

Supporting Information

Mineralogy Controlled Dissolution of Uranium from Airborne Dust in Simulated Lung Fluids (SLFs) and Possible Health Implications

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Locations of the Collected Dust

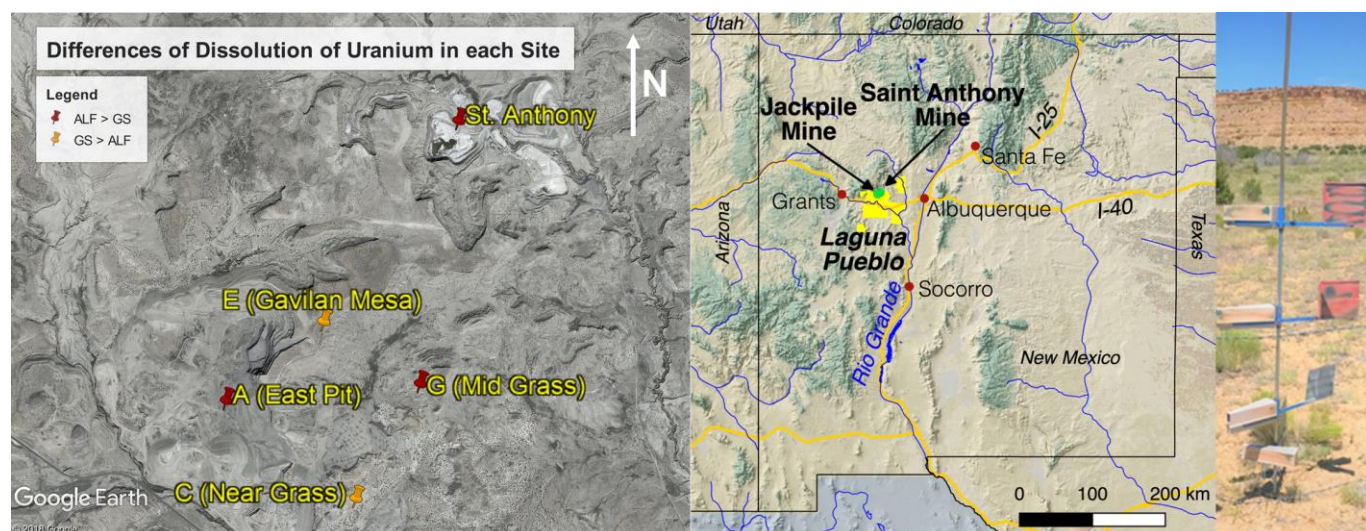


Figure S1. Dust sample collection locations (left) and area map (center). Picture of four Big Spring Number Eight passive dust collectors at four sampling heights (right). Only the dust from the highest collector, at 1.5 m height, was used in this study. The area map was created using QGIS 2.18 software. Data source: National Map Small Scale Collection, hosted by USGS.

Dust and sediment samples were collected from different topographies (**Figure S1**) in the vicinity of Jackpile Mine on Laguna Pueblo, New Mexico, and St. Anthony Mine to the north. Overall, the climate is semiarid, with the area receiving most of its annual rainfall during the monsoon season from July to August. The rest of the year is generally dry with occasional snow in the winter. The spring is dominated by winds from the west and northwest. Throughout the year winds from these directions consistently average the highest wind speeds and the highest gust speeds. Though winds come from all directions, the majority of the aeolian transport was expected to be to the southeast of the main pit. During mining operation, sources of the dust included the pit, rock crushers, uncovered waste rock piles, and onsite ore stockpiles.

The Jackpile Mine is located in the Paguate River valley, with mountains to the west and a series of mesas and valleys to the east. Site A, or East Pit, is 40 m thick backfilled with proto-ore and waste rock above the mineralized uranium deposits. This waste rock does not necessarily contain uranium above background.¹ Site E is located at very top of a Gavilan mesa. Site C is located immediate downwind whereas Site G is located the farthest downwind from the Jackpile mine. A continuing major uranium source in the study area is the St. Anthony Mine (0.87% of uranium by mass), an inactive but completely un-remediated mine located ~5 km to the northeast, just north of the Laguna Pueblo boundary.¹

