SUPPORTING INFORMATION

for

Enrichment of Cesium and Rubidium in Weathered Micaceous Materials at the Savannah River Site, South Carolina

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1. SOIL AND SEDIMENT SAMPLES

1.1 Soil Samples. The soil samples are from three cores taken by hand auger from locations shown in Figure S1, which also shows the location of the BGO-3A borehole. The core from the location identified as 'Fuquay,' hereinafter identified as 'FQ,' is from Fuquay soil developed on the Miocene or Oligocene Upland Unit. FQTRS is a core from Fuquay soil formed on the upper Eocene Tobacco Road Sand, and FQQAL is a core from soil formed on Quaternary alluvium. The depth range of each core segment and the corresponding mean depth are given in Table S1.

1.2 Sediment Samples. Seven sediment samples were taken from the large-diameter core from the BGO-3A borehole[S1] (Figure S1) at the depths shown in Table S2, which also shows the minerals in the clay fraction of each sample as determined by conventional X-ray diffractometry.[S2]

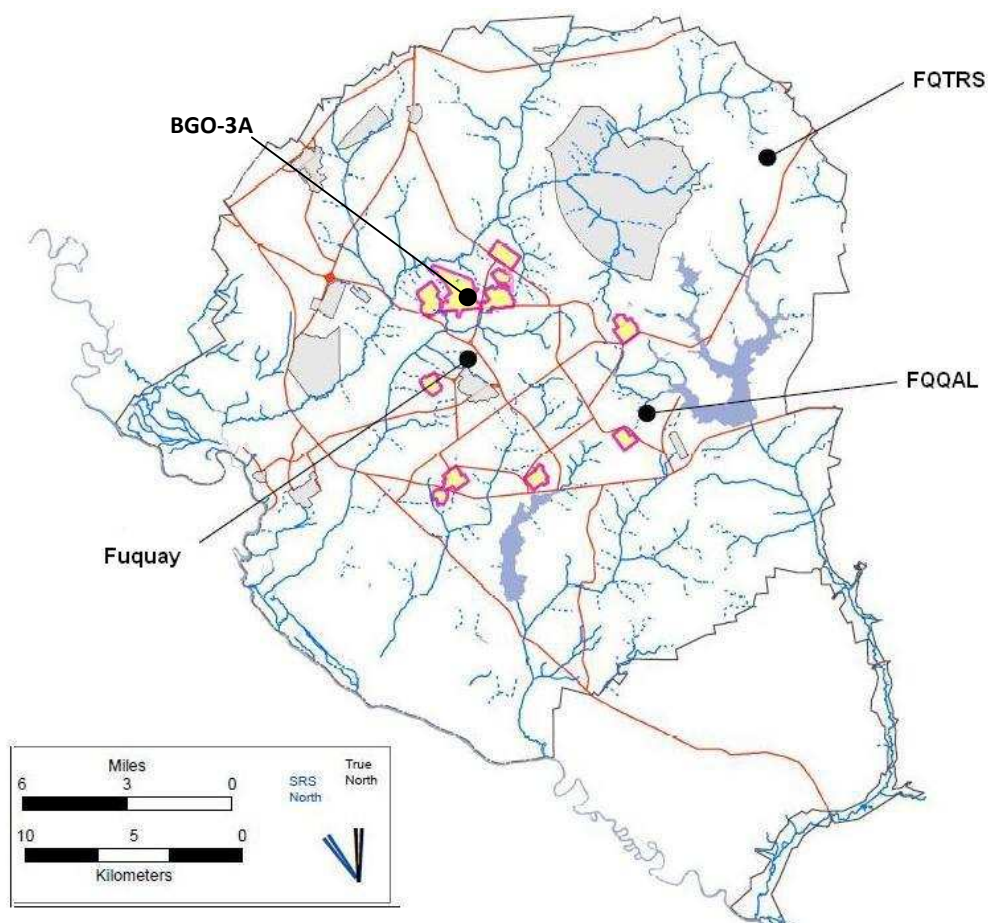**Figure S1**. Map of the Savannah River Site showing the locations of the three soil cores and the BGO-3A borehole. Areas of yellow with pink outline are operations areas where nuclear waste was produced. Non-nuclear operations areas are in gray, major roads in are in red, and streams are in blue. Map altered from original by Laura Bagwell[S3] of Savannah River National Laboratory.

