Uranium hydrogeochemistry of the Hanford caliche layer

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Introduction

Uranium (U) contamination is widespread in the U.S., particularly at Department of Energy (DOE) sites. The Hanford site in Richland, WA is underlain by Hanford (HF) and Ringold (RG) silts and sands and the $CaCO_3$-cemented Plio-Pleistocene (PP) unit. Recent studies have identified a "new" aqueous complex, $Ca_2UO_2(CO_3)_3$. These related Hanford samples provide an opportunity to investigate the coupled fate and transport of U(VI) in unconsolidated silts and sands compared to $CaCO_3$-cemented sediments.

Results and discussion

Kinetic batch studies demonstrated signficantly greater sorption of U onto PP ($K_f = 3.59$) in comparison to the HF and RG ($K_f = 1.67$ and $K_f = 1.19$, respectively). U sorption is generally associated with Fe-oxides, but measured Fe contents are not consistent with the observed U sorption (Table 2). Thus there are three hypotheses regarding increased sorption in PP: higher surface area, organic-facilitated sorption, and/or differential sorption of the $Ca_2UO_2(CO_3)_3$ complex (Table), which is likely to form given the high carbonate content of the caliche. In support of hypothesis 3 is the observation of increased U sorption in the presence of 2 mM $CaCl_2$ for all sediments.

Sample	*% TIC*	*% TOC*	*Fe (g kg^{-1})*	*SA (m^{2}g^{-1})*
HF	0.22	0.02	4.90	6.13
PP	2.68	0.32	1.61	11.2-14.6
RG	0.26	0.02	7.49	9.11

Conclusions

Additional studies will be undertaken involving selective extractions and X-Ray Absorption Near-Edge Spectroscopy (XANES) to resolve this conundry. Our results will have implications relevant to DOE nuclear facilities and uranium mines.

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The dynamic hydrochemical environment of Lake Tiberias, Israel

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Lake Tiberias (Israel) is a monomictic freshwater lake, situated in one of the pull-apart basins of the Dead Sea transform fault. Known and unspecified inputs of brackish to saline groundwaters enter the reservoir, chiefly through the bottom sediments. From mass balance calculations on a monthly base for the hydrological year 2002/03 an annual supply of unknown inputs of $84*10^6$ m³, $66*10^6$ kg of Cl and $0.5*10^6$ kg of Br is derived. Hydrochemical data of springs and of the lake, combined with discharge patterns and a volume and chemical mass balance of the lake, show that unspecified inputs are hydraulically connected to systems of known onshore spring cluster (Tabgha, Fuliya, Tiberias, eastern springs) and direct subsurface flow from the Hermon area and/or the Golan Heights. Hydrochemical characteristics of unknown inputs differ over the year and are not attributed to a single source. This is supported by the monthly changing Cl/Br ratios (108-163) of unspecified inputs, representing different seasonal start up of individual spring systems, mainly Tabgha (90-110) and Fuliya (140-200).

The influx of water through the bottom sediments is proved by seasonal changing Ce-anomalies at the sediment/water interface in the deeper parts of the lake, irrespective of the prevailing redox conditions. Small changes of Ce-anomalies are less recognisable along shallow lake floor. Changes of rare earth abundances, Cl/Br ratios and $\delta^{18}O$ in spring water are also traced in deep lake waters. Saline water infiltrates all across the lakes floor. However, major inputs occur along faults outside the Dead Sea Graben *sensu stricto*.