Electronic Supplementary Material (ESI) for Environmental Science: Processes & Impacts.

Supplementary Information for

The Fate of Inhaled Uranium-Containing Particles upon Clearance to Gastrointestinal Tract

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Table S1: The compositions of the Simulated Gastrointestinal Fluids[1]

	Simulated Gastric Fluid	Simulated Intestinal Fluid
Sodium taurocholate (mM)	0.08	3
Lecithin (mM)	0.02	0.2
Pepsin (mg/mL)	0.1	-
Sodium chloride (mM)	34.2	68.62
Maleic acid (mM)	-	19.12
Sodium hydroxide	-	34.8
pH	1.6	6.5

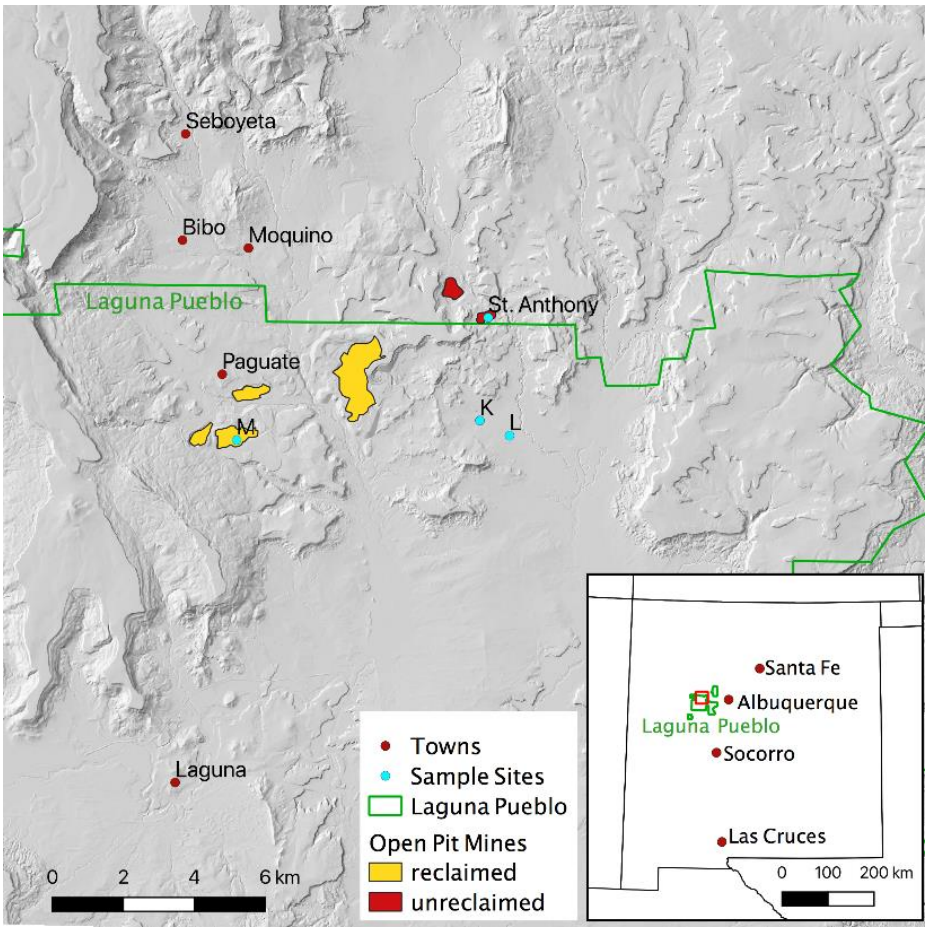

Figure S1: A map of the sampling area.

