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THE URANIUM WEATHERING RATE AS AN INDICATOR  
OF URANIUM MOBILITY IN CONTAMINATED SOIL  
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Richard A. Sturn  
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of the requirements for  
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**THE URANIUM WEATHERING RATE AS AN INDICATOR OF URANIUM  
MOBILITY IN CONTAMINATED SOIL**

**By**

**Richard A. Sturn**

**A THESIS**

**Submitted to  
Michigan State University  
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## ABSTRACT

### THE URANIUM WEATHERING RATE AS AN INDICATOR OF URANIUM MOBILITY IN CONTAMINATED SOIL

By

Richard A. Sturn

The goals of this study were to determine the rate of uranium weathering in contaminated soil from the Fernald Environmental Project (FEMP) site near Cincinnati Ohio and to estimate from this rate the maximum solution-phase U concentrations expected during typical leaching events at the site. Uranium release rates were measured in four experiments with different soil-to-solution ratios and flow rates using a mixed-flow reactor and a pH 7.4 1.67 mM CaCl<sub>2</sub> influent solution. The experimental uranium weathering rate,  $0.20 \pm 0.01 \mu\text{g U h}^{-1} \text{g}_{\text{soil}}^{-1}$ , was independent of the steady-state uranium concentration over the range in these experiments, 0.038 to 0.235 ppm U, and applied to a relatively small labile fraction of the total soil uranium. Qualitative calculations based on this rate predict that the uranium concentrations of the soil solution should be about 1 ppm during leaching events, far above that typical of natural waters.

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## **INTRODUCTION**

Soils containing inorganic contaminants such as uranium are potential sources of groundwater contamination. The contaminant phases present in the soil and the rate at which they weather largely control contaminant mobility and the aqueous concentration of the contaminant entering the shallow groundwater system. Thus, an estimate of a contaminant's weathering rate can provide a measure of mobility that is useful for risk assessment and remediation decisions.

Uranium-contaminated soil is a potential source of groundwater contamination at the Fernald Environmental Management Project, located approximately 30 miles northwest of Cincinnati, Ohio. The Fernald Environmental Management Project (FEMP) was previously called the Feed Materials Production Center at Fernald, Ohio, which from 1954 to 1989 produced various uranium and thorium products for the U.S. weapons complex. As a consequence of the metallurgical and chemical processes that occurred at FEMP, approximately 2,000,000 m<sup>3</sup> of soil was contaminated with uranium. The uranium-contaminated soil overlies approximately 50 feet of glacial till that caps a regional buried-valley aquifer (Sminchak et. al., 1996). Though composed predominantly of low permeability clay and silt, the till contains interconnected sand and gravel lenses that provide potential migration pathways into this important groundwater resource (Sminchak et. al., 1996).

Uranium, which was released through effluent leaks, solid product spills, and by deposition of airborne particulates from plant incinerators, exists mainly as discrete uranium-rich particles rather than sorbed on clays or organic matter (Francis et al., 1993; Lee and Marsh, 1992). X Ray absorption spectroscopy indicates that uranium is mostly hexavalent, but large (>10 μm) U(IV)-bearing phases, presumably uraninite (UO<sub>2</sub>), are also present (Bertsch et al., 1994). The following uranium-bearing phases have been identified in FEMP soils by a combination of SEM-EDS and analytical electron

microscopy: uranyl phosphate minerals, uranyl silicates, uranium (IV) oxides (presumably uraninite), a calcium uranium (VI) oxide, and uranium associated with amorphous iron oxides (Buck et al. 1993, Lee and Marsh, 1992).

Many U(VI) phases are soluble in oxidizing, carbonate-rich environments such as surface soils at FEMP (Table 1). Of the U(VI) minerals identified in FEMP soil, the uranyl phosphate minerals are the least soluble, followed by uranyl silicates. The solubility of U(VI) phases increases in the presence of carbonate and other oxygen-containing ligands, which form strong complexes with the uranyl ion (Table 2). Anionic uranyl carbonate complexes are the most abundant form of aqueous U(VI) with concentrations typically several orders of magnitude greater than that of the free uranyl ion. Uranyl complexes are predominately anionic, which minimizes uranyl ion sorption by negatively charged clays and organic matter.

Table 1. Common uranium (VI) minerals and their solubility products.

