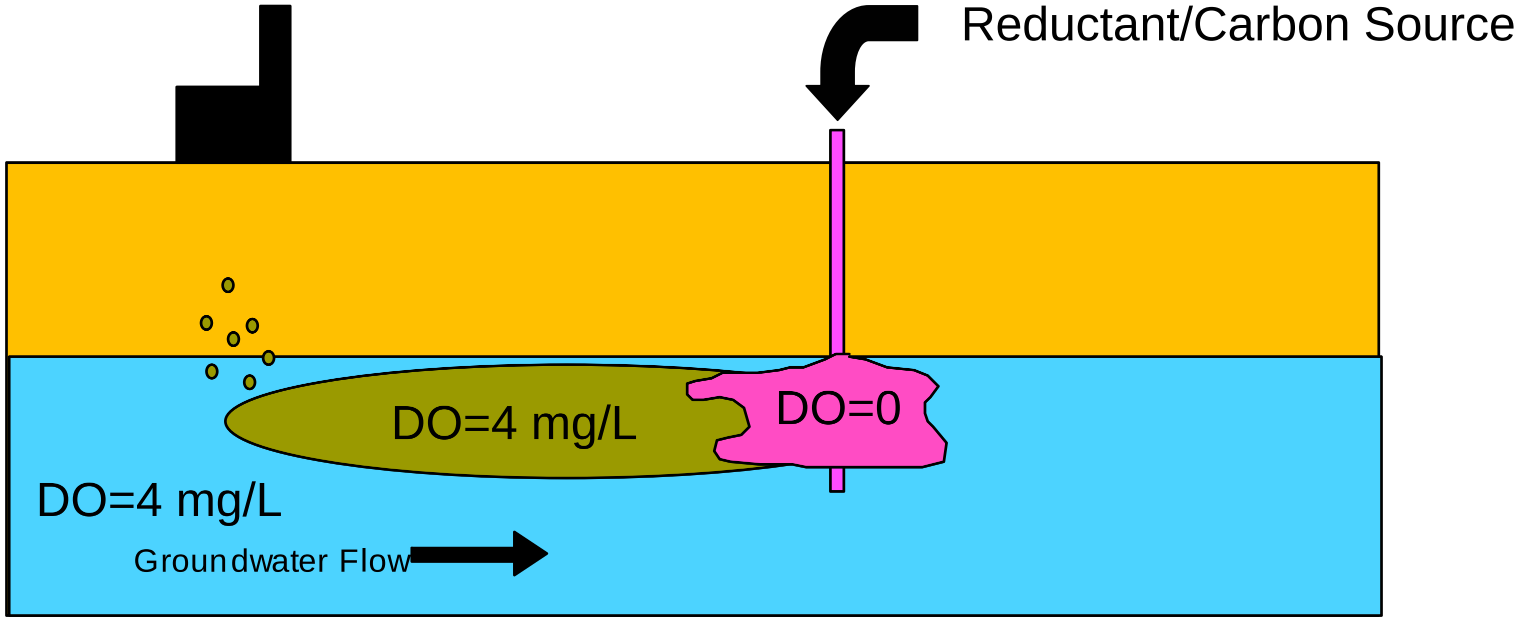


Goal: Increase pH to stop acid sensitive contaminants

pH Rebound of Untreated Plume Area



Treatment By Microbial or Abiotic Induced Reduction



Goal: Reduce redox potential to stop redox sensitive contaminants