Table S1. Depth Ranges and Textures of the Soil Samples

Core	Depth range (cm)	Mean depth (cm)	Sand	Silt	Clay	Soil texture
				(Percent by mass)		
FQ	0-20	10	87.2	5.8	7.0	Loamy sand
FQ	20-36	28	91.3	5.6	3.2	Sand
FQ	36-56	46	91.2	6.1	2.7	Sand
FQ	56-76	66	88	6.8	5.3	Sand
FQ	76-97	86	90.6	5.5	3.9	Sand
FQ	97-117	107	89.5	7.1	3.4	Sand
FQ	117-137	127	82.3	5.8	11.9	Loamy sand
FQ	137-157	147	65.6	4.6	29.8	Sandy clay loam
FQ	157-178	168	50.3	8.3	41.5	Sandy clay
FQ	178-203	191	41.7	13.3	45.1	Clay
FQTRS	0-15	8	92.4	3.9	3.7	Sand
FQTRS	15-30	23	91.3	5.3	3.4	Sand
FQTRS	30-46	39	91.6	5.9	2.5	Sand
FQTRS	46-61	53	87.1	7.1	5.9	Loamy sand
FQTRS	61-76	69	80.2	4.2	15.6	Sandy loam
FQTRS	76-89	83	74.1	3.3	22.6	Sandy clay loam
FQQAL	0-15	8	85.1	7.3	7.6	Loamy sand
FQQAL	15-38	22	80.2	13.6	6.2	Loamy sand
FQQAL	28-38	33	58.8	3.6	37.6	Sandy clay
FQQAL	38-46	42	41.6	12.2	46.2	Clay
FQQAL	46-61	53	28.7	8.2	63.2	Clay
FQQAL	61-76	69	19.2	20.3	60.4	Clay
FQQAL	76-84	80	27.7	7.4	65	Clay
FQQAL	84-91	88	51.4	7.1	41.6	Sandy clay
FQQAL	91-104	98	60.5	3	36.5	Sandy clay

Table S2. Depths and Clay Minerals of Sediment Samples from the BGO-3A Core

Depth (m)	Major mineral(s) in clay	Minor mineral(s) in clay
8.5	kaolinite	quartz
14.0	kaolinite	quartz
23.2	kaolinite	quartz, illite or mica
28.7	kaolinite, illite or mica, quartz	smectite
29.7	smectite, kaolinite, quartz	illite or mica
30.0	smectite, kaolinite, quartz	illite or mica
30.7	smectite, kaolinite, quartz	illite or mica

1.3 Soil Texture Analysis. About 25 g of a <2 mm portion of each soil sample was treated to remove organic matter, iron oxides, and carbonate cements by well-known methods[S4] yielding a Na-saturated sample. These treated samples were then separated into sand (2–0.05 mm), coarse silt (50-20 µm), fine silt (20-2 µm), and clay (<2 µm) fractions by standard timed settling techniques. The percentage of each size fraction except the clay fraction, relative to the mass of <2 mm starting material, was calculated from its mass after drying (Table S1). A maximum value for the mass of the clay fraction was calculated as the difference between the mass of the starting material (25 g) and the sum of the masses of the fractions coarser than clay and used to calculate the clay contents shown in Table S1. Because the uppermost soil segments probably contained more than 1% organic matter,[S5] the true clay contents of these segments likely are significantly less than the values shown for them in Table S1.

The amounts of fine silt and coarse silt were combined for texture classification according the *Glossary of Soil Science Terms*, Soil Science Society of America.[S6] The soil texture class to which each sample belongs is named in Table S1 and depicted graphically in Figure S2.
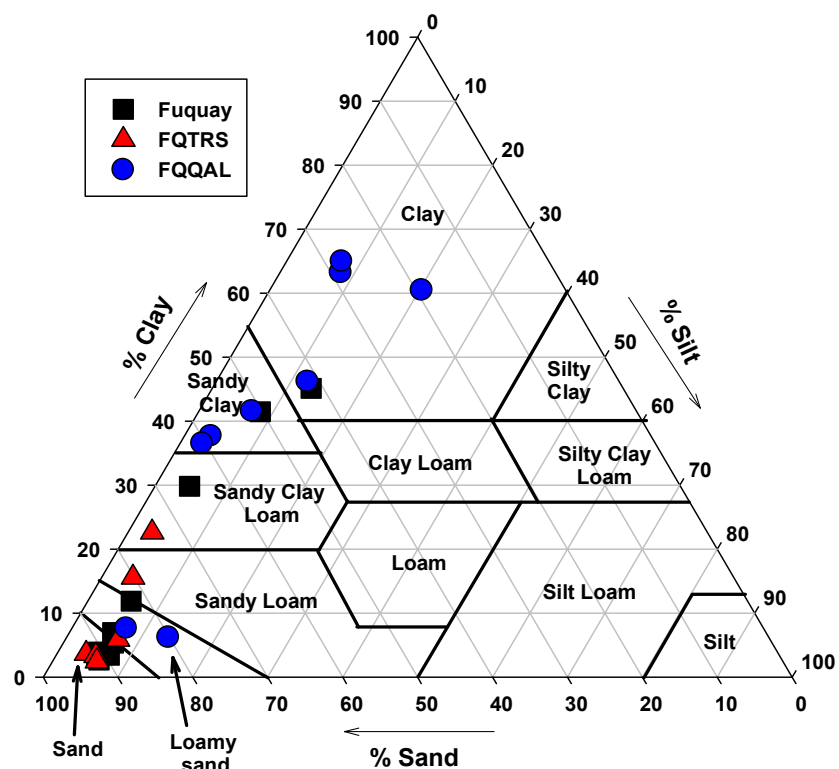
Figure S2. Ternary diagram of soil textures for the FQ (black squares), FQTRS (red triangles), and FQQAL (blue circles) core segments. Percentages are by mass. Soil textures are according to the *Glossary of Soil Science Terms*, Soil Science Society of America.[S6]

