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EFFECTS OF SOLUTION pH AND COMPLEXING REAGENTS ON URANIUM AND THORIUM DESORPTION UNDER SATURATED EQUILIBRIUM CONDITIONS

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ABSTRACT

Three contaminated bulk surface soils were used for investigating the effect of solution pH and complexing reagents on uranium and thorium desorption. At a low solution pH, the major chemical species of uranium and thorium, uranyl [UO_2^{+2}], thorium dihydroxide [$Th(OH)_2^{+2}$], and thorium hydroxide [$Th(OH)^{+3}$], tend to [illegible] complexes with acetates in the solution phase, which increases the fractions of uranium and thorium desorbed into this phase. At a high solution pH, important uranium and thorium species such as uranyl tricarbonate complex [$UO_2(CO_3)_3^{-4}$] and thorium tetrahydroxide complex [$Th(OH)_4^0$] tend to resist complexation with acetates. The presence of complexing reagents in solution can release radionuclides such as uranium and/or thorium from the soil to the solution by forming soluble complexes. Sodium bicarbonate ($NaHCO_3$) and diethylenetriaminepentaacetic acid (DTPA) are strong complex formers that released 38% to 62% of total uranium activity and 78% to 86% of total thorium activity, respectively, from the soil samples investigated. Solutions of 0.1 molar sodium nitrate ($NaNO_3$) and 0.1 molar sodium sulfate (Na_2SO_4) were not effective complex formers with uranium and thorium under the experimental conditions. Fractions of uranium and thorium desorbed by 0.15g/200ml humic acid ranged from 4.62% to 6.17% and 1.59% to 7.09%, respectively. This work demonstrates the importance of a knowledge of solution chemistry in investigating the desorption of radionuclides.

INTRODUCTION

In contaminated soils, radionuclides such as uranium and/or thorium may be associated with different chemical species on soil surfaces or inside soil grains with consequent differences in leachability and mobility. Chemical species in contacting solutions can react with soil contaminants by dissolution, ion exchange, or complexation to release contaminants from the soil to the solution. Understanding the effect of chemical species in solution is important in investigating the distribution of uranium and thorium between the soil and the solution under desorption conditions. In this work, the effects of the solution pH and the complexing reagents on the desorption of uranium and thorium under saturated equilibrium conditions were investigated.

EXPERIMENTAL METHODS

Three surface soils — Hazelwood A, Hazelwood B, and Weldon Spring — from St. Louis, Missouri, were sampled, air-dried, and sieved (< 2 mm). Soil properties measured on the prepared material were pH (1:1 in water), cation exchange capacity (CEC),[1] amount of organic carbon,[2] and saturated moisture content.[3] Concentrations of uranium and thorium were measured by dissolving uranium and thorium from soil into a solution by using an acid dissolution procedure.[4] After solubilization, uranium and thorium were separated on an anion exchange column. Uniform, thin-film, solid counting samples were then prepared for alpha-spectrometry. Triplicate measurements were carried out for the chemical and radioactivity analyses. The concentrations of metal ions on the surface of soil particles were measured with inductively coupled plasma-atomic emission spectrometry (ICP-AES).[5] Inorganic anions such as fluoride (F^-), chloride (Cl^-), nitrate (NO_3^-), phosphate (PO_4^{-3}), and sulfate (SO_4^{-2}) in soil were determined by ion chromatography. Concentrations of carbonate (CO_3^{-2}) and bicarbonate (HCO_3^-) in soil were quantitatively determined by potentiometric titration.[6]

A kinetic study was performed to search for the equilibrium contacting time for the subsequent saturated batch experiments. The Hazelwood B soil was used in the kinetic study because of its relatively high

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concentrations of uranium and thorium compared with the other two soil samples. Two contact solutions were used in the kinetic study: laboratory distilled deionized water with a pH of 6.1 and a buffer solution with a pH of 6.5. The latter was prepared by diluting a solution of 200 ml of 1.0 molar potassium dihydrogen phosphate (KH_2PO_4) plus 55.6 ml of 1.0 molar sodium hydroxide (NaOH) to 4.0 L with laboratory-distilled deionized water. The pH of this buffer solution was adjusted with NaOH and hydrochloric acid (HCl) solutions. In the kinetic study, 5-g samples of soil were each agitated with 200 ml of contacting solution for 30 minutes, 2 hours, 2 days, and 7 days, respectively. Then the soil and the solution were separated by centrifugation at 2,000 rpm for 30 minutes. Activities of uranium and thorium in the solution phase were measured by alpha-spectrometry.[4]

