Variability in uranium adsorption in saturated-zone sediments from the IFRC tracer-test well array in Hanford’s 300 area uranium plume

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Modeling adsorption variability on contaminated sediments

- Quantify adsorbed uranium
- Determine impact of variable chemistry on uranium adsorption
- Quantify reactive surface area
- Optimize surface complexation model parameters
TEM vadose zone sample
Sediment sample locations
Carbonate extractions

- Sodium carbonate/bicarbonate
- Alkalinity = 20 meq/L
- pH 9.45
- < 2 mm size fraction
- 50 g/L

Carbonate extractions: 2-5
Adsorbed U(VI): 2-5

- Ringold
- Smear zone

Graph showing elevation (m above sea level) against [U$_{Extracted}$] (mol/g) for Adsorbed U(VI) with values ranging from 2 to 5.
Carbonate extractions: composites
Adsorbed U(VI): all sites

Smear zone

Elevation (m above sea level)

[U$_{Extracted}$] (mol/g)

2-5

3-32

3-31

2-26

2-27

2-30

2-31

TTWA

Ringold
Total U vs. adsorbed U(VI)

\[ y = -1.1 \times 10^{-9} + 0.43x \quad R = 0.98 \]

\[ y = -5.9 \times 10^{-10} + 0.17x \quad R = 0.59 \]
Modeling adsorption variability on contaminated sediments

- Quantify adsorbed uranium
- **Determine impact of variable chemistry on uranium adsorption**
- Quantify reactive surface area
- Optimize surface complexation model parameters
U(VI) aqueous speciation

pH = 7.87; calcite equilibrium assumed
U(VI)$_{tot}$ = 2 x 10$^{-6}$ M

- log of concentration (moles/L)

Alkalinity (meq/L)

300 Area Groundwater

Columbia River
U(VI) adsorbs predominantly according to:

1. \( \equiv \text{Fe(OH)}_2 + \text{UO}_2^{2+} + \text{CO}_3^{2-} = \equiv \text{FeO}_2 \text{UO}_2 \text{CO}_3^{2-} + 2\text{H}^+ \)

2. \( \equiv \text{Fe(OH)}_2 + \text{UO}_2^{2+} + 2\text{CO}_3^{2-} = \equiv \text{FeO}_2 \text{UO}_2 (\text{CO}_3)_2^{4-} + 2\text{H}^+ \)

Bargar, Reitmeyer, Lenhart, and Davis (2000)  
U(VI) adsorption experiments

- Artificial groundwater
- Variable alkalinity
- Variable solid/liquid ratio
- With or without added U(VI)
U(VI) desorption: 2-31 shallow
Fraction U(VI) desorbed vs. alkalinity
Modeling U(VI) adsorption in presence of carbonate

\[ >\text{SOH} + \text{UO}_2^{2+} + \text{H}_2\text{O} \rightleftharpoons >\text{SOUO}_2\text{OH} + 2\text{H}^+ \]

\[ >\text{SOH} + \text{UO}_2^{2+} + \text{CO}_3^{2-} \rightleftharpoons >\text{SOUO}_2\text{HCO}_3 \]

\[ >\text{SOH} + \text{UO}_2^{2+} + \text{CO}_3^{2-} \rightleftharpoons >\text{SOUO}_2\text{HCO}_3 \]

Parameters: \( >\text{SOHT} \), \( K_{i,\text{ads}} \)

Independent of chemical conditions
Fraction U(VI) desorbed vs. alkalinity
Next steps

• Expand range of chemical conditions to constraint SCM
• Examine factors influencing variability
• Examine grain-scale U(VI) desorption rate
Conclusions

• Maximum in adsorbed U(VI) in smear zone
• Adsorbed U(VI) ~ 40% of total U in smear zone
• Variability in smear-zone adsorbed U(VI) on the scale of the tracer test well array
• Variability in adsorption properties ~ reactive surface area
• Desorption time-scales similar to U-contaminated sediments from deep vadose zone