1.4 Quantitative Determination of Crystalline Phases in Clay Fractions. High intensity synchrotron radiation was used to obtain high resolution X-ray diffractograms necessary to quantify the crystalline phases in the clay fraction.[S2] Table S3 lists the results of Rietveld phase quantification of the crystalline phases in the clay fraction of each core segment and the corresponding estimate of the cation exchange capacity (CEC) of the fine fraction (<50 μm). The compositions of the phases quantified are $(Fe,Mg,Al)_6(Si,Al)_4O_{10}(OH)_8$ for HIV, $K(Al,Mg,Fe)_2(Si,Al)_4O_{10}[(OH)_2 \cdot (H_2O)]$ for illite, $Al_2Si_2O_5(OH)_4$ for kaolinite, $Al(OH)_3$ for gibbsite and SiO_2 for quartz. χ^2 represent the goodness of fit on three-point smoothed patterns and corresponds to

$$\chi^2 = [\Sigma_i \, (I_{obs} - I_{calc})_i^2 \, / \, \sigma^2(I_{obs})_i] \, / \, (n - p),$$

with I the intensity, $\sigma(I_{obs})$ the estimated error of the measure (fixed to 10% of the counts), n the number of points used for simulation and p the number of parameters estimated. (The procedure used to estimate CEC for the fine fractions is described under 'Fine Fractions,' below.)

Table S3. Rietveld Phase Quantification of Minerals and Estimated CEC Values

Core	Mean depth (cm)	HIV (%)	Illite (%)	Kaolinite (%)	Gibbsite (%)	Quartz (%)	χ^2	Estimated CEC (cmol$_c$ kg^{-1})
FQ	10	26.0	0.0	38.9	7.5	27.5	4.2	8.0
FQ	28	16.4	0.0	53.0	9.9	20.8	17.0	3.8
FQ	46	24.7	0.0	32.6	9.7	33.0	7.2	4.2
FQ	66	19.0	0.0	28.6	3.6	48.9	6.9	4.7
FQ	86	16.2	0.0	35.5	6.9	41.4	8.6	3.9
FQ	107	19.1	0.0	40.4	7.0	33.5	9.0	3.6
FQ	127	12.7	0.0	39.4	10.4	37.5	12.3	5.3
FQ	147	2.1	0.0	84.7	10.4	2.8	9.1	3.8
FQ	168	2.3	0.0	84.5	11.1	2.1	24.2	3.8
FQ	191	1.3	0.0	84.8	12.8	1.0	19.8	3.1
FQTRS	8	19.1	7.3	37.3	4.7	31.6	2.4	6.1
FQTRS	23	7.7	6.7	42.6	11.5	31.5	3.2	2.7
FQTRS	39	12.6	8.7	41.1	9.5	28.1	4.4	2.9
FQTRS	53	8.5	9.2	35.8	12.6	33.9	4.2	3.4
FQTRS	69	14.7	13.2	38.6	10.5	23.0	1.3	9.1
FQTRS	83	20.2	11.5	42.2	18.3	7.8	6.4	12.3
FQQAL	8	6.1	22.1	47.2	0.0	24.5	5.2	4.8
FQQAL	22	5.3	27.3	34.8	0.0	32.6	4.2	3.0
FQQAL	33	9.2	13.4	39.2	0.0	38.2	1.9	8.1
FQQAL	42	12.0	17.1	69.1	0.0	1.8	2.3	9.6
FQQAL	53	12.7	16.1	50.6	0.0	20.6	7.8	10.3
FQQAL	69	12.9	12.7	52.4	0.0	21.9	7.0	8.3
FQQAL	80	6.7	18.5	68.9	0.0	5.9	2.2	8.8
FQQAL	88	14.0	13.6	64.9	0.0	7.6	3.4	10.5
FQQAL	98	1.9	13.2	72.7	0.0	12.0	4.0	6.0

1.5 Fine Fractions. Fine fractions of the soil and sediment samples were prepared by sieving the air-dried soil (after removal of material >2 mm) and sediment samples on a 63 μm polyester monofilament screen manually until continued shaking was no longer effective in causing fine material to pass the sieve. The CEC of each soil fine fraction was estimated from the mass fraction of clay in the fine fraction (calculated as the ratio of the clay percentage to the sum of the clay and silt percentages, both from Table S1) times a weighted sum of the CECs of the phyllosilicate minerals in the clay fraction (weighted by their abundances in the clay fraction from Table S3). The mineral CEC values used were 50 cmol$_c$/kg for HIV,[S7] 25 cmol$_c$/kg for illite,[S8] and 4 cmol$_c$/kg for kaolinite.[S9,S10]

2. CHEMICAL PROCEDURES

2.1 Reagents and Laboratory Ware. Trace metal grade reagents and high purity de-ionized water were used throughout except as otherwise noted. Fluorocarbon vials were used for all reactions with hot acid, and new polystyrene or polyethylene containers were used otherwise. The fluorocarbon vials were cleaned in a nitric acid bath at ~80°C before each use. The new containers did not require cleaning. Evaporations were done from fluorocarbon vials under flowing clean air.