The effect of the solution pH on the uranium and thorium desorption under batch extraction was investigated by adjusting the solution pH of 1 molar sodium acetate (NaOAc) solution to 4.0, 5.6, and 8.0. The effects of the complexing reagents were investigated under equilibrium batch extraction with 0.1 molar sodium nitrate ($NaNO_3$), 0.1 molar sodium sulfate (Na_2SO_4), 0.1 molar sodium bicarbonate ($NaHCO_3$), 0.15g/200ml humic acid, and 0.1 molar diethylenetriaminepentaacetic acid (DTPA) with a pH of 8.0.

RESULTS AND DISCUSSION

Properties of the experimental soils are listed in Table 1. The soils are moderately alkaline, which suggests the presence of free carbonate. Uranium and thorium concentration measurements are summarized in Table 2. At both the Weldon Spring and Hazelwood sites, uranium concentrations vary from 0.74 to 12.45 Bq/(g soil). Concentrations of thorium-230 in Hazelwood soils [17.09 and 85.36 Bq/(g soil)] are much higher than in Weldon Spring soil [0.22 Bq/(g soil)]. The results of the acid soluble elemental analysis listed in Table 3 indicate that aluminum (Al^{+3}), ferrous iron (Fe^{+2}), calcium (Ca^{+2}), and magnesium (Mg^{+2}) are important metal ions in the soil solution systems investigated. In Hazelwood B soil, the metal concentrations of cobalt (Co) and nickel (Ni) were about 30 to 100 times larger than those found in Hazelwood A and Weldon Spring soils, respectively. Soil concentrations of inorganic anions as measured by ion chromatography are summarized in Table 4. The F^- concentration in Weldon Spring soil was about 20 times larger than that found in Hazelwood soils, and the HCO_3^- concentration in Weldon Spring soil was 1.5 times larger than that in Hazelwood soils.

Results of the kinetic study, which are plotted in Figure 1, indicate that the fractions of uranium and thorium desorbed approached a constant value after two days of agitation. Therefore, two days contacting time was used for the subsequent batch experiments. In Table 5, the fractions of total uranium and thorium activity desorbed by 1 molar NaOAc solutions at different solution pHs and the related distribution coefficients are summarized. Distribution coefficients for uranium and thorium under desorption conditions were calculated as the ratio of uranium and/or thorium activities remaining in soil to that released into the solution. Table 5 shows that desorption of uranium and thorium increased by lowering the pH of the contacting solution. Therefore, the calculated distribution coefficients for uranium and thorium desorption in an acidic soil solution would be decreased. A possible explanation is based on the major uranium species in a natural water system as predicted by Langmuir.[7] In a typical groundwater system with a solution pH of 4.0, 5.6, and 8.0, the important uranium species are uranyl [UO_2^{+2}], uranium carbonate complex [$UO_2CO_3^0$], and uranyl tricarbonate complex [$UO_2(CO_3)_3^{-4}$], respectively.[7] Compared to $UO_2CO_3^0$ and $UO_2(CO_3)_3^{-4}$, UO_2^{+2} has a strong tendency to form complexes with acetate with cumulative formation

TABLE 1 Physical and Chemical Properties of Soil Samples[a]

Soil Sample	pH	CEC (meq/100g)	Organic Carbon (wt %)	Saturated Moisture Content (wt %)
HWA	8.47	43.59	4.28	37.5
HWB	8.35	45.89	5.77	47.0
WS	8.14	69.28	3.80	64.0

[a] HWA = Hazelwood A; HWB = Hazelwood B; WS = Weldon Spring.

