Impact of Cr(VI) as a Co-Contaminant on the Sorption and Desorption of U(VI) in Hanford Sediments Under Mildly Alkaline Oxic Conditions

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Background

Hanford Site

- Site built during the Manhattan Project for plutonium production in Benton County, Washington.
- Multiple contamination plumes across the site were created from past operations[1]
- Plumes of co-mingled contaminants including uranium and chromium exist in the 200 Area of the Central Plateau. [3]

Monitored Natural Attenuation/Benefits

- Once active remediation is complete, there is still a need to understand contaminant fate in the subsurface as some contaminants will remain.
- Results will minimize the knowledge gap with respect to U(VI) mobility under site relevant conditions.
- A fundamental understanding of U in the subsurface will help to quantify its long-term mobility and the possibility for monitored natural attenuation (MNA – the controlled use of naturally occurring degradation and retardation processes of contaminants in the subsurface) after active remediation (pump and treat technology) has been completed.

Objectives

Evaluate the impact Cr (VI) has on U(VI) sorption and desorption in Hanford sediments under slightly alkaline conditions, in the presence of major groundwater components at the site.

Specific aims:

- Evaluate change in adsorption-desorption distribution coefficient (K_d) of U(VI) in the presence of Cr(VI)
- Evaluate the change in retardation of U(VI) in the presence of Cr(VI)
- Simulate U(VI) breakthrough data using a two-domain, first order nonequilibrium transport model

Methodology

Materials

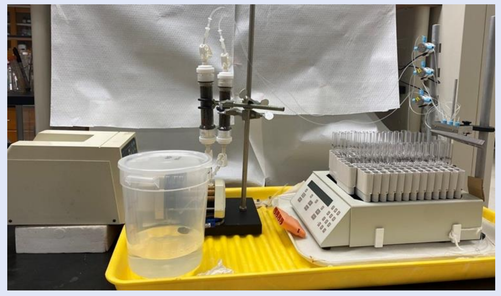
- Uncontaminated sediment (sieved ≤ 2 mm) collected at the Tristate Asphalt gravel pit in Pasco, WA
- U(VI) spiked artificial groundwater (AGW: Na-K-Mg-Ca with Cl-HCO_3-SO_4, pH: 8.10 ± 0.07) – [U(VI)] source: solid uranyl nitrate hexahydrate (International Bio-Analytical Industries, Inc.) Cr(VI) source: $K_2Cr_2O_7$ (High Purity Standards).

Batch Experiments

- U(VI) concentration (168 µmol/L – U:Cr ratio of 1:10)
- 14 days on an end over end tube revolver

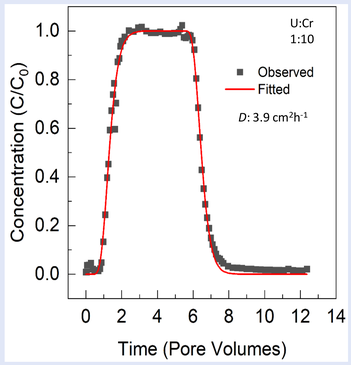
Column Experiments

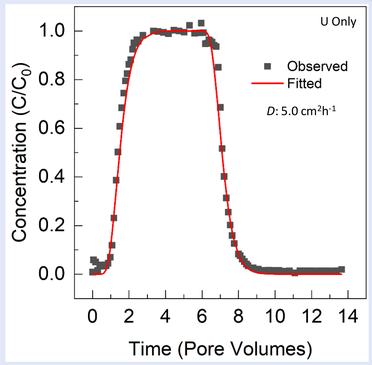
- U(VI) concentration (168 µmol/L – U:Cr ratio of 1:10)
- Nonreactive bromide tracer test to characterize the hydrodynamic flow properties of the sediment
- Stop flow events to distinguish between dispersion and non-equilibrium effects in the columns
- U analysis – ICP-MS, Br analysis – IC