Fate of Iron Minerals

- Hematite Reduction

  Hematite + $6H^+ + 2e^- = 2Fe^{+2} + 3H_2O$

  Hematite + $0.67H^+ + 0.67e^-$ = 0.67Magnetite + $0.33H_2O$

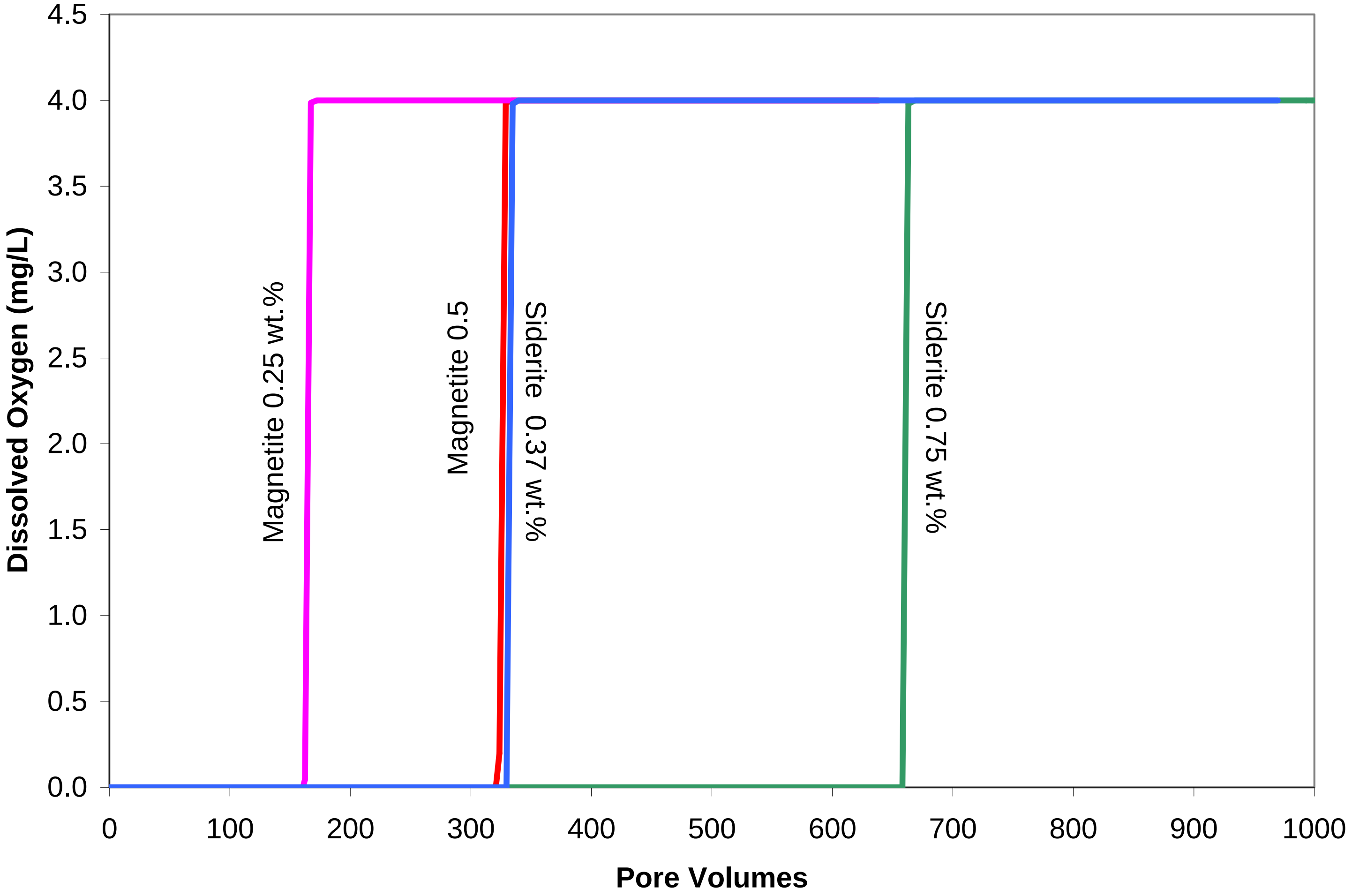
  Hematite + $4H^+ + 2HCO_3^- + 2e^-$ = 2Siderite + $3H_2O$