TABLE 2 Uranium and Thorium Concentrations in Soil Samples[a]

	Concentration × 10^{-2} Bq/g		
Radionuclide	HWA	HWB	WS
Th-228	3.54 ± 0.15	7.84 ± 0.45	6.01 ± 0.31
Th-230	1,700 ± 70	8,520 ± 510	10.8 ± 0.5
Th-232	5.31 ± 0.23	8.55 ± 0.49	5.36 ± 0.27
U-234	36.9 ± 1.8	446 ± 15	611 ± 23
U-235	0.41 ± 0.04	16.8 ± 0.6	25.6 ± 1.0
U-238	37.0 ± 1.8	433 ± 15	608 ± 23

[a] HWA = Hazelwood A; HWB = Hazelwood B; WS = Weldon Spring.

TABLE 3 Elemental Analysis of Soils by ICP-AES (average from duplicate measurements)

	Soil Sample					
	Hazelwood A		Hazelwood B		Weldon Spring	
Element	mg/g soil	mmole/g soil	mg/g soil	mmole/g soil	mg/g soil	mmole/g soil
Ag	<0.006	$<6 \times 10^{-5}$	0.016 ± 0.010	$(1.5 \pm 0.9) \times 10^{-4}$	<0.006	$<6 \times 10^{-5}$
Al	7.32 ± 0.50	0.271 ± 0.019	8.84 ± 0.40	$(3.28 \pm 0.15) \times 10^{-1}$	12.68 ± 1.60	0.470 ± 0.059
As	<0.060	$<8 \times 10^{-4}$	0.14 ± 0.01	$(1.9 \pm 0.1) \times 10^{-3}$	0.070 ± 0.001	$(9.3 \pm 0.1) \times 10^{-4}$
Ba	3.88 ± 0.20	$(2.83 \pm 0.15) \times 10^{-2}$	0.75 ± 0.02	$(5.5 \pm 0.10) \times 10^{-3}$	0.21 ± 0.00[a]	$(1.5 \pm 0.0)^{a} \times 10^{-3}$
Be	<0.001	$< 1 \times 10^{-4}$	0.002 ± 0.00[a]	$(2 \pm 0)^{a} \times 10^{-4}$	<0.001	$<1 \times 10^{-4}$
Ca	8.92 ± 0.04	0.222 ± 0.001	17.22 ± 0.88	0.430 ± 0.022	12.48 ± 0.02	0.311 ± 0.001
Cd	<0.002	$<2 \times 10^{-5}$	0.004 ± 0.00[a]	$(4 \pm 0)^{a} \times 10^{-5}$	<0.020	$<1.8 \times 10^{-4}$
Co	0.06 ± 0.00[a]	$(1 \pm 0)^{a} \times 10^{-3}$	1.98 ± 0.06	$(3.26 \pm 0.10) \times 10^{-2}$	0.012 ± 0.002	$(2.0 \pm 0.0)^{a} \times 10^{-4}$
Cr	0.02 ± 0.00[a]	$(4.0 \pm 0.0)^{a} \times 10^{-4}$	0.04 ± 0.00[a]	$(8.0 \pm 0.0)^{a} \times 10^{-4}$	0.02 ± 0.00[a]	$(4.0 \pm 0.0)^{a} \times 10^{-4}$
Cu	0.07 ± 0.00[a]	$(1 \pm 0)^{a} \times 10^{-3}$	1.34 ± 0.04	0.021 ± 0.001	0.02 ± 0.00[a]	$(4 \pm 0)^{a} \times 10^{-4}$
Fe	13.94 ± 0.34	0.250 ± 0.006	19.30 ± 0.92	0.346 ± 0.016	19.44 ± 0.96	0.348 ± 0.017
K	1.01 ± 0.04	$(2.59 \pm 0.10) \times 10^{-2}$	1.39 ± 0.08	$(3.55 \pm 0.20) \times 10^{-2}$	2.24 ± 0.18	$(5.73 \pm 0.46) \times 10^{-2}$
Mg	3.66 ± 0.14	0.151 ± 0.006	10.38 ± 0.48	0.427 ± 0.020	3.68 ± 0.20	0.151 ± 0.008
Mn	0.72 ± 0.02	0.013 ± 0.000[a]	1.00 ± 0.04	0.018 ± 0.001	0.60 ± 0.00[a]	0.011 ± 0.000[a]
Na	0.19 ± 0.03	$(8.2 \pm 1.4) \times 10^{-3}$	0.22 ± 0.04	$(9.4 \pm 1.6) \times 10^{-3}$	0.16 ± 0.01	$(7.0 \pm 0.4) \times 10^{-3}$
Ni	0.09 ± 0.00[a]	$(1.5 \pm 0.0)^{a} \times 10^{-3}$	2.42 ± 0.10	0.041 ± 0.002	0.026 ± 0.000[a]	$(4.4 \pm 0.0)^{a} \times 10^{-4}$
Pb	0.14 ± 0.01	$(6.8 \pm 0.0)^{a} \times 10^{-4}$	0.19 ± 0.01	$(9.2 \pm 0.0)^{a} \times 10^{-4}$	0.05 ± 0.01	$(2.4 \pm 0.0)^{a} \times 10^{-4}$
Sb	<0.060	$<4.90 \times 10^{-4}$	<0.060	$<4.90 \times 10^{-4}$	<0.060	$<4.90 \times 10^{-4}$
Se	<0.060	$<7.6 \times 10^{-4}$	0.082 ± 0.004	$(1.0 \pm 0.1) \times 10^{-3}$	<0.060	$<7.6 \times 10^{-4}$
Ti	0.19 ± 0.01	$(4.0 \pm 0.2) \times 10^{-3}$	0.26 ± 0.02	$(5.4 \pm 0.3) \times 10^{-3}$	0.16 ± 0.02	$(3.4 \pm 0.3) \times 10^{-3}$
V	0.05 ± 0.00[a]	$(1.0 \pm 0.0)^{a} \times 10^{-3}$	0.51 ± 0.02	0.010 ± 0.000[a]	0.038 ± 0.002	$(7.5 \pm 0.0)^{a} \times 10^{-4}$
Zn	0.09 ± 0.00[a]	$(1.4 \pm 0.0)^{a} \times 10^{-3}$	0.12 ± 0.01	$(1.8 \pm 0.1) \times 10^{-3}$	0.094 ± 0.004	$(1.4 \pm 0.1) \times 10^{-3}$