Mineral Name	Formula	Log ( $K_{sp}$ )
carnotite	$K_2(UO_2)_2(VO_4)_2 \bullet 1-3H_2O$	-56.9
tyuyamunite	$Ca(UO_2)_2(VO_4)_2 \bullet 7-11H_2O$	-53.41
K-autunite	$K_2(UO_2)_2(PO_4)_2 \bullet 8-12H_2O$	-48.099
autunite	$Ca(UO_2)_2(PO_4)_2 \bullet 8-12H_2O$	-43.927
uranophane	$Ca(UO_2)_2(SiO_3)_2(OH)_2 \bullet 5H_2O$	-17.490
rutherfordite	$UO_2CO_3$	-14.434
haweite	$Ca(UO_2)_2(Si_6O_{15})$	-6.329
schoepite	$UO_2(OH)_2 \bullet H_2O$	-4.724
soddite	$(UO_2)(SiO_4) \bullet 2H_2O$	-0.512

Solubility products for tyuyamunite and caronite are from Langmuir (1978); all others are from the MINTEQA2 database (U.S. EPA; CREAM 1991)

Table 2. Equilibria for some important U(VI) complexes in solution.

Reaction	log K (25C)	Reference
$2\text{UO}_2^{2+} + 3\text{OH}^- + \text{CO}_3^{2-} = (\text{UO}_2)_2(\text{OH})_3(\text{CO}_3)^-$	41	Maya, 1982
$\text{UO}_2^{2+} + 2\text{CO}_3^{2-} = \text{UO}_2(\text{CO}_3)_2^{2-}$	16.2	Maya, 1982
$\text{UO}_2^{2+} + \text{PO}_4^{3-} = \text{UO}_2\text{PO}_4^-$	13.25	Sandino, 1992
$\text{UO}_2^{2+} + 2\text{H}_2\text{O} = \text{UO}_2(\text{OH})_2 + 2\text{H}^+$	12	Lemire, 1980
$\text{UO}_2^{2+} + \text{CO}_3^{2-} = \text{UO}_2(\text{CO}_3)$	9.87	Civatta, 1979
$\text{UO}_2^{2+} + \text{HPO}_4^{2-} = \text{UO}_2\text{HPO}_4^-$	7.28	Sandino, 1992
$\text{UO}_2^{2+} + \text{CH}_3\text{COO}^- = \text{UO}_2(\text{CH}_3\text{COO})^+$	6.4	Aharland, 1953
$2\text{UO}_2^{2+} + \text{H}_2\text{O} = (\text{UO}_2)_2(\text{OH})_2^{2+} + 2\text{H}^+$	-5.6	Lemire, 1980
$\text{UO}_2^{2+} + \text{H}_2\text{O} = \text{UO}_2(\text{OH})^+ + \text{H}^+$	-5.8	Lemire, 1980
$3\text{UO}_2^{2+} + 5\text{H}_2\text{O} = (\text{UO}_2)_3(\text{OH})_5^+ + 5\text{H}^+$	-15.6	Lemire, 1980

In contrast to the high solubility and mobility of U(VI) in carbonate-rich environments, U(IV) minerals such as uraninite, which may persist in oxidizing environments, are highly insoluble. At circumneutral to acidic pH, groundwater at equilibrium with uraninite typically contains less than 0.01 ppb (Langmuir, 1979).

Since uranium exists mainly as discrete U-rich particles in FEMP soil, uranium release will occur largely by dissolution of these U-bearing phases. If dissolution rates and surface areas of each uranium mineral are known, a composite weathering rate could be derived by summing the individual dissolution rates. Several workers have used this approach for silicate minerals to model watershed response to acid deposition (Sverdrup, 1989, Sverdrup and Warfvinge 1993) and to calculate the composition of weathering solutions (Made and Fritz, 1989, 1990). However, since this soil contains a mixture of crystalline and noncrystalline uranium phases with a range of particle sizes, it is not possible to measure the surface area of each phase. Also, uraninite is the only uranium

mineral for which an empirical dissolution rate has been determined (Grandstaff, 1976; Bruno et al., 1991). An alternative approach is to measure the overall uranium weathering rate directly from a soil sample.

Mixed-flow reactors, which consist of a well-stirred reaction vessel through which solution flows at a constant rate, are suitable for measuring the net rate of uranium release from FEMP soil. They have been used to measure the dissolution kinetics of silicate minerals (Chou and Wollast, 1985; Mast and Drever 1987; Dove and Crerar, 1990), of uranium oxides (Bruno et al., 1991), and of mineral mixtures (Swoboda-Colberge and Drever, 1993). The primary advantage of mixed-flow reactors over batch systems is that they allow calculation of reaction rates directly from the steady-state mass balance condition of the effluent rather than fitting concentration vs. time data to an assumed rate law (Dove and Crerar, 1990). The steady-state mass balance condition for the uranium concentration of the effluent is

$$[U]_{ss}Q/M = [U]_{in}Q/M + R \quad (1)$$

where  $Q$  is the solution flux through the system ( $g_{soln} h^{-1}$ );  $M$  is the soil mass contained in the reaction vessel ( $g_{soil}$ );  $[U]_{ss}$  and  $[U]_{in}$  are the steady-state and influent uranium concentrations ( $\mu g U g^{-1}_{soln}$ ), and  $R$  is the uranium release rate ( $\mu g U g^{-1}_{soil} h^{-1}$ ).