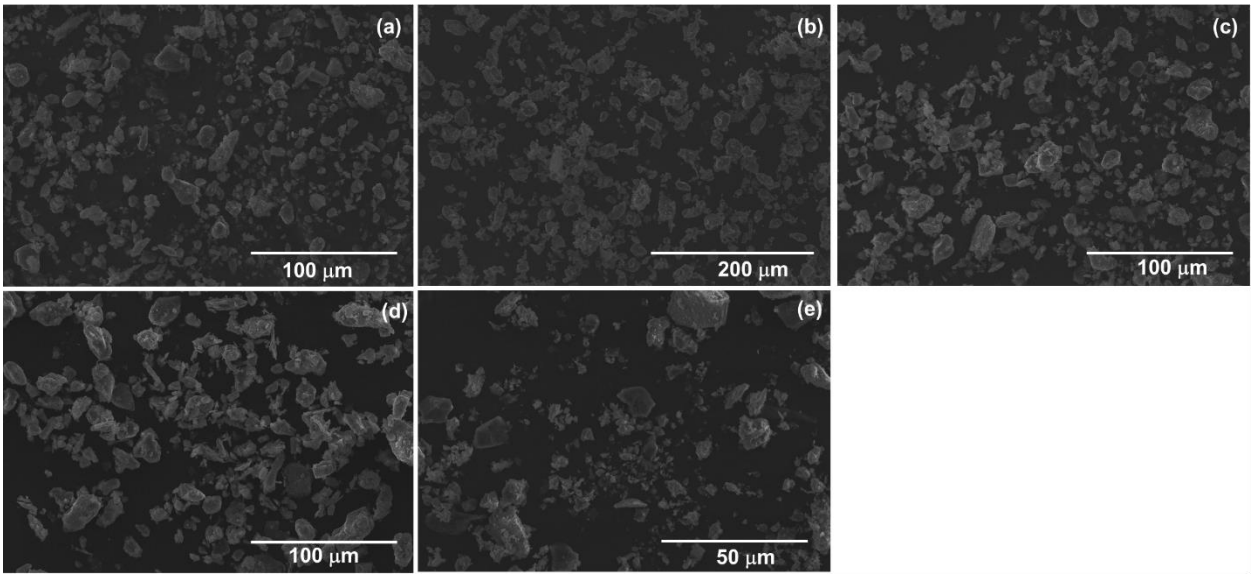

Figure S2: SEM images of the samples from (a) Site K (b) Site L (c) Site M (d) St. Anthony Mine sediment, and (e) St. Anthony rock

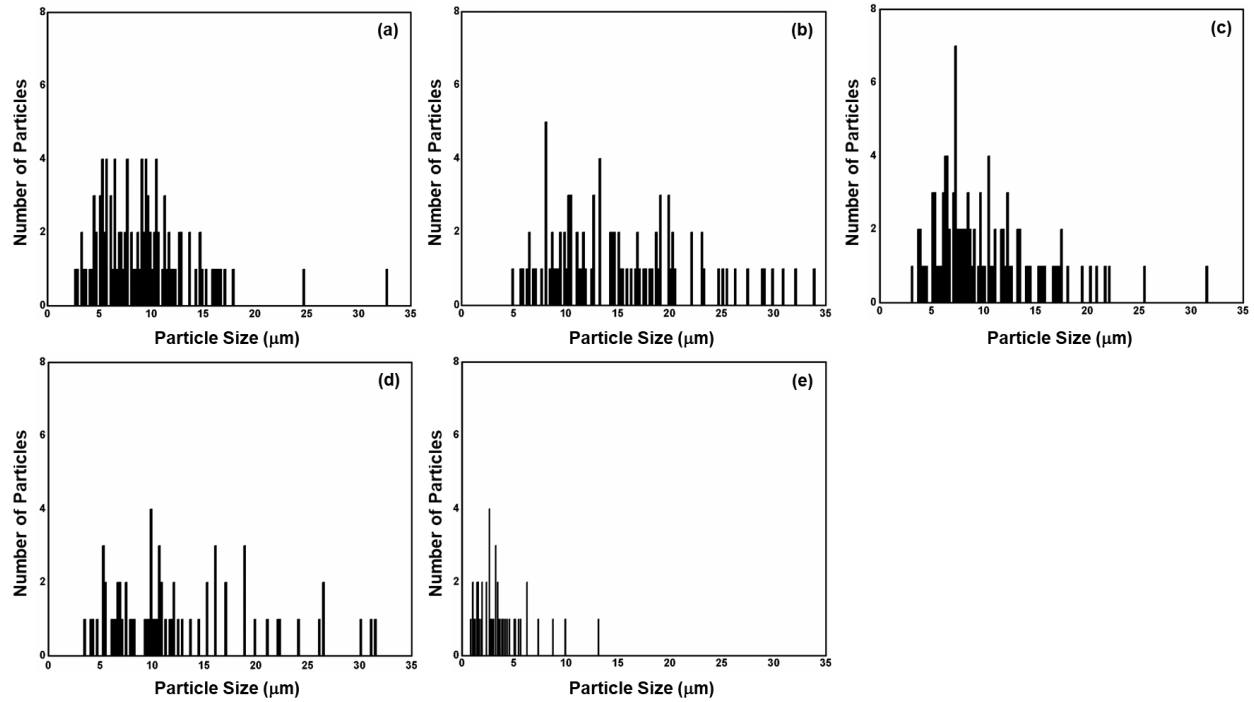

Figure S3: Particle-size distribution of the samples from (a) Site K (b) Site L (c) Site M (d) St. Anthony sediments, and (e) St. Anthony rock