[a] A standard deviation of 0.00 is the result of rounding.

TABLE 4 Inorganic Anion Concentration in Soil Samples (average from duplicate measurements)

	Soil Sample					
	Hazelwood A		Hazelwood B		Weldon Spring	
Inorganic Anion	µg/g soil	µmole/g soil	µg/g soil	µmole/g soil	µg/g soil	µmole/g soil
F^-	20 ± 0	1.0 ± 0.0	25 ± 0	1.3 ± 0.0	393 ± 8	20.7 ± 0.5
Cl^-	10 ± 0	0.28 ± 0.00	10 ± 0	0.28 ± 0.00	17.5 ± 2.5	0.49 ± 0.07
HCO_3^-[a]	1.42×10^6	2.32×10^4	1.38×10^6	2.26×10^4	2.28×10^6	3.73×10^4
NO_3^-	95 ± 5	3.17 ± 0.17	67.5 ± 2.5	2.25 ± 0.08	125 ± 0	4.17 ± 0.00
PO_4^{-3}	180 ± 0	1.90 ± 0.00	155 ± 5	1.63 ± 0.05	210 ± 5	2.21 ± 0.05
SO_4^{-2}	147.5 ± 2.5	1.54 ± 0.03	205 ± 0	2.13 ± 0.00	315 ± 0	3.28 ± 0.00

[a] Single measurement.

constants such as Log β_1 = 2.52, Log β_2 = 4.40, and Log β_3 = 6.20.[8] Therefore, when the pH of the contact solution is lowered to 4.0, more free UO_2^{+2} can be released into the solution phase by forming a soluble complex with acetate. The cumulative formation constants for Th^{+4} with acetate are reported as Log β_1 = 4.88, Log β_2 = 6.91, and Log β_3 = 9.05.[9] Because thorium has a strong tendency to hydrolyze with OH^-, the important species present in natural water systems are expected to be thorium dihydroxide ($ThOH_2^{+2}$) and thorium hydroxide ($ThOH^{+3}$) for a solution with pH 4.0 and thorium tetrahydroxide complex ($ThOH_4^0$) for solutions with pH 5.6 and 8.0.[10] It was concluded that, at pH = 8.0, thorium was not present in the appropriate form to complex with acetate. When the pH was lowered to 5.6 and 4.0, $ThOH^{+3}$ and $ThOH_2^{+2}$

became more important, thus allowing thorium complex-formation with acetate. It can also be predicted that, at pH< 3.0, a high fraction of thorium species will be released to the solution phase as free Th^{+4} or Th^{+4}-acetate complexes.