  Hematite + $4SO_4^{-2} + 38H^+ + 30e^-$ = 2Pyrite + $19H_2O$

- Order of O_2 buffer capacity

  Pyrite > Siderite > Magnetite > Fe^{+2}

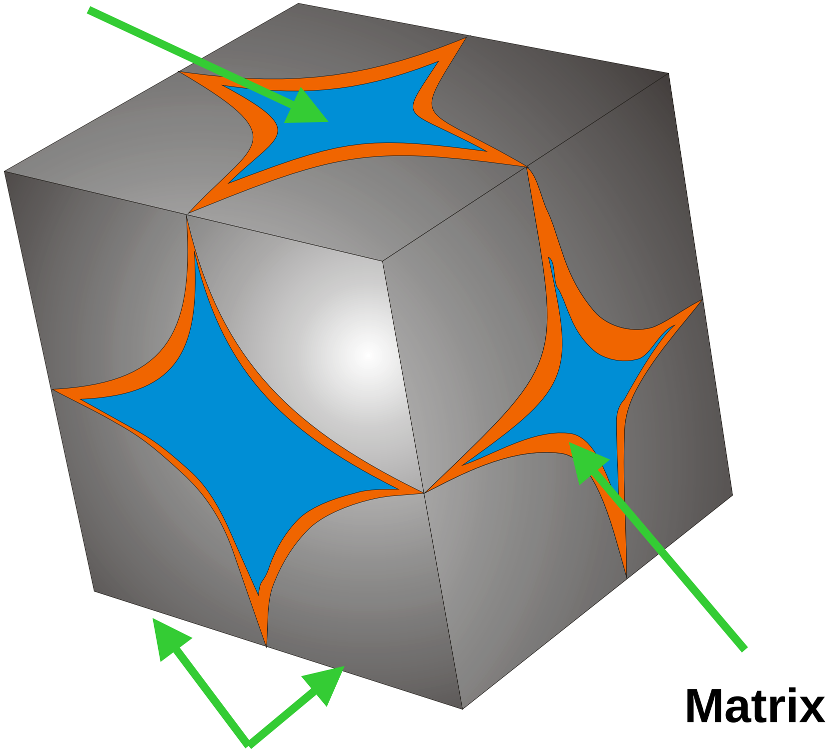
O_2 Rebound After Reductive Treatment



Evenly Distributed Matrix

Greater reactive surface area