Computation Calculations – Input Files for the SGIF Solutions

SGF Solution

```
PHASES
Uranophane
    Ca(UO2)2(SiO3OH)2 + 6H+ = 2UO2+2 + Ca+2 + 2H4SiO4
    log_k     17.49
    delta_h   0 kcal
    -Vm       68.32 cm3/mol
PHASES
Coffinite
    USiO4 + 4H+ = H4SiO4 + U+4
    log_k     -7.62
    delta_h   -14.548 kcal
    -Vm       68.32 cm3/mol
SURFACE_MASTER_SPECIES
    Uranophane    Ca(UO2)2(SiO3OH)2
SURFACE_MASTER_SPECIES
    Rutile        TiO2
SURFACE_MASTER_SPECIES
    Coffinite     USiO4
    Quartz        SiO2
SURFACE_SPECIES
SiO2 = SiO2
    log_k     -3.9993
    delta_h   32.949 kJ
SURFACE_SPECIES
TiO2 +2.0000 H2O  =  + 1.0000 Ti(OH)4
    log_k     -9.6452
    delta_h   -226.107 kJ
PHASES
Uranyl_carbonate
    Na4UO2(CO3)3 +3.0000 H+  =  + 1.0000 UO2++ + 3.0000 HCO3- + 4.0000 Na+
    log_k     4.0395
    delta_h   0 kcal
    -Vm       68.32 cm3/mol
PHASES
Rutile
    TiO2 +2.0000 H2O  =  + 1.0000 Ti(OH)4
    log_k     -9.6452
    delta_h   -226.107 kcal
    -Vm       68.32 cm3/mol
SURFACE_MASTER_SPECIES
    Uranyl_carbonate Na4UO2(CO3)3
SURFACE_SPECIES
Na4UO2(CO3)3 = Na4UO2(CO3)3
    log_k     0
SURFACE_SPECIES
TiO2 = TiO2
    log_k     0
PHASES
Microcline
    KAlSi3O8 + 4H2O + 4H+ = 3H4SiO4 + Al+3 + K+
    log_k     0.616
    delta_h   -12.309 kcal
SURFACE_MASTER_SPECIES
```

```
    Microcline    KAlSi3O8
SURFACE_SPECIES
KAlSi3O8 = KAlSi3O8
    log_k     0
SOLUTION_MASTER_SPECIES
    Lactate       Lactate-         0     90.08           90.08
SOLUTION_MASTER_SPECIES
    Ti            Ti(OH)4          0     47.88           47.88

SOLUTION_MASTER_SPECIES
    Taurine       Taurine-         0     125.15          125.15
SURFACE_SPECIES
Ca(UO2)2(SiO3OH)2 = Ca(UO2)2(SiO3OH)2
    log_k     0
    delta_h   0 kJ
SOLUTION_SPECIES
Lactate- = Lactate-
    log_k     0
Lactate- + H+ = LactateH
    log_k     3.86
    delta_h   -1361.9 kJ
SOLUTION_SPECIES
Ti(OH)4 = Ti(OH)4
    log_k     0
SOLUTION_SPECIES
Taurine- = Taurine-
    log_k     0
H+ + Taurine- = TaurineH
    log_k     8.82

SOLUTION_MASTER_SPECIES
    Pyruvate      Pyruvate-        0     88.06           88.06
SOLUTION_SPECIES
Pyruvate- = Pyruvate-
    log_k     0
Pyruvate- + H+ = PyruvateH
    log_k     2.5
EQUILIBRIUM_PHASES 1
    Autunite  0 0
    Calcite   0 0
    Carnotite 0 0
    Coffinite 0 0
    Dolomite(disordered) 0 0
    Kaolinite 0 0
    Microcline 0 0
    Quartz    0 0
    Schoepite 0 0
    Torbernite 0 0
    Tyuyamunite 0 0
    Uraninite 0 0
    Uranophane 0 0
    Uranyl_carbonate 0 0
    Rutile    0 0
SOLUTION 2
    temp      37
    pH        1.6
    pe        4
```

```
    redox     pe
    units     mmol/l
    density   1
    Cl        34.2
    Glycine   0.003
    Na        34.28
    P         0.02
    Taurine   0.08
    -water    0.1 # kg
GAS_PHASE 1
    -fixed_volume
    -equilibrium with solution 2
    -pressure 1
    -volume 1
    -temperature 37
    O2(g)     1
SURFACE_SPECIES
USiO4 = USiO4
    log_k     0
```