Fractions of total soil uranium and thorium activities desorbed by different complexing agents and related empirical distribution coefficients are summarized in Table 6. Solutions of 0.1 molar $NaNO_3$ and 0.1 molar Na_2SO_4 were not effective complex formers with uranium and thorium under the experimental conditions. Sodium bicarbonate ($NaHCO_3$) removed about 38% to 62% of uranium from the soils, and DTPA removed 78% to 86% of thorium from the soils. Fractions of uranium and thorium desorbed by 0.15g/200ml humic acid ranged from 4.62% to 6.17% and 1.59% to 7.09%, respectively.

The distribution of different carbonate species in solution is controlled by the solution pH. In a closed batch system, at pH>8.0, the major carbonate species in the carbonate system is CO_3^{-2},[11] which acts as a bridging ligand for complexation.[12] The structure of the solid phases between UO_2^{+2} and the carbonate anions shows that the carbonate anions are bidentate. Stability constants for the 1:1, 1:2, and 1:3 complexes reported in the literature agree very well with the best value for Log β_1, Log β_2, and Log β_3 as 10.0, 17.0, and 21.6, respectively.[13] In the soil solution system, metal ions adsorbed on the soil surface or bound within soil aggregates can be released to the solution phase and compete with UO_2^{+2} to form complexes with CO_3^{-2}. Table 3 shows that the important acid soluble metal ions in the soil samples investigated were Al^{+3}, Fe^{+2}, Ca^{+2}, and Mg^{+2}. Because Al^{+3} has a strong tendency to form hydroxide compounds, interactions between Al^{+3} and carbonates are not important for the system investigated here. The equilibrium constants listed in Table 7 indicate that Fe^{+2} and Mg^{+2} have stronger tendencies to precipitate with OH^- than with CO_3^{-2}.[14] Therefore, in the soil solution, Ca^{+2} and fractions of Fe^{+2} and Mg^{+2} will compete with UO_2^{+2} for the available CO_3^{-2} species in solution.

The 0.1 molar DTPA solution with a pH of 8.0 was effective for desorbing uranium and thorium from the soil samples under the given experimental conditions. It has been reported that DTPA can form stable multidentate complexes with metal cations with the three amino nitrogens and five carboxylate groups available for bonding.[15] Complexes of DTPA with thorium ions are very strong because both nitrogen and oxygen atoms in DTPA coordinate to the thorium. In addition, the ionic radius of thorium, 0.85Å, is very small for coordination number 8, which makes the Th-DTPA complexes very