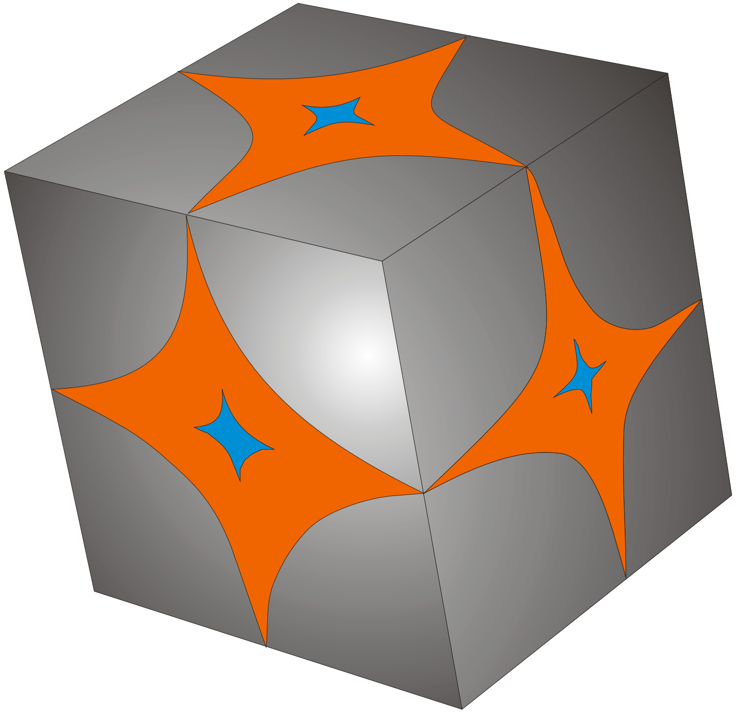
Pore



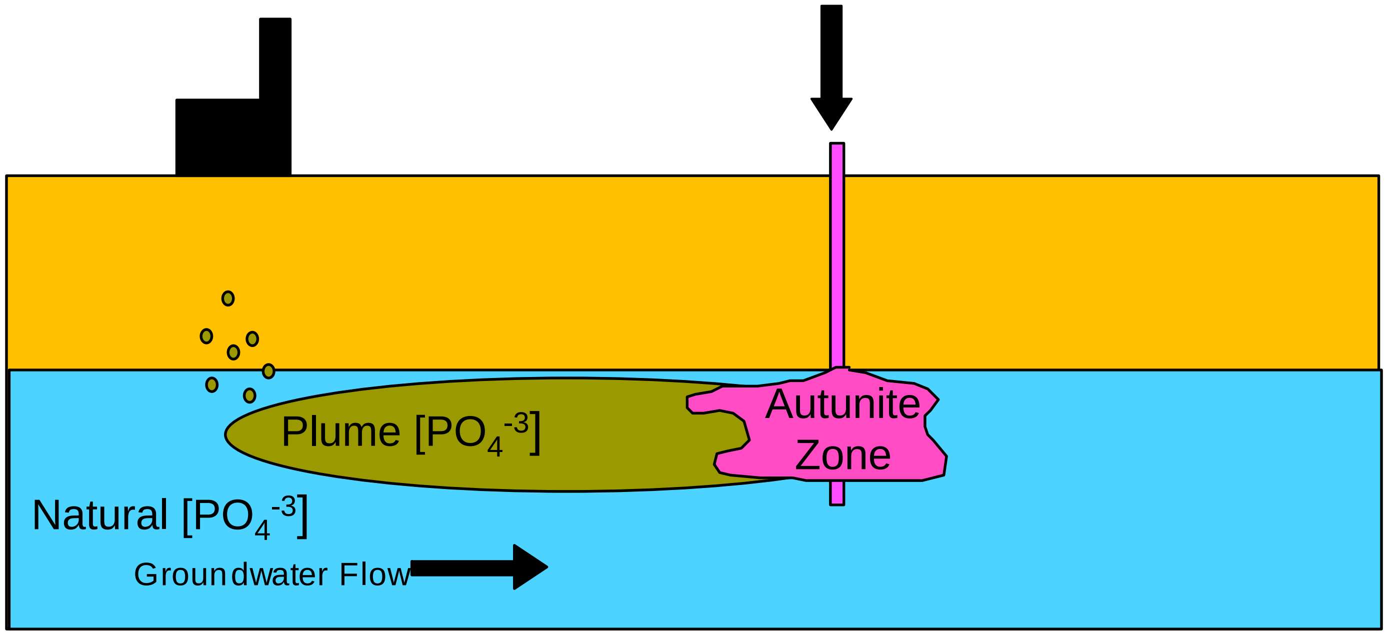
Matrix

Detrital Grains

Greater mass



Uranium Treatment by Addition of Phosphate



Phosphate to induce autunite precipitation

Autunite Zone

Plume [PO_4^{-3}]

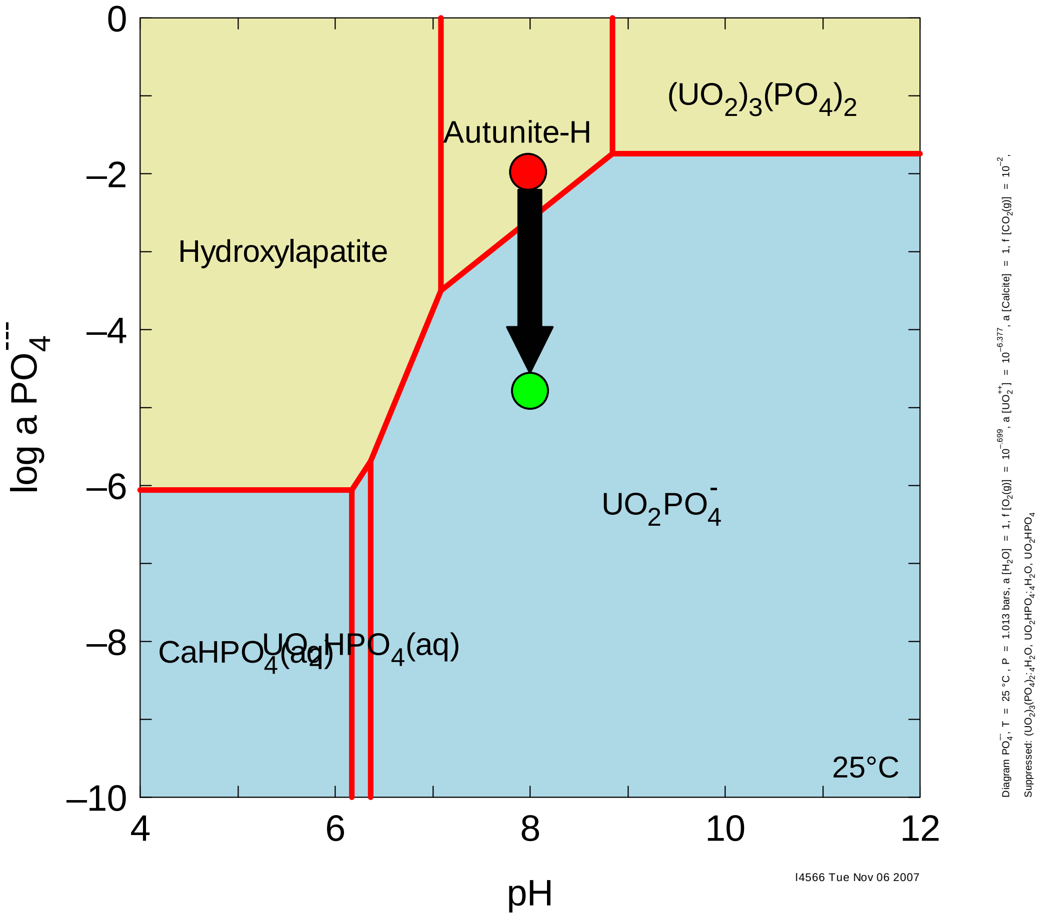
Natural [PO_4^{-3}]