SIF Solution

```
PHASES
Uranophane
    Ca(UO2)2(SiO3OH)2 + 6H+ = 2UO2+2 + Ca+2 + 2H4SiO4
    log_k     17.49
    delta_h   0 kcal
    -Vm       68.32 cm3/mol
PHASES
Coffinite
    USiO4 + 4H+ = H4SiO4 + U+4
    log_k     -7.62
    delta_h   -14.548 kcal
    -Vm       68.32 cm3/mol
SURFACE_MASTER_SPECIES
    Uranophane    Ca(UO2)2(SiO3OH)2
SURFACE_MASTER_SPECIES
    Rutile        TiO2
SURFACE_MASTER_SPECIES
    Coffinite     USiO4
    Quartz        SiO2
SURFACE_SPECIES
SiO2 = SiO2
    log_k     -3.9993
    delta_h   32.949 kJ
SURFACE_SPECIES
TiO2 +2.0000 H2O  =  + 1.0000 Ti(OH)4
    log_k     -9.6452
    delta_h   -226.107 kJ
PHASES
Uranyl_carbonate
    Na4UO2(CO3)3 +3.0000 H+  =  + 1.0000 UO2++ + 3.0000 HCO3- + 4.0000 Na+
    log_k     4.0395
    delta_h   0 kcal
    -Vm       68.32 cm3/mol
PHASES
```

```
Rutile
    TiO2 +2.0000 H2O  =  + 1.0000 Ti(OH)4
    log_k     -9.6452
    delta_h   -226.107 kcal
    -Vm       68.32 cm3/mol
SURFACE_MASTER_SPECIES
    Uranyl_carbonate Na4UO2(CO3)3
SURFACE_SPECIES
Na4UO2(CO3)3 = Na4UO2(CO3)3
    log_k     0
SURFACE_SPECIES
TiO2 = TiO2
    log_k     0
PHASES
Microcline
    KAlSi3O8 + 4H2O + 4H+ = 3H4SiO4 + Al+3 + K+
    log_k     0.616
    delta_h   -12.309 kcal
SURFACE_MASTER_SPECIES
    Microcline    KAlSi3O8
SURFACE_SPECIES
KAlSi3O8 = KAlSi3O8
    log_k     0
SOLUTION_MASTER_SPECIES
    Lactate       Lactate-         0     90.08          90.08
SOLUTION_MASTER_SPECIES
    Malate        Malate-          0     116            116
SOLUTION_MASTER_SPECIES
    Ti            Ti(OH)4          0     47.88          47.88

SOLUTION_MASTER_SPECIES
    Taurine       Taurine-         0     125.15         125.15
SURFACE_SPECIES
Ca(UO2)2(SiO3OH)2 = Ca(UO2)2(SiO3OH)2
    log_k     0
    delta_h   0 kJ
SOLUTION_SPECIES
Lactate- = Lactate-
    log_k     0
Lactate- + H+ = LactateH
    log_k     3.86
    delta_h   -1361.9 kJ
SOLUTION_SPECIES
Malate- = Malate-
    log_k     0
H+ + Malate- = MalateH
    log_k     1.9
SOLUTION_SPECIES
Ti(OH)4 = Ti(OH)4
    log_k     0
SOLUTION_SPECIES
Taurine- = Taurine-
    log_k     0
H+ + Taurine- = TaurineH
    log_k     8.82

SOLUTION_MASTER_SPECIES
```

```
    Pyruvate      Pyruvate-        0     88.06           88.06
SOLUTION_SPECIES
Pyruvate- = Pyruvate-
    log_k     0
Pyruvate- + H+ = PyruvateH
    log_k     2.5
EQUILIBRIUM_PHASES 1
    Autunite  0 0
    Calcite   0 0
    Carnotite 0 0
    Coffinite 0 0
    Dolomite(disordered) 0 0
    Kaolinite 0 0
    Microcline 0 0
    Quartz    0 0
    Schoepite 0 0
    Torbernite 0 0
    Tyuyamunite 0 0
    Uraninite 0 0
    Uranophane 0 0
    Uranyl_carbonate 0 0
    Rutile    0 0
SOLUTION 2
    temp      37
    pH        6.5
    pe        4
    redox     pe
    units     mmol/l
    density   1
    Cl        34.2
    Glycine   0.003
    Malate    19.12
    Na        34.28
    P         0.02
    Taurine   0.08
    -water    0.1 # kg
GAS_PHASE 1
    -fixed_volume
    -equilibrium with solution 2
    -pressure 1
    -volume 1
    -temperature 37
    O2(g)     1
SURFACE_SPECIES
USiO4 = USiO4
    log_k     0
```

Table S2: Mineralogy Input to PHREEQC for the Computational Calculations.