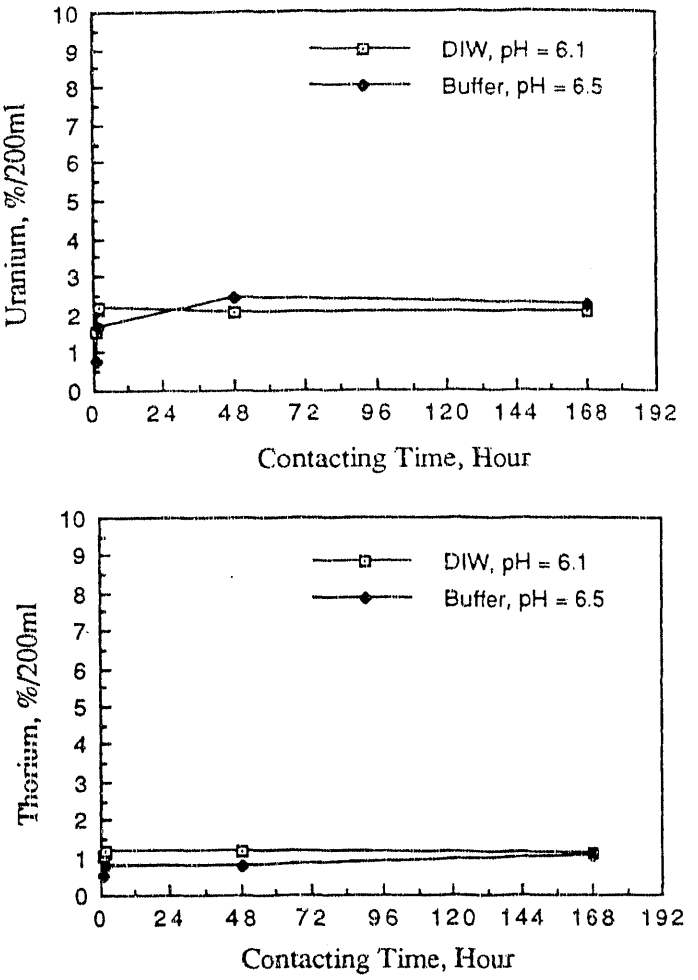

FIGURE 1 Fractions of (a) Total Uranium Activity and (b) Total Thorium Activity Released into 200 ml of Contact Solution at Different Contacting Times (DIW = laboratory distilled deionized water; Buffer = potassium dihydrogen phosphate solution with pH adjusted to 6.5)

stable in the solution phase.[15] When complexes between DTPA and uranium were investigated, it was reported that DTPA is potentially capable of satisfying the coordination number 8 for the U^{+4} species in simple 1:1 complexes. The uranyl ion UO_2^{+2} forms complexes with DTPA with a lower stability and smaller coordination number than the U^{+4} species because of its geometry and lower effective metal charge. The presence of oxygen atoms in the UO_2^{+2} ions is a steric hindrance to higher than tridentate chelation with a single ligand molecule.[15]

The humic acids constitute an extremely complex group of compounds: the phenolic and carboxylic groups are two common functional groups. Because the structure of humic acids is not well defined, there is no compound that can act as a true model for humic acids. Although there is no doubt about the ability of humic

TABLE 5 Fraction (%) of Total Uranium and Total Thorium Activities Desorbed by 1 Molar NaOAc Solution with pH Adjusted to 4.0, 5.6, and 8.0, and Calculated Distribution Coefficients[a]

Soil Sample	Solution pH	Uranium		Thorium	
		% Desorption	K_d, ml/g	% Desorption	K_d, ml/g
HWA	4.0	55.4 ± 3.2	32.2 ± 1.9	7.07 ± 0.10	526 ± 7
	5.6	16.3 ± 1.8	205.4 ± 22.7	0.081 ± 0.004	(4.93 ± 0.24) × 10^4
	8.0	3.53 ± 0.66	1,090 ± 200	0.04 ± 0.02	(10.0 ± 5.0) × 10^4
HWB	4.0	67.7 ± 8.3	19.1 ± 2.3	23.9 ± 3.1	127 ± 17
	5.6	43.4 ± 2.7	52.2 ± 3.2	0.33 ± 0.05	(1.21 ± 0.18) × 10^4
	8.0	7.37 ± 1.21	503 ± 83	0.06 ± 0.02	(6.66 ± 2.22) × 10^4
WS	4.0	37.2 ± 1.1	67.5 ± 2.0	36.9 ± 1.4	68.4 ± 2.6
	5.6	19.5 ± 3.4	165 ± 29	11.4 ± 2.5	311 ± 68
	8.0	5.41 ± 0.50	699 ± 65	3.82 ± 1.33	1,010 ± 350

[a] HWA = Hazelwood A; HWB = Hazelwood B; and WS = Weldon Spring.

TABLE 6 Fraction (%) of Total Uranium and Total Thorium Activities Desorbed by Different Complexing Agents and Calculated Distribution Coefficients[a]