The goals of this study are to determine the rate of uranium weathering in contaminated soil at FEMP and to estimate from this rate uranium mobility in FEMP soil. The uranium weathering rate is measured from a soil sample with a mixed-flow reactor.

In this study, all uranium phases are regarded collectively as a bulk soil property. Thus, it is not necessary to estimate surface areas of each U-bearing solid phase.

## **MATERIALS AND METHODS**

### **Soils**

The soil used in this study was a homogenized mixture of highly contaminated ( $>1200 \mu\text{g U g}^{-1}$ ) soil from the incinerator area at FEMP and uncontaminated background soil. The total soil uranium concentration of  $< 2\text{-mm}$  diluted soil (hereafter "soil") determined by microwave digestion/ICP-MS ( $465 \pm 12 \mu\text{g U g}^{-1}$ ) and by gamma spectroscopy ( $475 \pm 55 \mu\text{g U g}^{-1}$ ) were very close to the average soil U concentration ( $500 \mu\text{g U g}^{-1}$ ) of soils at the FEMP site (Francis et al., 1993).

The soil contained  $2.3 \pm 0.1 \%$   $\text{CaCO}_3$ ,  $1.7 \pm 0.1 \%$  organic carbon,  $16 \pm 1 \text{ g Fe}_{\text{DCB}} \text{ kg}^{-1}$ , and  $1.9 \pm 0.1 \text{ g Al}_{\text{DCB}} \text{ kg}^{-1}$  [DCB = extractable by dithionite-citrate-bicarbonate (Jackson et al., 1986)]. Mineralogical and grain size analyses were not performed specifically for this soil, but these characteristics are known for FEMP soils in general (Elless et al., 1993). Soils at FEMP are fine-grained ( $\geq 75\%$  silt and clay). Quartz is the most abundant mineral on a per-mass basis with lesser amounts of calcite, dolomite, and, in the clay fraction, kaolinite, illite, vermiculite, and chlorite. Carbonate gravels, which were added to the soils during construction and erosion-control activities prior to 1989, constitute 20% to 30% of the total mass of surface soils at the site.

## Measurement of Uranium Release Rates

The rate of uranium release from FEMP soils was determined using a mixed-flow reactor (Figure. 1). Depending on the experiment, 125-mL or 50-mL Erlenmeyer flasks were used as the reaction vessel. A variable-speed, multi-channel, peristaltic pump was used to maintain a constant solution flux in and out of the reaction vessel. The soil was kept in suspension with a magnetic stir bar. Because most mixed-flow reactors are designed for material with grain-sizes much coarser than FEMP soils, the major design challenge in these experiments was to keep the fine silt and clay in the reactor. The best approach was to use a stainless steel, 2- $\mu\text{m}$ -pore-size HPLC pump-inlet filter to filter the effluent solution. The filter was connected to the output line and submerged in the reaction vessel. Effluent solutions drawn through the filter contained no visible clay, though they could have contained  $<2\text{-}\mu\text{m}$  colloids.

Four uranium weathering experiments with different flow rates and soil-to-solution ratios (Table 3) were performed to determine the uranium weathering rate of the soil as a function of effluent uranium concentration. The feed solution for all experiments was a pH 7.4 solution of 1.67 mM  $\text{CaCl}_2$ . For each experiment, the desired soil and solution masses were added to the reaction vessel. The feed solution was then continuously pumped through the reactor. Experiments lasted 50 to 120 h. A 10 to 30-mL sample of effluent was collected at least once every four residence times. Steady-state uranium concentrations were determined by plotting the uranium concentration of