Mineral	Chemical Formula	St. A. S	St. A. R	Site K	Site L	Site M
Quartz	SiO_2	99	99	99	99	99
Dolomite	$CaCO_3.MgCO_3$	1	0	1	1	1
Microcline	$KAlSi_3O_8$	0	1	1	1	1
Kaolinite	$Al_2Si_2O_5(OH)_4$	1	1	1	1	1
Calcite	$CaCO_3$	0	0	0	0	0
Rutile	TiO_2	1	0	0	0	0
Uraninite	UO_2	0.01	0	0.7	0.8	0.8
Coffinite	$U(SiO_4)_{1-x}(OH)_{4x}$	0.01	1	0	0.4	0
Andersonite	$Na_2Ca(UO_2)(CO_3)_3 \cdot 6H_2O$	0	0.45	0.0003	0	0
Torbernite	$Cu(UO_2)_2(PO_4)_2 \bullet 12H_2O$	0	0.1	0	0.02	0.15
Tyuyamunite	$Ca(UO_2)_2V_2O_8 \cdot (5\text{-}8)H_2O$	0	0	0.01	0.02	0.01
Carnotite	$K_2(UO_2)_2(VO_4)_2 \cdot 3H_2O$	0	0.2	0.04	0	0
Uranophane	$(Ca(UO_2)_2(SiO_3OH)_2 \cdot 5H_2O)$	0.001	0.03	0.02	0.015	0
Schoephite	$(UO_2)_8O_2(OH)_{12} \bullet 12(H_2O)$	0.1	0	0	0	0
Autunite	$Ca(UO_2)_2(PO_4)_2 \cdot 10\text{–}12H_2O$	0.08	1	0.2	0.15	0.25

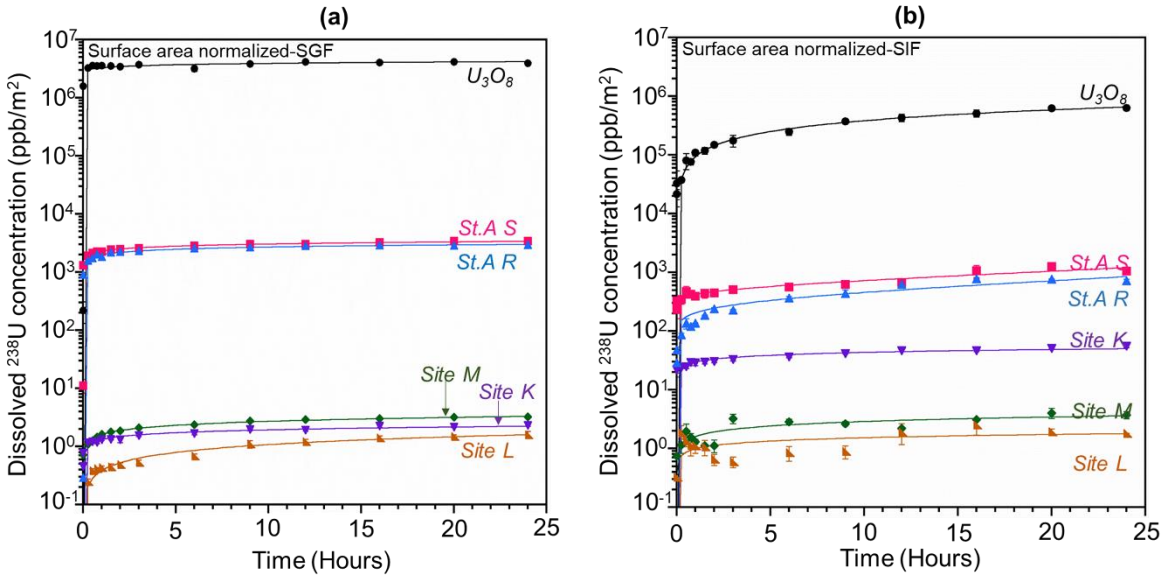

Figure S4: The surface-area-normalized dissolutions of uranium from natural dust and sediment samples in (a) SGF solution and (b) SIF solution

Table S 3: The ratio of %U normalized dissolved U in SGF to SIF after 24 hours of dissolution experiment.

Sample	Dissolution ratio after 24 hours SGF/SIF
U_3O_8	6.85
St.A. S	4.20
St.A. R	4.35
Site K	0.06
Site L	0.88
Site M	0.53

Table S4: Final pH (after 24 hours passed) of the dust-treated SGIF Solutions. The initial pH of the SGF solution is 1.60 and that of the SIF solution is 6.50.

Sample	**Final pH**	
	SGF	SIF
U_3O_8	1.46 ± 0.05	6.42 ± 0.09
STAS	1.89 ± 0.06	6.02 ± 0.16
STAR	1.63 ± 0.05	5.93 ± 0.16
Site K	1.63 ± 0.04	6.28 ± 0.04
Site L	1.69 ± 0.06	6.12 ± 0.07
Site M	1.68 ± 0.09	6.23 ± 0.09

Uranyl Cation Detection[2]

The colorimetric analysis of the Uranyl-Curcumin-Triton-X System was used to qualitatively determine (orange coloration) the presence of the uranyl cation (UO_2^{2+}). Smaller quantities of dust in SGIF solutions did not show a visible color development. Therefore, to confirm the presence of

uranyl cation in dust-treated SGIF solutions, a higher quantity of dust (~50 mg in 3 mL of fluid) was treated for 24 hours using the same dissolution conditions.

Table S5: Masses of natural dust and sediment samples used in uranyl cation detection.