2.2 Acid Extractions. Portions of the soil and sediment fine fractions were treated for 3 h with hot, strong nitric acid solution. These extractions, under conditions similar to those of US EPA Method 3050B,[S11] were intended to remove most of the alkali metal atoms not bound in silicate structures. To a decigram of each fine fraction in a fluorocarbon vial, 10 cm^3 aqueous HNO_3 solution (~10 mol dm^{-3}) was added and the vial was closed and shaken vigorously. The vials were then heated to about 100°C with frequent manual shaking, every 5 minutes for the first half hour, and then every 15 minutes for the next 2.5 hours. After 3 hours of extraction, vials were centrifuged at 2000 revolutions per minute (rpm) for 10 minutes. The supernatant liquids were poured into pre-weighed, acid-cleaned fluorocarbon vials and about 5 cm^3 of high purity deionized water was added to each of the extraction residues. The vials were then closed, shaken, and centrifuged for an additional 10 minutes at 2000 rpm, and each supernatant rinse solution was added to the corresponding original supernatant liquid. The mass of each of the combined liquids was determined by weighing the liquid in the pre-weighed vial. [Another set of decigram portions of the soil fine fractions (but including no sediment fine fractions) was treated more moderately with dilute HNO_3 (~2 mol dm^{-3}) at 80°C for 3 hours in a procedure that was the same as the one just described except for the smaller acid concentration and lower temperature. Results of the more moderate acid extraction are not being reported in the present study, except that the results from one sample

(FQTRS, 39 cm) are used below to support our evaluation of the accuracy of the total of each element from the corresponding strong acid extraction.]

2.3 Digestion of Acid-Extraction Residues. The solid residues remaining after the acid extractions (except those from BGO-3A sediment samples from 14 m depth and below) were rinsed and then digested by gentle heating in closed fluorocarbon vials with a 5:1 (molar) mixture of concentrated HF and HNO_3. After evaporation of SiF_4 and excess acid, these digestates were taken up in dilute HNO_3 (10 cm^3, 0.32 mol dm^{-3}).

2.4 Preparation of Test Solutions for Instrumental Determinations. Two drops of a 'spike' solution containing highly enriched (to 99.85%) ^{85}Rb were added to each combined extract and rinse solution, and one drop of the same solution was added to each solution containing the digested extraction residues. In each case the mass of added spike solution was determined by weighing the spike-solution container (a polyethylene dropping bottle) before and after the solution was added. The amount of added ^{85}Rb was more than the amount of natural ^{85}Rb in all of the extract solutions. The amount of added ^{85}Rb was less than the amount of natural ^{85}Rb in most of the solutions containing digested extraction residue, but in no case less than 44% of the amount of natural ^{85}Rb. Eirik Krogstad, then of Georgia State University, had calibrated the mass fraction of ^{85}Rb in the added solution by duplicate analyses of National Institute of Standards and Technology (NIST) standard reference material (SRM) 612.

Each solution with added ^{85}Rb was divided into two portions. One portion was used for trace element determinations by inductively coupled plasma mass spectrometry (ICP-MS), and the remainder was used for potassium determination by flame atomic absorption spectrophotometry (FAAS). For ICP-MS, a 4 cm^3 portion of each combined extract and rinse solution was heated until dry and the residue was taken up in dilute HNO_3 (0.32 mol dm^{-3}). A 5 cm^3 portion of each solution containing the digested extraction residues was used for ICP-

MS. These did not require further processing because the digestates already had been taken up in 0.32 mol dm^{-3} HNO$_3$. The solution portions to be used for K determination by FAAS were heated to dryness, and the remaining solids were taken up in a weighed amount of an acidic CsCl solution (0.1 mol kg^{-1} HNO$_3$, 0.01 mol kg^{-1} CsCl, both reagent grade). The Cs served as an ionization suppressant in the flame.

2.5 Instrumental Determinations. A Thermo Scientific Element 2 high resolution, sector magnetic field, inductively coupled plasma mass spectrometer was used for determination of Rb, Cs, Sr, and Ba. Rb was determined by stable isotope dilution from signals at mass to charge ratios (*m/z*) of 85 and 87 after correction of the latter for the contribution of ^{87}Sr, which was assumed to be 8.48% (the molar ratio of ^{87}Sr to ^{88}Sr in common Sr) of the beam intensity of ^{88}Sr at *m/z* = 88. Cs, Sr, and Ba were determined by comparing the signals at *m/z* = 133, 88, and 137, respectively, to the signal at *m/z* = 85, using sensitivities for ^{85}Rb, ^{88}Sr, ^{133}Cs, and ^{137}Ba determined from bracketing runs of a solution containing 1.00 µg kg^{-1} of each element (of natural isotopic composition). Corrections for mass spectrometer background signals were applied to all runs but were significant only for the bracketing runs.

K was determined with a Perkin-Elmer atomic absorption spectrophotometer, after test solutions had been diluted to <2.0 mg kg^{-1} K with the acidic CsCl solution, by comparison of readings (absorbance at 766.5 nm) for test solutions with those for reference solutions of known K concentration prepared from standard KCl (NIST SRM 999).