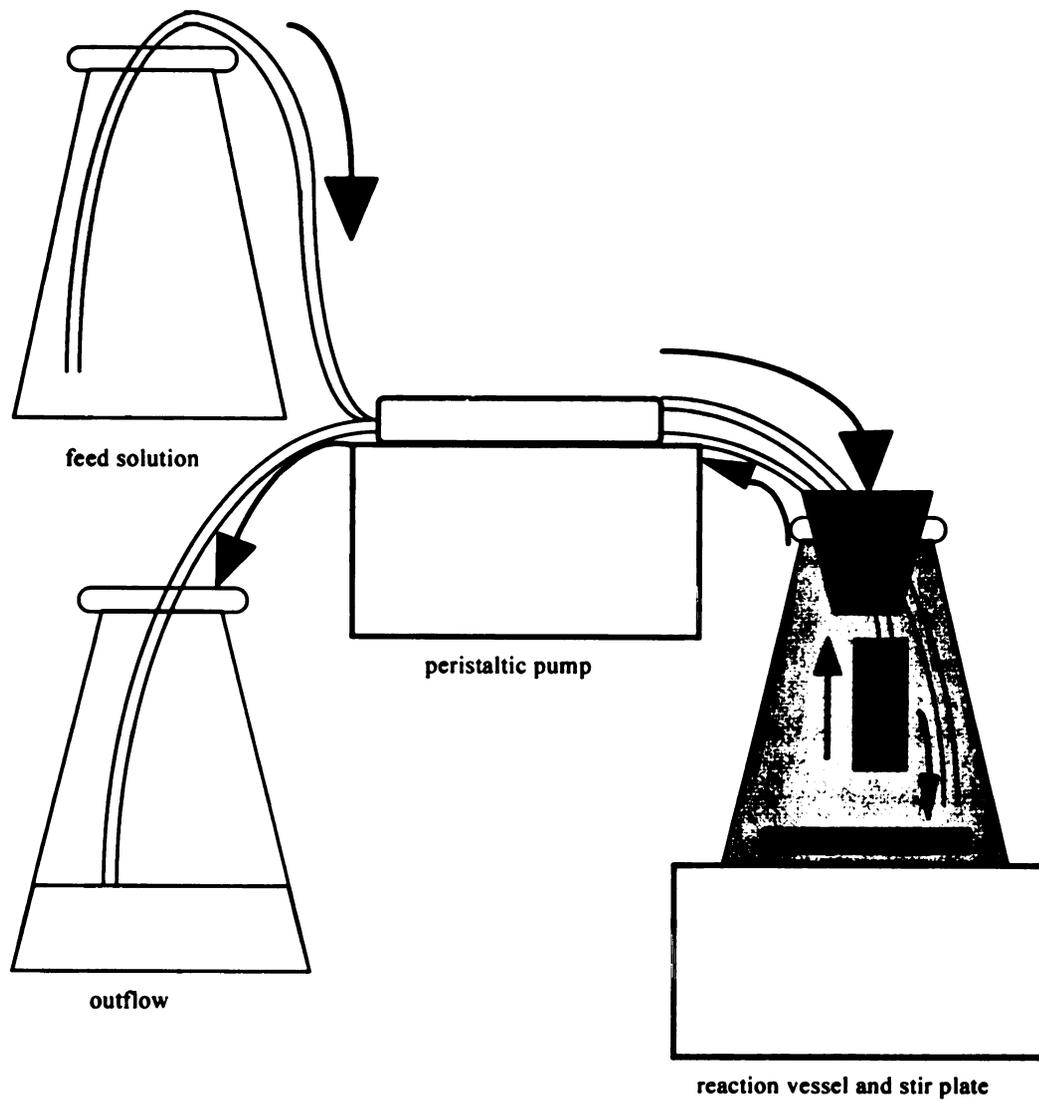


Figure 1. Schematic diagram of the mixed-flow reactor.

Table 3. Experimental conditions for mixed-flow reactor experiments.

Soil Mass g	Solution Mass g	Flow Rate g h <sup>-1</sup>	Q/M g <sub>soil</sub> h <sup>-1</sup> g <sup>-1</sup> soil	Residence Time h
10.40	104.00	9.46	0.90	11.0
4.94	47.32	6.60	1.32	7.2
5.00	50.41	10.30	2.06	4.9
4.90	100.01	23.23	4.73	4.3

the effluent as a function of residence time and averaging the U concentrations of samples from the plateau region of the graph (see Figure 2). Uranium release rates for each experiment were calculated from the steady-state uranium concentration using Eq. 1.

### Chemical Analyses

The uranium concentration of each effluent sample was determined by ICP-MS. For ICP-MS analysis of uranium, samples were diluted in 1% HNO<sub>3</sub> and spiked with <sup>209</sup>Bi as an internal standard. The pH, alkalinity and major cation concentrations of effluent solutions were also determined to verify that the solution composition was similar in all experiments. Solution pH was measured with a semi-micro glass combination electrode. Alkalinity was measured by acid titration. The concentrations of Ca and Mg in selected samples were measured by atomic absorption spectroscopy, Na by atomic emission spectroscopy, and Fe, Al, and Si by directly coupled plasma-atomic emission spectroscopy.

## RESULTS

### Steady-State Effluent Concentrations

The uranium concentration of the reactor effluents increased rapidly to a maximum value and then decreased to steady-state, with the greatest peak concentrations in the experiments with the lowest Q/M values (Figure 2) and longest residence times. For the lowest Q/M, about 3% of the total soil U was released during the 8 residence times prior to steady state, whereas much less than 1% of soil U was released prior to steady state at the highest Q/M. The steady-state concentrations ranged from 0.038 to 0.235  $\mu\text{g U g}^{-1}$  (Table 4) and generally decreased with Q/M ( $\text{g}_{\text{soln}} \text{h}^{-1} \text{g}^{-1}_{\text{soil}}$ ).