Groundwater Flow

Goal: Force precipitation of insoluble U-phosphates

Effects of Rebound to Natural Phosphate Concentrations

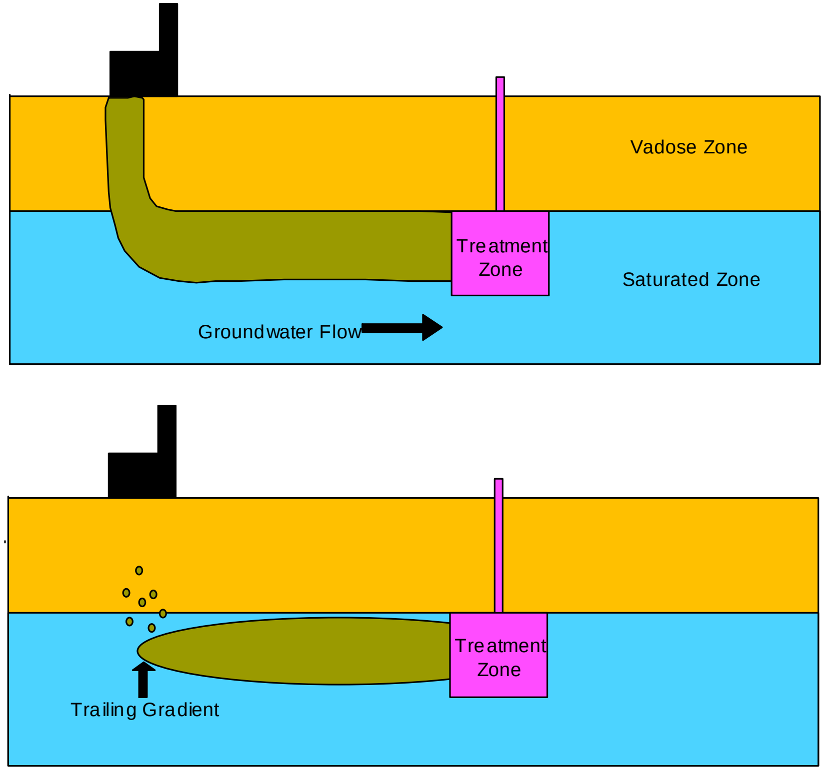
Assumed:
- calcareous sand
- PCO_2=0.01 atm.



Biogeochemical Gradients

- Migrating interface between two different sets of biogeochemical conditions in aquifer
  - Induced by differing conditions associated with natural setting vs. plume vs. remediation
    - In situ remediation is usually emplacement of artificial biogeochemical gradient
- Gradients can include pH, redox potential, mineralogy, activities of non-contaminant species, etc.
  - pH and redox gradients, in particular, can have strong influence on mobility of many metals and radionuclides
- Understanding evolution and migration of biogeochemical gradients provides the framework for using the wealth of basic science on contaminant specific attenuation mechanisms

Trailing Gradients



- Located at trailing edge of plume after source and vadose zone are depleted
- Length of time active remediation must operate depends on migration rate of trailing gradient through treatment zone
- Rate of trailing gradient migration depends on aquifer mineralogy, hydrogeology, plume chemistry, etc.
- Trailing gradients often migrate more slowly than leading gradients because plume chemistry ± remediation chemistry changes critical aquifer properties

DOE EM Initiative

- DOE Office of Environmental Management has funded multi-year initiative to develop the science and guidance necessary to further the use of natural attenuation strategies in closing metal and radionuclide waste sites
  - Will build on EPA guidance on MNA for metals and radionuclides
  - Will focus on overall biogeochemical evolution of waste sites using the gradient evolution conceptual model

Conclusions

- In situ remediation plans for metal and/or radionuclide contaminated sites should include in-depth evaluation of sustainability of proposed technologies
- Sustainability is determined by the balance between biogeochemical conditions induced by active remediation and long-term biogeochemical evolution of waste site
  - Controlling parameters include hydrogeology, differences between remediation - plume - natural biogeochemical conditions, mineralogy, and texture
- Biogeochemical gradient evolution is a simple framework for considering overall evolution of waste sites