Samples	Amount in 3 mL of SGF (mg)	Amount in 3 mL of SIF (mg)
U_3O_8	50.21	50.16
St. Anthony Sediments	50.22	50.89
St. Anthony Rock	50.53	50.51
Site K	50.15	51.17
Site L	47.19	50.09
Site M	50.13	50.67

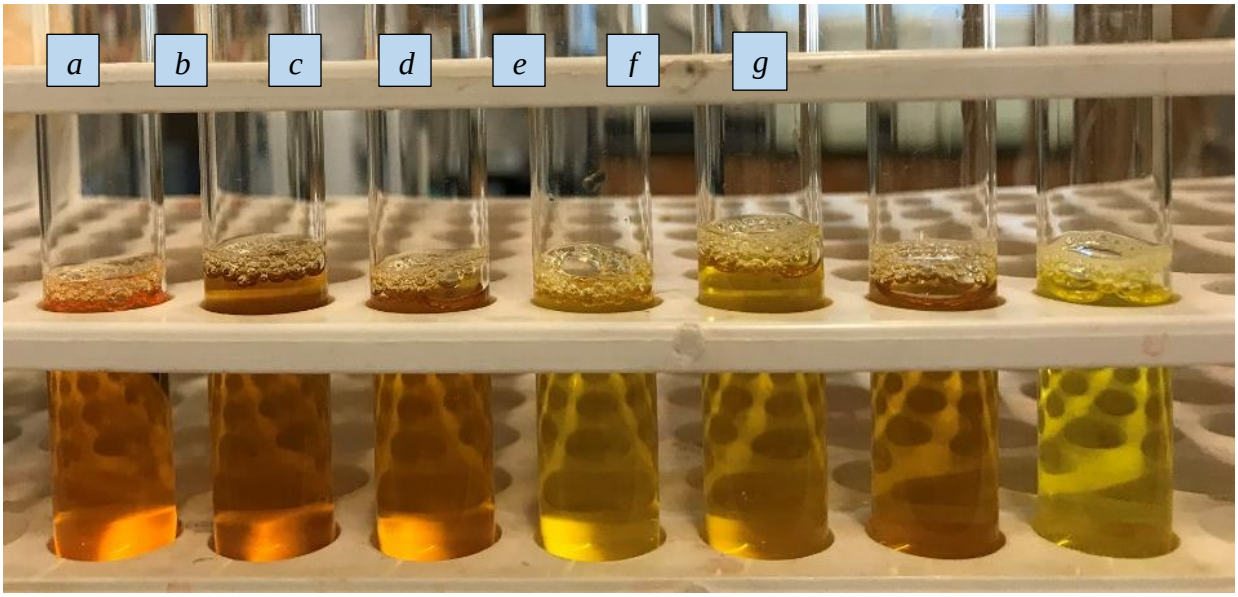

Figure S5: Orange coloration for Uranyl-Curcumin-Triton-X experiment for (a) U_3O_8, (b) St. Anthony sediment, (c) St. Anthony rock, (d) Site K, (e) Site L, and (f) Site M, while (g) is the Blank**.**

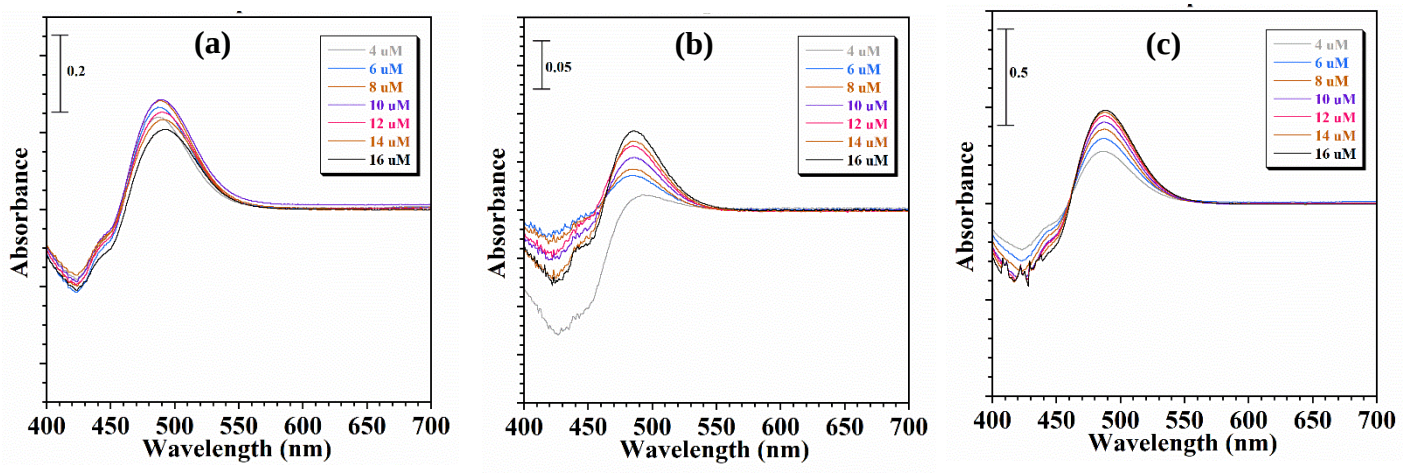

Figure S6: UV-VIS scanned spectra of uranyl-curcumin-triton-x system in (a) SGF, (b) SIF, & (c) water.