2.6 Commercial Elemental Analyses. The mass fractions of Cs, Rb, Ba, Sr, and K in the fine fractions were determined by Activation Laboratories Ltd., "Actlabs" (Ancaster, Ontario) by instrumental analysis of solutions obtained by a four-acid digestion (HF, HNO$_3$, HClO$_4$, HCl). The specific procedure used is identified by the company as Code UT-6 Total Digestion ICP & ICP/MS.[S12]

2.7 Cesium Extraction by Cation Exchange. The amounts of Cs removed from 1 g portions of bulk soil (<2 mm) by ion exchange with ammonium acetate (1 mol dm^{-3}) at room temperature were determined by ICP-MS at the Savannah River National Laboratory.[S2] For comparison, the total Cs in the bulk soil was calculated, by assuming that the sand-sized material in the soil contains no Cs, as the mass fraction of Cs in the fine fraction (Table S8, below) times the mass fraction of fine material (clay- and silt-sized material, Table S1).

3. PRECISION AND ACCURACY OF CHEMICAL DETERMINATIONS

3.1 Procedural Blank Runs. Amounts of Rb, Cs, Ba, and Sr found in procedural blank runs were negligible (Table S4). The 0.8 μg of K found in the first procedural blank run for acid extraction (Table S4) was 4% of the smallest amount of K determined for an acid extract of soil but was only 2% of the average amount of K in acid extracts. (The solution from that run was not spiked with the enriched [85]Rb, so no results were determined for the trace elements.) The 0.8 μg was near the limit of detection, and no K was detected in a second, spiked blank run on the extraction procedure. No blank corrections were applied.

Table S4. Results of Procedural Blank Runs

Procedure	K		Rb		Cs		Sr		Ba	
	(μg)	(%)	(ng)	(%)	(ng)	(%)	(ng)	(%)	(ng)	(%)
Acid extraction	0.8	3.5								
Acid extraction	0.0	0.0	3.8	0.4	−0.2	−0.1	1.9	0.6	1.0	0.1
Residue digestion			3.2	0.2	0.1	0.1	2.5	0.1	6.0	0.1

Note. The amount of each element determined in a procedural blank run is expressed as a mass value and as a percentage of the smallest amount of the element found when the procedure was applied to the soil samples.

3.2 Instrumental Precision. Test solutions prepared from residues of the acid extraction of fine fractions from eight of the ten FQ core segments were run by ICP-MS in duplicate. The relative differences in trace element concentrations from these runs are listed in Table S5. Only a few other test solutions were run by ICP-MS a second time, and these were run a second time because the first runs had anomalous characteristics (unusually low ion beam intensities or variable internal precision values). The results from such runs are not included in Table S5. Except for the unexplained large differences obtained for the 46 cm core

segment, the results indicate acceptable precision (considered to be ±10% or better for this study) for Rb, Cs, and Ba. The large relative differences for Sr are unexplained.

Table S5. Relative Differences Between Values from Duplicate ICP-MS Runs (First – Second)

Mean depth (cm)	Rb (%)	Cs (%)	Sr (%)	Ba (%)
10	−6.0	3.3	20	−4.6
28	−3.6	5.6	23	−0.5
46	−46.9	−7.8	−28	−48.6
66				
86	−1.6	7.4	31	8.6
107	−4.1	0.8	28	6.9
127	−1.7	7.7	35	8.9
147				
168	−4.9	−0.8	32	−7.3
191	−5.2	8.7	39	15.1
Mean	−9.2	3.1	22	−2.7
Standard deviation	15.3	5.6	21	20.0
Median	−4.5	4.5	30	3.2

Notes. The test solutions were from acid extraction residues of FQ fine fractions. The solutions from 66 cm and 147 cm mean depth were run only once.

No duplicate potassium determinations of test solutions were done in this study. Instrumental internal precision was noted by tabulating the relative standard deviation of the K concentrations calculated from two sets of bracketed absorbance readings that were taken for a single K determination. In no case was this value greater than 5%.

3.3 Precision of Acid Extractions. The strong acid extraction of fine fractions from the FQTRS and FQQAL cores was done twice. The relative differences in the specific amounts of the elements extracted are listed in Table S6. Given the precision of the ICP-MS

determinations suggested by the data of Table S5, the data of Table S6 show no evidence that different amounts of these elements were extracted from the FQQAL samples in the two different sets of strong acid extractions, but somewhat more K and Cs (and perhaps Sr) were extracted from FQTRS samples in the first set.

Table S6. Differences Between Values from Duplicate Strong Acid Extractions (First −
Second)

Core	Mean depth (cm)	K (%)	Rb (%)	Cs (%)	Sr (%)	Ba (%)
FQTRS	8	6.5	−7.1	11.2	33	12.4
FQTRS	23	11.6	3.1	25.2	27	9.6
FQTRS	38	11.5	−5.1	17.5	25	3.5
FQTRS	53	23.6	−2.9	23.2	33	5.1
FQTRS	69	11.2	−5.0	8.3	26	−5.6
FQTRS	83	12.1	−4.5	7.8	30	−4.9
FQQAL	8	7.4	−2.1	7.9	17	2.0
FQQAL	22	−12.9	−8.1	11.4	13	−4.2
FQQAL	33	0.4	−4.7	−1.6	10	−3.6
FQQAL	42	2.9	−1.2	−1.2	13	−2.6
FQQAL	53	5.6	−2.6	−1.9	14	−3.6
FQQAL	69	5.3	−6.0	−3.6	14	−3.6
FQQAL	80	5.5	−6.4	−5.5	16	−4.1
FQQAL	88	11.1	−2.5	0.4	26	2.0
FQQAL	98	9.8	1.2	3.2	30	5.3
Mean		7.4	−3.6	6.8	22	0.5
Standard deviation		7.8	3.0	9.6	8	5.7
Median		7.4	-4.5	7.8	25	−2.6

3.4 Accuracy of the Chemical Determinations. Determination of K, Rb, Cs, Sr, and Ba in the U. S. Geological Survey geochemical reference material SCo-1[S13] by a procedure essentially like that described above for the residues of acid extractions (but with considerably greater effort to get precise results for K) produced results in good agreement with the certificate values for the reference material (Table S7).