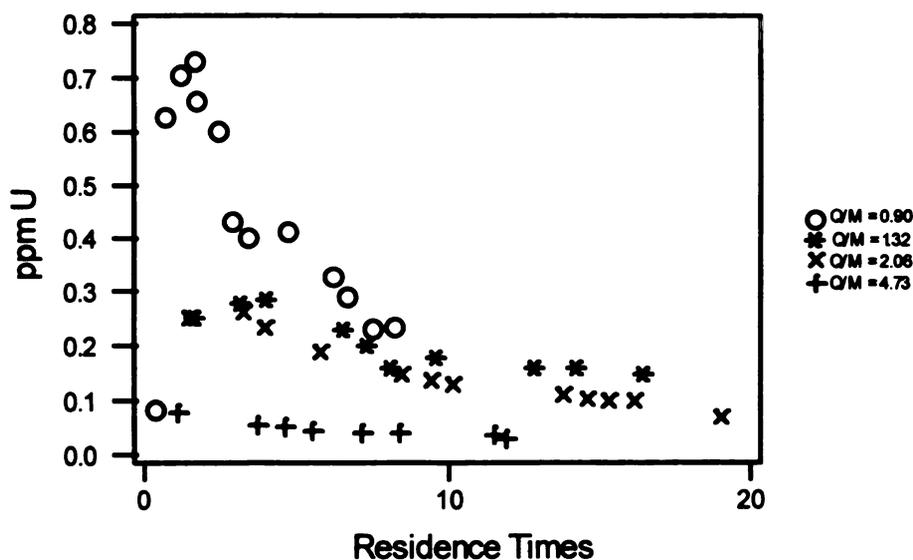
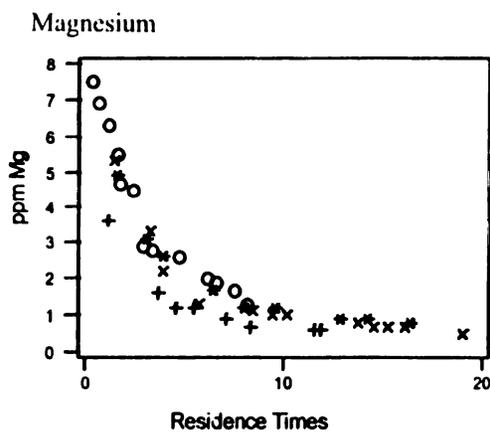
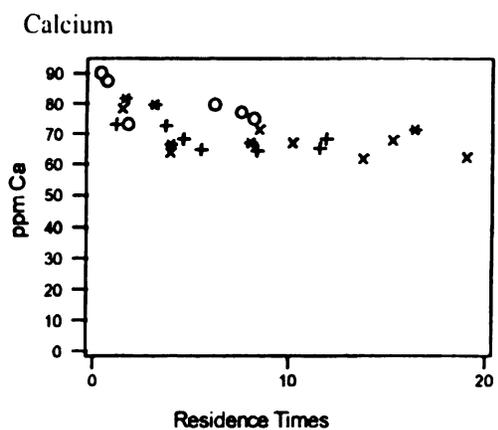


Figure 2. Time-dependence of effluent uranium concentrations.



X Q/M= 0.90  
 \* Q/M= 1.32  
 + Q/M= 2.06  
 O Q/M= 4.73

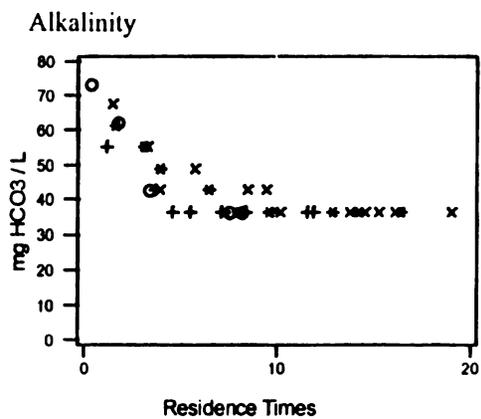


Figure 3. Time-dependence of effluent solution concentrations of Ca, alkalinity, and Mg.

The time-dependence of alkalinity, Ca, and Mg in effluent solutions is shown in Figure 3; steady-state concentrations are summarized in Table 4. Effluent pH in all four experiments attained a steady-state value of 7.8 within two residence times (not shown). Alkalinity attained steady-state ( $36 \text{ mg HCO}_3^- \text{ L}^{-1}$ ) prior to or at approximately the same time as uranium. The steady-state concentrations of Ca approached those of the input solution within a few residence times. The effluent Mg concentrations were initially high but then decreased to steady-state values (near 1 ppm Mg) after 8 to 10 residence times, similar to the time-dependence of U concentrations. Silica concentrations were measured only for the experiment with Q/M of 1.32, which had a steady-state silica concentration of approximately 12 ppm (Appendix 1). Iron and aluminum were below detection limits for all samples, as expected for the high solution pH.