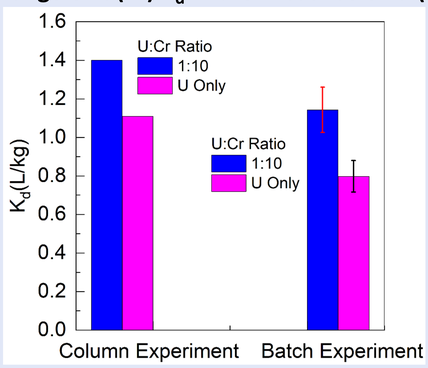
Results/Discussion

Bromide tracer Tests



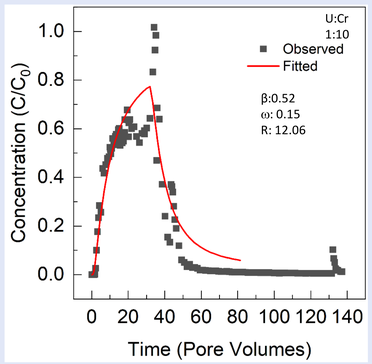


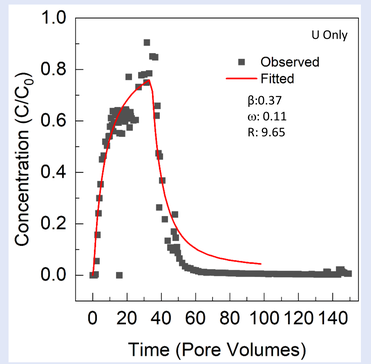
Change of U(VI) K_d in the Presence of Cr(VI)



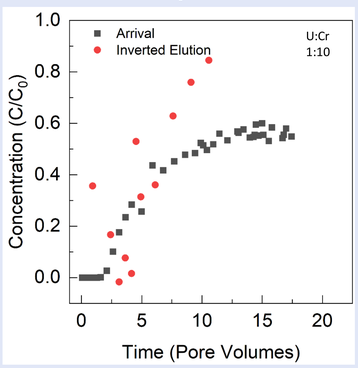
- K_d values obtained from moment analysis of the U(VI) breakthrough curves agree well with those calculated from the results of batch-equilibrium experiments.
- The K_d increased in the presence of Cr at a 1:10 U:Cr ratio from 0.8±0.1 to 1.1±0.1 and 1.1 to 1.4 L/kg in batch and column experiments respectively.

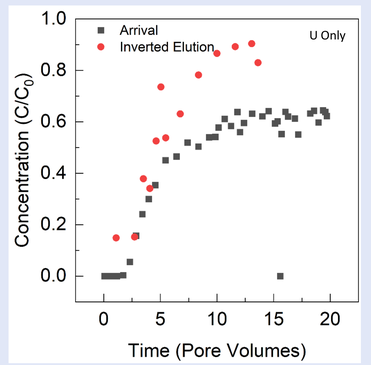
U(VI) Breakthrough Curves





Comparison of Arrival to Inverted Elution Curve





- Impact of rate-limited sorption

Summary

- U(VI) adsorption to Hanford sediment (K_d=1.4 L/kg) and retardation (12.06) in the presence of Cr(VI) is minimal but not further reduced compared to when U(VI) is present alone. This should be considered while developing future MNA procedures.
- **Future Work**: (i) investigate adsorption of U with the inclusion of other contaminants to establish the true adsorption capacity of Hanford sediments; (ii) use of more robust modeling to simulate the impact of stop flow events in each column.

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(1) Gephart, R. E. *Hanford: A Conversation about Nuclear Waste and Cleanup*; Battelle Press: Columbus, Ohio, **2003**.
(2) ResearchGate,https://www.researchgate.net/publication/335043099_Challenges_with_vitrification_of_Hanford_High-Level_Waste_HLW_to_borosilicate_glass_-_An_overview/figures?lo=1.
(3) Hanford Site Groundwater Monitoring Report for 2021, 2022. . Richland, Washington.