Table S7. Results from Geochemical Reference Sample SCo-1

	K (g kg^{-1})	**Rb** (mg kg^{-1})	**Cs** (mg kg^{-1})	**Sr** (mg kg^{-1})	**Ba** (mg kg^{-1})
This study	23.0	107	7.4	163	557
Certificate	23.0 ± 0.7	110 ± 4	7.8 ± 0.7	170 ± 16	570 ± 30

Note: The value for K$_2$O on the certificate[S13] has been converted to the equivalent value for K.

The good agreement with the certificate values for the trace elements shows that the [85]Rb-enriched spike solution had been accurately calibrated. The accuracy of the element determinations in the present study is not limited by systematic error. Because logarithmic scales (base 10) are used in presentation of nearly all of the data in the accompanying manuscript, uncertainties of about 10% for most of the elements and of about 20% for Sr are of no consequence to the interpretations of this work.

The Actlabs results were reported with results from several different geochemical reference materials for quality assurance. These results are in the quality control section of a report identified as A13-07873.[S14] Nearly all results for Cs, Sr, and Ba were within 10% of the certificate values. Most of the K and Rb determinations were within about 10% of the certificate values, but there are large deviations from the certificate values for several of the reference materials in several cases each.

4. RESULTS OF CHEMICAL DETERMINATIONS

4.1 Large-Ion Alkali and Alkaline-Earth Elements in Fine Fractions. The values for the mass fractions of K, Rb, Cs, Sr, and Ba in the soil fine fractions (<63 μm) used to calculate the data plotted in Figures 3 and 5 of the accompanying manuscript are those determined by Actlabs (Table S8) with two exceptions: (1) The fine fraction of the deepest FQ segment (191 cm) was not submitted for analysis by Actlabs, (2) The values from Actlabs for the 39 cm segment of FQTRS were judged to be systematically too large. In each of those two cases, the sum of values from the acid extract (Table S9) and the corresponding residue (Table S10) was used as the total of each element in the fine fraction.

The judgment that the values from Actlabs for the 39 cm segment of FQTRS were systematically too large is based on the observation that the Actlabs' value for each element was much larger than the total of the element from the strong acid extract and corresponding residue (Tables S9 and S10). The decision that the totals determined in-house are the better ones is based on good agreement (relative differences 8% or less) of those totals with corresponding totals from a more moderate acid extraction, which are not otherwise reported in the present study.

Table S8. Mass Fractions of K, Rb, Cs, Sr, and Ba in Soil Fine Fractions (<63 µm).

Core	Mean depth (cm)	K (g kg^{-1})	Rb (mg kg^{-1})	Cs (mg kg^{-1})	Sr (mg kg^{-1})	Ba (mg kg^{-1})
FQ	10	3.5	26	3.4	28	150
FQ	28	5.1	41	5.0	31	202
FQ	46	5.6	43	5.2	31	214
FQ	66	5.9	46	5.4	34	220
FQ	86	6.5	55	6.5	37	232
FQ	107	6.0	58	7.4	36	224
FQ	127	6.1	81	11.6	44	268
FQ	147	4.6	63	10.6	42	217
FQ	168	3.6	42	7.4	34	141
FQ	191					
FQTRS	8	4.3	42	4.8	28	174
FQTRS	23	4.6	38	3.9	28	180
FQTRS	39	6.0	49	4.7	42	247
FQTRS	53	5.4	46	4.5	32	206
FQTRS	69	5.8	85	10.1	48	302
FQTRS	83	5.0	71	9.4	46	259
FQQAL	8	20.6	66	1.4	56	566
FQQAL	22	20.9	76	2.2	70	688
FQQAL	33	22.4	106	3.9	61	622
FQQAL	42	13.0	103	5.7	52	468
FQQAL	53	13.1	90	4.9	49	416
FQQAL	69	17.4	92	4.0	52	442
FQQAL	80	13.5	81	4.2	44	378
FQQAL	88	8.6	67	4.9	36	288
FQQAL	98	9.6	69	4.8	37	306
UCC		23.2	84	4.9	320	628

Note. The values are those determined by Actlabs except that those for the upper continental crust (UCC) are from Rudnick and Gao.[S15]

4.2 Results of Acid Extractions. The masses of elements extracted by hot, strong acid from soil and sediment fine fractions, each divided by the mass of fine fraction subjected to extraction, are listed in Table S9. The corresponding amount remaining in the solid residues of the extractions are listed in Table S10. The values in Table S10 are not mass fractions of the elements in the residues. Rather, each value is the mass of an element in a residue divided by the original mass of fine material (<62 μm) subjected to the strong acid extraction. The values for the third segment of the FQ core (46 cm) were not measured directly because material was accidentally spilled; these values are based on the total for each element determined by Actlabs less the amount extracted by the strong acid treatment.