Table S6: Calculated Equilibrium U Concentrations

	Dissolved [U]/ M								
	Without Kaolinite			With Kaolinite			With Microcline		
Mineral	**SGF**	**SIF**	**SGF/SIF**	**SGF**	**SIF**	**SGF/SIF**	**SGF**	**SIF**	**SGF/SIF**
Andersonite	6.76E-03	9.87E-03	0.68	6.76E-03	9.87E-03	0.68	1.00E-02	9.87E-03	1.01
Autunite	2.00E-02	5.53E-04	36.17	1.02E-02	5.58E-04	18.23	8.04E-04	5.43E-04	1.48
Carnotite	4.70E-03	8.51E-06	552.50	1.67E-04	8.89E-06	18.97	2.08E-06	1.12E-06	1.86
Coffinite	1.99E-09	1.88E-11	106.35	5.84E-11	1.87E-11	3.12	1.62E-11	2.46E-09	6.6E-03
Schoepite	1.00E-02	1.93E-05	518.14	9.99E-03	2.42E-05	412.42	2.44E-04	8.65E-06	28.26
Torbentite	2.00E-02	2.29E-04	87.41	3.48E-03	2.30E-04	15.10	2.61E-04	5.99E-04	0.43
Tyuyamunite	6.18E-03	7.49E-06	824.70	1.52E-04	7.96E-06	19.06	1.75E-06	1.10E-06	1.60
Uranophane	1.91E-02	5.93E-06	3216.34	1.91E-02	6.22E-06	3063.69	9.24E-05	6.22E-06	14.86
U_3O_8	1.44E-02	8.61E-06	1676.54	1.05E-03	9.08E-06	115.22	9.57E-06	7.32E-07	13.079
Uraninite	1.99E-09	1.89E-11	105.39	5.84E-11	1.72E-11	3.40	1.62E-11	2.46E-09	6.6E-03
For Natural dust and sediment samples									
STS	4.31E-03	9.41E-04	4.585371	--	--	--	--	--	--
STR	5.81E-01	5.54E-01	1.04871	--	--	--	--	--	--
K	6.95E-04	3.98E-03	0.174768	--	--	--	--	--	--
L	9.24E-04	7.52E-03	0.123179	--	--	--	--	--	--
M	7.29E-04	7.37E-03	0.098955	--	--	--	--	--	--