Table 4. Steady-state effluent solution compositions, and calculated U release rate (R).

Q/M ( $\frac{\text{g}_{\text{soil}}}{\text{h}^{-1} \text{g}^{-1} \text{soil}}$ )	U (ppm)	pH	Alkalinity ( $\text{mgHCO}_3^- \text{ L}^{-1}$ )	Na (ppm)	Ca (ppm)	Mg (ppm)	R ( $\mu\text{g U h}^{-1} \text{g}^{-1} \text{soil}$ )
0.90	0.235	7.8	36.6	8	76.3	1.5	0.21
1.32	0.155	7.8	36.6	7	69	0.9	0.20
2.06	0.106	7.8	36.6	7.5	65.6	0.7	0.21
4.73	0.038	7.8	36.6	6.6	64.7	0.7	0.18

### Uranium Speciation

Geochemical modeling results indicate that in all experiments the steady-state effluent was undersaturated with respect to common uranyl oxides, hydroxides, and rutherfordite,  $\text{UO}_2\text{CO}_3$ , and that >99% of the total dissolved uranium was uranyl

carbonate complexes (Table 5). Modeling results for the experiment with  $Q/M = 1.32$  show that effluent was also undersaturated with respect to uranophane,  $\text{Ca}(\text{UO}_2)_2(\text{SiO}_3\text{OH})_2$ , ( $\text{Log IAP}/K_{sp} = -3.89$ ) and that aqueous concentration of U-silica complexes ( $2 \times 10^{-15} \text{ M kg}^{-1}$ ) was a minor portion of the total dissolved uranium. The saturation state of the effluent with respect to uranyl phosphate minerals was also investigated by assuming a total dissolved phosphate concentration of 0.1 ppm  $\text{PO}_4$ . In all cases the steady-state effluent was undersaturated with respect to autunite ( $\text{Log IAP}/K_{sp}$  decreased from -3.98 to -6.48 with increasing  $Q/M$ ), other autunite group minerals, and  $(\text{UO}_2)_2(\text{PO}_4)_2$ . Thus, it is likely that the solution residence times in these experiments were sufficiently rapid to ensure that the effluent remained undersaturated with respect to pure, crystalline uranium minerals in FEMP soil.

Table 5. Uranium speciation and saturation state of the steady-state effluent.

$Q/M$ ( $\frac{\text{g}_{\text{soil}} \text{ h}^{-1}}{\text{g}^{-1}_{\text{soil}}}$ )	% U bound to $\text{CO}_3$	$[\text{UO}_2^{2+}]_{\text{aq}}$ $\text{M kg}^{-1}$	log IAP/ $K_{sp}$ $\text{UO}_4(\text{OH})_2$	log IAP/ $K_{sp}$ gummite	log IAP/ $K_{sp}$ rutherfordite	log IAP/ $K_{sp}$ $\text{UO}_3$	log IAP/ $K_{sp}$ schoepite
0.90	99.8	$3.56 \times 10^{-12}$	-1.53	-6.39	-2.93	-3.70	-1.39
1.32	99.9	$2.26 \times 10^{-12}$	-1.72	-6.58	-3.16	-3.87	-1.58
2.06	99.9	$1.47 \times 10^{-12}$	-1.90	-6.76	-3.30	-4.08	-1.77
4.73	99.9	$5.53 \times 10^{-13}$	-2.21	-7.51	-3.74	-4.51	-2.21

Saturation indices were calculated with MINTEQA2 V. 3.10 (CREM, US EPA 1990) at the solution compositions listed in Table 4.

### Uranium Release Rates

Since the pH, alkalinity and the concentrations of the major cations were nearly identical in all experiments, the net uranium release rate of the soil for each experiment

can be compared as a function of  $[U]_{ss}$ , independent of these other solution parameters. Although the steady-state uranium concentration of these experiments varies by a factor of  $\sim 6$ , from 0.038 to 0.235  $\mu\text{g U g}^{-1}$ , the uranium release rates calculated with Eq. 1 for each Q/M are independent of uranium concentration (Table 2). The average net experimental uranium weathering rate in FEMP soil at the solution composition these experiments (R) is  $0.20 \pm .01 \mu\text{g U h}^{-1} \text{g}^{-1}_{\text{soil}}$ .