4.3 Elements Removed by Cation Exchange with Ammonium Acetate. In the second column under Cs of Table S9 are values for the mass of Cs extracted from 1 g portions of bulk soil (<2 mm) by ion exchange with ammonium acetate, divided by the mass of soil subjected to ion exchange.

Table S9. Elements Extracted by Acid and Exchanged by Ammonium Acetate

Core	Mean depth	K	Rb	Cs		Sr (mg kg⁻¹)	Ba (mg kg⁻¹)
	(cm)	(g kg⁻¹)	(mg kg⁻¹)	(mg kg⁻¹)			
FQ	10	0.24	11	2.6	0.022	7.0	44
FQ	28	0.19	17	4.2	0.026	3.4	50
FQ	46	0.28	19	4.1	0.024	4.0	49
FQ	66	0.30	20	4.3	0.032	4.3	47
FQ	86	0.24	24	4.9	0.028	3.4	48
FQ	107	0.34	26	5.5	0.037	4.7	57
FQ	127	0.60	40	8.3	0.066	6.5	101
FQ	147	0.83	37	7.8	0.109	9.8	101
FQ	168	0.82	24	5.5	0.136	8.6	62
FQ	191	0.32	11	3.0	0.133	4.3	29
FQTRS	8	0.34	21	3.8	0.021	3.9	51
FQTRS	23	0.27	19	3.2	0.019	3.7	54
FQTRS	39	0.26	16	2.5	0.022	3.2	40
FQTRS	53	0.40	21	3.4	0.041	3.9	54
FQTRS	69	1.04	45	7.0	0.053	9.8	153
FQTRS	83	1.10	40	6.7	0.061	10.6	141
FQQAL	8	0.20	8	0.8	0.043	2.7	44
FQQAL	22	0.24	13	1.2	0.015	3.2	73
FQQAL	33	0.57	26	2.7	0.117	10.3	134
FQQAL	42	0.98	38	3.9	0.193	19.7	179
FQQAL	53	0.92	31	3.3	0.146	17.6	130
FQQAL	69	0.83	24	2.6	0.157	13.4	85
FQQAL	80	0.84	25	2.8	0.253	14.1	85
FQQAL	88	0.72	26	3.3	0.182	10.2	71
FQQAL	98	0.73	25	3.2	0.136	8.9	67
	(m)						
BGO-3A	8.5	0.13	1.6	0.26		3.4	20
BGO-3A	14.0		3.1	0.51		8.0	93
BGO-3A	23.2	0.89	9.4	2.2		249	2820
BGO-3A	29.7	2.50	25	3.6		247	1280
BGO-3A	30.0	1.88	16	3.0		420	1967
BGO-3A	30.7	3.82	34	4.5		180	990

Note. For the second column under Cs only, values are the mass of Cs extracted from unit mass of bulk soil by ion exchange with ammonium acetate relative to the soil mass.

Table S10. Elements in Residues of Strong Acid Extractions

Core	Mean depth (cm)	K (g kg⁻¹)	Rb (mg kg⁻¹)	Cs (mg kg⁻¹)	Sr (mg kg⁻¹)	Ba (mg kg⁻¹)
FQ	10	2.8	15	1.1	19	95
FQ	28	4.3	21	1.3	25	134
FQ	46	4.7	19	0.9	21	140
FQ	66	5.1	21	0.9	22	140
FQ	86	4.8	28	1.9	28	158
FQ	107	4.4	28	2.1	28	144
FQ	127	5.1	36	3.5	32	135
FQ	147	2.6	24	2.0	21	84
FQ	168	2.2	22	2.6	23	63
FQ	191	2.3	22	2.5	24	70
FQTRS	8	3.5	17	1.0	20	109
FQTRS	23	3.8	17	0.8	21	120
FQTRS	39	4.6	18	0.8	23	138
FQTRS	53	4.0	21	1.1	21	132
FQTRS	69	3.9	32	2.4	26	118
FQTRS	83	3.3	29	2.4	28	105
FQQAL	8	16.1	47	0.6	45	505
FQQAL	22	20.0	62	0.8	51	574
FQQAL	33	16.2	61	1.5	38	450
FQQAL	42	10.1	55	1.9	21	255
FQQAL	53	7.2	45	1.8	21	258
FQQAL	69	11.5	48	2.2	24	329
FQQAL	80	7.0	44	1.6	21	272
FQQAL	88	7.7	43	1.7	17	184
FQQAL	98	8.1	43	1.6	19	195
	(m)					
BGO-3A	8.5		24	0.7	28	154

Note. The amount of each element in each extraction residue is reported relative to the original mass of soil fine fraction subjected to the extraction. K in the extraction residue from BGO-3A, 8.5 m depth, was not determined.