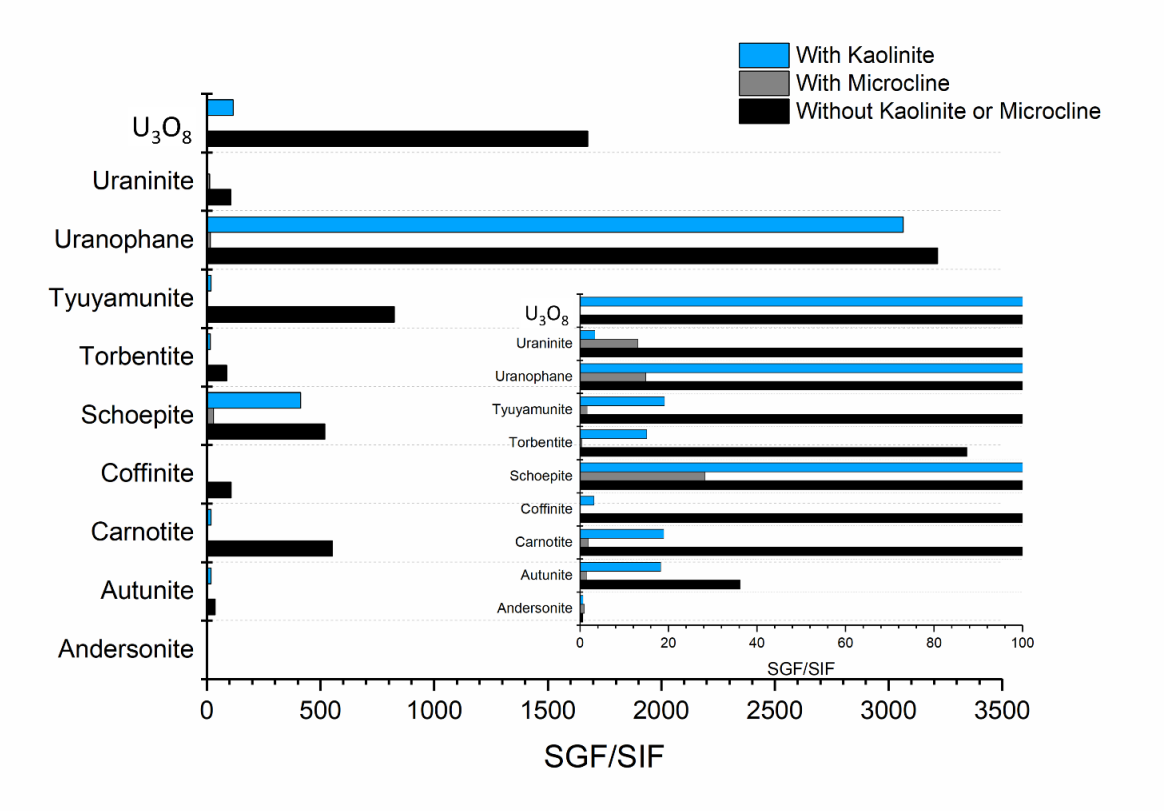

Figure S7: Single-phase uranium solubility ratio of SGF/SIF (a) with and without kaolinite (b) percent decrease in SGF/SIF ratio as a f(Kaolinite).

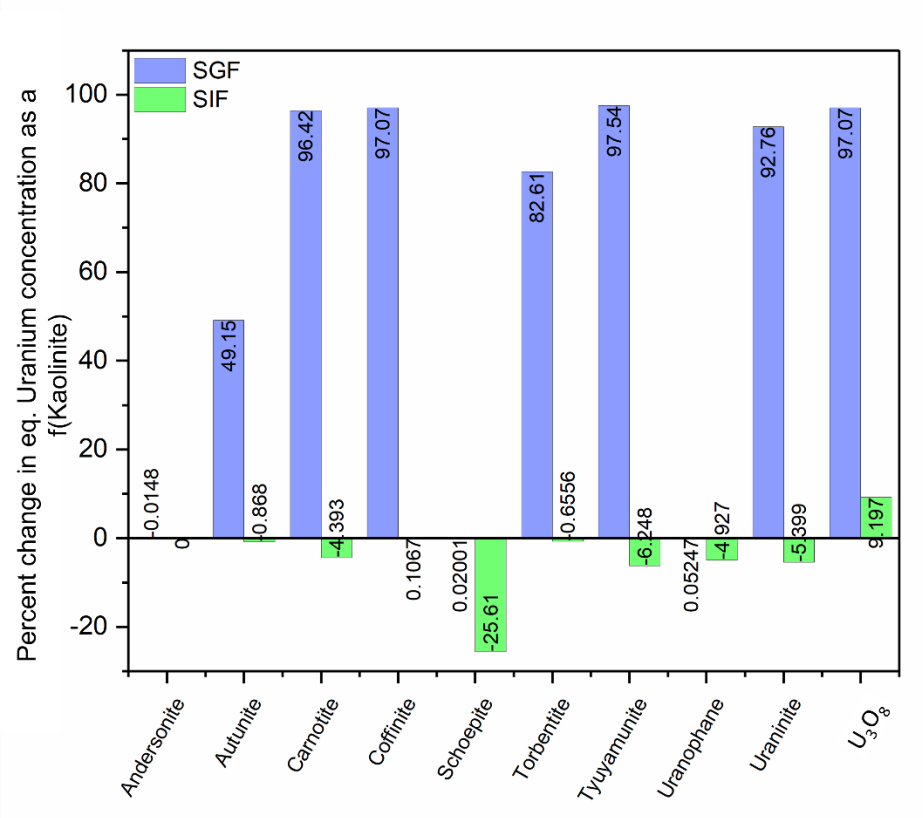

Figure S8: The percent change in uranium dissolution for each mineral in each fluid upon addition of kaolinite. The positive numbers indicate a decrease in concentration while the negative numbers indicate an increase in concentration.

References

(1) Marques, M. R. C.; Loebenberg, R.; Almukainzi, M. Simulated Biological Fluids with Possible Application in Dissolution Testing. *Dissolution Technol.* **2011**, *18* (3), 15–28. https://doi.org/10.14227/DT180311P15.

(2) Zhu, J. H.; Zhao, X.; Yang, J.; Tan, Y. T.; Zhang, L.; Liu, S. P.; Liu, Z. F.; Hu, X. L. Selective Colorimetric and Fluorescent Quenching Determination of Uranyl Ion via Its Complexation with Curcumin. *Spectrochim. Acta - Part A Mol. Biomol. Spectrosc.* **2016**, *159*, 146–150. https://doi.org/10.1016/j.saa.2016.01.021.