## DISCUSSION

The soil used in this study had a total uranium concentration near 470  $\mu\text{g U g}^{-1}_{\text{soil}}$ , which is comparable to the average soil U concentration at FEMP. Thus, although the soil was a mixture of contaminated soil and uncontaminated soil, the results of this study should be applicable to the FEMP site. The fraction of total soil uranium released during these experiments, 8%, 4%, 6%, 2% in order of decreasing Q/M, consisted of a labile fraction of the total soil uranium. The high uranium concentrations during the period prior to steady-state may be due mainly to rapid dissolution of very fine grained and/or amorphous uranium phases, possibly uranyl carbonates, oxides, and/or hydroxides. Once these dissolved, the effluent uranium concentration decreased to a steady state value controlled by a less reactive uranium phase(s). However, the uranium concentration decreased slightly after an initial steady state period in two of the experiments (Q/M of 2.06 and 4.73), indicating that U phase(s) controlling  $[U]_{ss}$  had dissolved completely.

Thus, it is likely that a new steady-state concentration corresponding to a more inert uranium mineral(s) would have been obtained for all Q/M values if the experiments had lasted longer. The uranium weathering rate measured here applies to a relatively small labile fraction of the total soil uranium rather than the bulk of U contamination.

Laboratory-derived weathering rates typically are one to three orders of magnitude greater than mineral weathering rates derived from elemental mass balances of watersheds because all the mineral surface area interacts with the influent solution in the mixed flow reactor, whereas only a small portion of the mineral surface area interacts directly with advective water in the soil profile (Velbel 1993, Swoboda-Colberge and Drever, 1993). Thus, to correct for partial soil-solution contact in the soil profile, it is reasonable to assume that 10% of the soil interacts directly with advective water. Thus, the uranium weathering rate in the field should be about 10% that measured in the mixed-flow reactor, or  $0.02 \mu\text{g U h}^{-1} \text{g}^{-1} \text{soil}$ .

Uranium contamination at FEMP mainly occurs in the upper 10-15 cm of the soil profile and leaches from this upper layer to the underlying soil horizons and ultimately to groundwater. Assuming saturated or nearly saturated conditions during main leaching events, the time-dependent uranium concentration of the soil solution during leaching,  $[\text{U}]_t$ , can be calculated from the following expression:

$$[\text{U}]_t = 0.1Rt\theta_g^{-1} \quad (2)$$

where the factor of 0.1 corrects the laboratory-derived weathering rate  $R$  for partial soil-solution contact in the field,  $[\text{U}]_t$  represents  $\mu\text{g U g}^{-1} \text{solution}$  after  $t$  hours of soil-solution

contact, and  $\theta_g$  is the gravimetric water content of the soil,  $\text{g H}_2\text{O g}^{-1}_{\text{soil}}$ . The concentrations predicted by Equation 2 are considered maximum estimates because it does not account for many of the complex chemical and physical processes occurring in the soil profile. Saturation water content for FEMP soil is about  $0.38 \text{ g H}_2\text{O g}^{-1}_{\text{soil}}$ . The hydraulic conductivity as a function of water content is unknown for these soils, but a range from  $0.5$  to  $2 \text{ cm hr}^{-1}$  is a reasonable estimate for near-saturated conditions. Assuming piston flow, the corresponding solution residence time in the upper 10 cm of the profile is 5 to 20 hr. Assuming that the solution pH and alkalinity the field are similar to those in the effluent solutions from the mixed-flow reactors, and that the U weathering rate is independent of U concentration as it was in the mixed-flow reactors, the uranium concentration in the soil solution will range from 0.26 to 1.1 ppm U, which corresponds to much less than 1% of soil U. If the factor of 0.1 is not used to correct for the fact that bulk water contacts only a fraction of the soil, then the U concentration of the soil solution will range from 2.6 to 11 ppm U. Although it is not known whether the experimental weathering rate is valid at total dissolved uranium concentrations greater than 0.235 ppm, the free  $[\text{UO}_2^{2+}]$  will remain very low at high total dissolved [U] because of the relatively high alkalinity of the soil solution (Table 4), so the U weathering rate is likely valid at high total dissolved [U]. The uranium concentrations predicted by this model are similar to uranium concentrations of shallow groundwater (1 to 12 ppm U) measured at FEMP, (Cunnane, 1995), even though the simple model used here greatly oversimplifies the complex chemical and hydraulic process that occur in the field.

## CONCLUSIONS

The experimental uranium weathering rate, which applies to a relatively small labile fraction of the total soil uranium, was  $0.20 \pm 0.01 \mu\text{g U h}^{-1} \text{g}^{-1}_{\text{soil}}$  and was independent of steady-state uranium concentration. Simple qualitative calculations based on this rate predict that the uranium concentrations of the soil solution are at ppm levels during leaching events. These results indicate FEMP soil contains a relatively labile and therefore mobile fraction of soil uranium that is capable of producing soil solutions with uranium concentrations far above those of typical natural waters, 0.01 ppm U (Langmuir, 1979).