Table S11. Percentages Extracted by Acid and Exchanged by Ammonium Acetate

Core	Mean depth (cm)	K (%)	Rb (%)	Cs (%)		Sr (%)	Ba (%)
FQ	10	7.6	42	72	5.1	27	31
FQ	28	4.1	46	76	6.1	12	25
FQ	46	5.6	48	81	5.2	15	26
FQ	66	5.8	52	85	5.0	17	26
FQ	86	4.4	48	76	4.6	11	23
FQ	107	6.6	49	73	4.8	15	28
FQ	127	10.9	53	71	3.2	18	43
FQ	147	23	61	80	3.0	32	54
FQ	168	27	55	68	3.7	27	50
FQ	191	12.3	33	55	4.1	16	29
FQTRS	8	8.9	56	78	5.9	16	32
FQTRS	23	6.7	53	80	5.6	15	32
FQTRS	39	5.4	46	75	8.3	13	22
FQTRS	53	8.6	51	77	7.0	16	29
FQTRS	69	21	60	71	2.7	27	57
FQTRS	83	25	57	72	2.5	30	61
FQQAL	8	1.2	15	54	20.7	5.9	8.0
FQQAL	22	1.2	17	58	3.4	5.9	11
FQQAL	33	3.4	30	63	7.3	22	22
FQQAL	42	9.0	42	67	5.8	48	40
FQQAL	53	9.4	40	64	4.1	46	33
FQQAL	69	6.6	36	56	4.9	36	20
FQQAL	80	7.2	35	64	8.4	41	24
FQQAL	88	8.5	38	65	7.6	37	28
FQQAL	98	8.5	38	65	7.1	32	24

Notes. Except for the second column under Cs, values are the percentages of elements extracted by hot, strong acid from soil fine fractions. The percentages are relative to the total of each element determined by Actlabs (Table S10), with two exceptions explained at the beginning of this section (§4). For the second column under Cs only, values represent the amount of Cs extracted from bulk soil expressed as a percentage of the amount of Cs in the soil, the latter calculated as explained above (§2.7) by the assumption that there is no Cs in the sand-sized fraction.

Table S12. Mass Ratios of Trace Elements to K in Extracts and Residues

Core	Mean depth (cm)	Extracts				Residues			
		Rb/K	Cs/K	Sr/K	Ba/K	Rb/K	Cs/K	Sr/K	Ba/K
FQ	10	0.045	0.0108	0.029	0.19	0.0052	0.00038	0.0068	0.033
FQ	28	0.086	0.0218	0.018	0.26	0.0048	0.00031	0.0057	0.031
FQ	46	0.068	0.0145	0.014	0.18	0.0047	0.00021	0.0049	0.032
FQ	66	0.068	0.0143	0.015	0.16	0.0041	0.00018	0.0043	0.027
FQ	86	0.102	0.0206	0.014	0.20	0.0058	0.00039	0.0059	0.033
FQ	107	0.078	0.0165	0.014	0.17	0.0064	0.00049	0.0063	0.033
FQ	127	0.067	0.0138	0.011	0.17	0.0070	0.00068	0.0063	0.027
FQ	147	0.044	0.0093	0.012	0.12	0.0095	0.00078	0.0082	0.033
FQ	168	0.030	0.0067	0.010	0.08	0.0100	0.00117	0.0101	0.028
FQ	191	0.035	0.0094	0.013	0.09	0.0099	0.00111	0.0106	0.031
FQTRS	8	0.063	0.0113	0.012	0.15	0.0049	0.00028	0.0058	0.031
FQTRS	23	0.069	0.0115	0.013	0.20	0.0045	0.00022	0.0056	0.031
FQTRS	39	0.061	0.0097	0.012	0.15	0.0040	0.00018	0.0051	0.030
FQTRS	53	0.053	0.0085	0.010	0.13	0.0051	0.00026	0.0053	0.033
FQTRS	69	0.044	0.0067	0.009	0.15	0.0081	0.00061	0.0067	0.030
FQTRS	83	0.037	0.0061	0.010	0.13	0.0089	0.00074	0.0084	0.032
FQQAL	8	0.043	0.0040	0.014	0.22	0.0029	0.00004	0.0028	0.031
FQQAL	22	0.053	0.0050	0.013	0.30	0.0031	0.00004	0.0025	0.029
FQQAL	33	0.045	0.0046	0.018	0.23	0.0038	0.00009	0.0023	0.028
FQQAL	42	0.038	0.0040	0.020	0.18	0.0054	0.00019	0.0021	0.025
FQQAL	53	0.033	0.0036	0.019	0.14	0.0051	0.00021	0.0023	0.029
FQQAL	69	0.029	0.0031	0.016	0.10	0.0041	0.00019	0.0021	0.029
FQQAL	80	0.029	0.0034	0.017	0.10	0.0041	0.00014	0.0019	0.025
FQQAL	88	0.036	0.0046	0.014	0.10	0.0056	0.00023	0.0022	0.024
FQQAL	98	0.034	0.0043	0.012	0.09	0.0052	0.00020	0.0023	0.024
	(m)								
BGO-3A	8.5	0.012	0.0019	0.025	0.15				
BGO-3A	23.2	0.011	0.0025	0.28	3.2				
BGO-3A	29.7	0.010	0.0015	0.10	0.51				
BGO-3A	30.0	0.009	0.0016	0.22	1.05				
BGO-3A	30.7	0.009	0.0012	0.05	0.26				

Note. No data were obtained from the residues of acid extraction of fine fraction of the BGO-3A core samples.

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