Complexing Agents	Soil Samples	Uranium		Thorium	
		% Desorption	K_d, ml/g	% Desorption	K_d, ml/g
0.1M $NaNO_3$ pH = 8.0	HWA	2.45 ± 0.05	1,590 ± 40	0.048 ± 0.020	(10.1 ± 4.4) × 10^4
	HWB	1.91 ± 0.33	2,120 ± 420	(8.2 ± 1.6) × 10^{-3}	(5.1 ± 1.2) × 10^5
	WS	5.95 ± 1.16	658 ± 134	4.90 ± 3.88	1,320 ± 690
0.1M $NaHCO_3$ pH = 8.0	HWA	62.0 ± 9.7	26.2 ± 10.0	0.135 ± 0.078	(9.1 ± 6.9) × 10^4
	HWB	51.9 ± 5.5	38.0 ± 8.4	0.032 ± 0.024	(2.0 ± 1.0) × 10^5
	WS	38.0 ± 3.2	67.0 ± 7.7	5.13 ± 3.19	1,050 ± 520
0.1M Na_2SO_4 pH = 8.0	HWA	2.42 ± 0.23	1,630 ± 170	0.051 ± 0.035	(11.6 ± 5.6) × 10^4
	HWB	6.40 ± 5.00	1,080 ± 700	0.011 ± 0.003	(3.80 ± 0.9) × 10^5
	WS	5.33 ± 0.15	711 ± 21	2.43 ± 0.53	1,700 ± 440
0.1 M DTPA pH = 8.0	HWA	13.0 ± 0.6	269 ± 15	81.9 ± 2.5	8.88 ± 1.51
	HWB	13.0 ± 0.3	269 ± 7	85.9 ± 3.0	6.62 ± 1.64
	WS	8.72 ± 1.40	432 ± 79	78.1 ± 11.8	12.5 ± 8.6
0.15g/200ml Humic acid pH = 8.0	HWA	5.90 ± 0.86	651 ± 96	1.59 ± 0.22	2,530 ± 390
	HWB	6.17 ± 0.38	610 ± 38	2.29 ± 0.52	1,300 ± 450
	WS	4.62 ± 0.32	829 ± 62	7.09 ± 1.84	558 ± 132

[a] HWA = Hazelwood A; HWB = Hazelwood B; and WS = Weldon Spring.

acids to combine with metals, the nature of the association is still open to investigation. Two modes of binding appear to be significant between the humic acids and the metals:[11] 1) the formation of complexes or chelates between the functional groups of the humic substance and the metal and 2) the association between the humic substance and a peptized colloidal particle of metal hydroxides possibly through sorption on the surface of the particle. Formation of one or another of these associations between humic acids and metals depends on the initial states of the humic acids and metals and their absolute and relative concentrations.

TABLE 7 Equilibrium Constants between Ca^{+2}, Mg^{+2}, Fe^{+2}, and CO_3^{-2}

Metal Ion	Log. Equilibrium Constant
$CaCO_3$	Kso: -8.22 (25°C)
$CaCO_3^0$	K_1: 4.39 (22°C), 5.34 (60°C)
$FeCO_3$	Kso: -11.04 (50°C), -11.21 (60°C),
$Fe(OH)_2$	-11.95 (100°C)
	Kso: -15.10
$MgCO_3$	Kso: -8.09 (25°C)
$Mg(OH)_2$	Kso: -11.23

Source: Ref. 14

CONCLUSION

Three contaminated surface soils (Hazelwood A, Hazelwood B, and Weldon Spring soils) were used to investigate the effects of solution pH and complexing reagents on uranium and thorium desorption. A study of the solution pH effect indicated that the fractions of total uranium and thorium activities released into the solution phase increased by lowering the solution pH. It was concluded that at a low solution pH, the major chemical species of uranium and thorium, UO_2^{+2}, $Th(OH)_2^{+2}$, and $Th(OH)^{+3}$, tend to form complexes with acetates in the solution phase. At a high solution pH, important uranium and thorium species such as $UO_2(CO_3)_3^{-4}$ and $Th(OH)_4^0$ tend to resist complexation with acetates.

The presence of complexing reagents in solution can cause the release of radionuclides such as uranium and/or thorium from the soil to the solution by forming soluble complexes. The 0.1 molar $NaNO_3$ and 0.1 molar Na_2SO_4 were not effective desorption or solubilizing agents for uranium and thorium, possibly due to low complex stability. Sodium bicarbonate and DTPA are strong complex formers that released 38% to 62% of total uranium activity and 78% to 86% of total thorium activity, respectively, from the soil samples investigated. Fractions of uranium and thorium desorbed by 0.15g/200ml humic acid ranged from 4.62% to 6.17% and 1.59% to 7.09%, respectively. Because the structure of humic acids is not well defined, the nature of the association between uranium and thorium with humic acids is still questionable. Therefore, an understanding of solution chemistry is important in investigating the desorption of radionuclides. A computer model capable of predicting possible reactions between radionuclides in soil and chemical species in solution has to be developed for investigating radionuclide mobility and transport.

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