## **APPENDIX**

## APPENDIX

## ANALYTICAL RESULTS

Table 6. Analytical results

$$Q/M = 0.90^{1,2}$$

TIME (H)	U (ppb)	Ca (ppm)	Mg (ppm)	Na (ppm)	K (ppm)	pH	Alkalinity (mgHCO <sub>3</sub> <sup>-</sup> L <sup>-1</sup> )
3.28	82	90.3	7.5	13.3	2.0	7.9	73.3
6.91	630	87.8	6.9	14.1	nd	7.9	nd
12.8	709	nd	6.3	nd	nd	7.9	nd
17.55	736	nd	5.5	nd	nd	7.8	nd
18.65	662	73.4	4.7	19.8	0.9	7.8	61.9
26.05	604	nd	4.5	nd	0.5	7.8	nd
31.58	432	nd	2.9	nd	nd	7.8	nd
36.70	404	nd	2.8	nd	nd	7.8	42.7
51.75	416	nd	2.6	nd	nd	7.8	nd
67.70	328	79.8	2	8.5	0.5	7.8	nd
72.70	291	nd	1.9	nd	nd	7.8	nd
82.46	233	77.3	1.7	8.5	nd	7.8	36.6
89.91	236	75.3	1.3	7.5	nd	7.8	36.6

<sup>1</sup> Iron and aluminum concentrations in all experiments were below the detection limit, ~ 8 ppb.

<sup>2</sup> nd = not determined

Table 6 (con'td).

Q/M = 1.32

TIME (H)	U (ppm)	Ca (ppm)	Mg (ppm)	Si (ppm)	Na (ppm)	K (ppm)	pH	Alkalinity (mgHCO <sub>3</sub> <sup>-</sup> L <sup>-1</sup> )
11.5	253	81.8	4.9	3.9	6.6	0.4	7.9	61.0
22.5	278	79.3	3.1	3.0	6.6	nd	7.9	54.9
28.48	284	66.5	2.6	19.0	7.5	0.9	7.8	48.8
46.63	227	nd	1.7	15.0	nd	nd	7.8	42.7
52.43	200	nd	nd	nd	nd	nd	7.8	36.6
58.10	159	67.0	1.2	19.9	7.5	0.2	7.8	36.6
69.06	178	nd	1.2	nd	7.5	nd	7.8	36.6
92.55	157	nd	0.9	12.7	6.6	0.9	7.8	36.6
102.4	157	nd	0.9	12.8	6.6	nd	7.8	36.6
118.2	148	71.4	0.8	12.0	nd	0.9	7.8	36.6

Table 6 (con'td).

Q/M = 2.06

TIME (H)	U (ppb)	Ca (ppm)	Mg (ppm)	Na (ppm)	K (ppm)	pH	Alkalinity (mgHCO <sub>3</sub> <sup>-</sup> L <sup>-1</sup> )
7.25	250	78.3	5.3	7.5	nd	7.8	67.1
16.00	263	nd	3.3	nd	0.2	7.8	54.9
19.31	231	63.8	2.2	7.5	1.4	7.8	42.7
28.28	187	nd	1.3	nd	nd	7.8	48.8
41.50	148	71.4	1.14	5.6	2.1	7.8	42.7
46.43	137	nd	1.0	nd	nd	7.8	42.7
49.86	130	67.1	1.0	6.5	2.4	7.8	36.6
67.59	110	61.9	0.8	7.5	2.9	7.8	36.6
71.44	103	nd	0.7	nd	nd	7.8	36.6
74.98	100	68.0	0.7	7.5	nd	7.8	36.6
79.12	98	nd	0.7	nd	nd	7.8	36.6
93.31	67	62.4	0.5	6.5	nd	7.8	36.6

Table 6 (con'td).

Q/M = 4.73

TIME (H)	U (ppb)	Ca (ppm)	Mg (ppm)	Na (ppm)	K (ppm)	pH	Alkalinity (mgHCO <sub>3</sub> <sup>-</sup> L <sup>-1</sup> )
4.77	76	72.9	3.6	nd	2.0	7.9	54.9
15.88	55	72.4	1.6	6.6	1.4	7.9	42.7
19.72	52	68.1	1.2	6.6	nd	7.8	36.6
23.63	41	64.8	1.2	6.6	0.6	7.8	36.6
30.67	39	nd	0.9	nd	0.5	7.8	36.6
36.05	39	64.2	0.7	6.6	nd	7.8	36.6
49.60	34	65.2	0.6	6.6	0.3	7.8	36.6
51.3	28	68.1	0.6	6.6	nd	7.8	36.6

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