List of Chemicals

The following chemicals were used to prepare SLFs according to the composition described in Pelfrene et al.² (**Table S1**): sodium chloride (NaCl, Acros, +99.0%), disodium hydrogenphosphate (Na₂HPO₄, Sigma-Aldrich, +99.0%), sodium bicarbonate (NaHCO₃, Sigma, 99.5%), trisodium citrate dihydrate (C₆H₅Na₃O₇, Sigma-Aldrich, +99%), ammonium chloride (NH₄Cl, VWR International, 99.5%), glycine (NH₂CH₂COOH, Aldrich Chemical Company, +99%), sodium dihydrogen phosphate (NaH₂PO₄, Sigma-Aldrich, +99.0%), L-cysteine (C₃H₇NO₂S, Aldrich Chemical Company, 99%), sodium hydroxide (NaOH, VWR International, 97%), citric acid monohydrate (C₆H₈O₇·H₂O, Fluka Analytical, +99%), calcium chloride dihydrate (CaCl₂·2H₂O, Fisher Scientific, +99%), sodium sulfate (Na₂SO₄, Sigma-Aldrich, +99%), magnesium chloride hexahydrate (MgCl₂·6H₂O, Sigma-Aldrich, +99%), disodium tartrate dihydrate (C₄H₄Na₂O₆·2H₂O, Honeywell Riedel – de Haen, 99.5%), sodium L- lactate (C₃H₅NaO₃, Sigma, 98%), sodium pyruvate (C₃H₃NaO₃, Sigma-Aldrich, +99%). Triuranium octaoxide (U₃O₈, National Bureau of Standards, 99.9%) was used in dissolution studies as a uranium-bearing standard reference material. Curcumin (C₂₁H₂₀O₆, TCI America, crystals), Triton-X-100 (C₁₄H₂₂O(C₂H₄O)_n(n=9-10), VWR International, Reagent grade), sodium acetate (NaOOCCH₃, Alfa Aesar, 99%), acetic acid (CH₃COOH, VWR International, 99.7%), and uranyl acetate(UO₂(CH₃COO)₂·2H₂O, Baker Analyzed, 99.4%) were used in the experiments of detecting uranyl cation according to the methods reported in Zhu et al.³

Table S1: Composition of the Simulated Lung Fluids²

Composition (g·L ⁻¹)	GS	ALF
NaCl	6.779	3.21
Na ₂ HPO ₄		0.071
NaHCO ₃	2.268	
Trisodium citrate dihydrate	0.055	0.077
NH ₄ Cl	0.535	
Glycine	0.375	0.059
NaH ₂ PO ₄	1.872	
L-cysteine	0.121	
NaOH		6.0
Citric acid		20.8
CaCl ₂ ·2H ₂ O	0.026	0.128
Na ₂ SO ₄		0.039
MgCl ₂ ·6H ₂ O		0.05
Disodium tartrate		0.09
Sodium lactate		0.085
Sodium pyruvate		0.172
Properties		
pH	7.3 ± 0.1	4.5 ± 0.1
Ionic strength (mol·L ⁻¹)	0.17	0.34

Human respiratory tract can be divided into three major regions. These are extrathoracic or upper respiratory tract, tracheobronchial region and pulmonary or alveolar region, both together named as the lower respiratory tract. Particles that are smaller than 100 μ m can be entered the respiratory tract via inhalation. However, only the particles that are smaller than 10 μ m have the possibility of passing to the lower respiratory tract. The bigger particles may adsorb by the mucus and will be removed eventually. Some of these particles have the possibility of swallowing and entering the gastrointestinal tract. However, the smaller particles will be able to pass through these barriers and will move down the lower respiratory tract. The finest fraction, particles smaller than 4 μ m and nano particle aggregates has the possibility of penetrating into the alveolar region of the deep lungs. The alveolar region is the lung compartment where the oxygen – carbon dioxide gas exchange occurs. Hence, this area is in close contact with the blood vessels. Alveolar region contains three major types of cells. Type I cells that made up the structure, the Type II cells that secrete pulmonary surfactants to the alveolus and, alveolar macrophages which contain lysosomes and ready to consume foreign bodies that reach this region. The Gamble's solution (GS) is simulating the pulmonary surfactants secreting from Type II cells. The Artificial Lysosomal Fluid (ALF) solution simulates the conditions inside these alveolar macrophages when they engulf the foreign body, a process known as phagocytosis. The combined body is known as phagolysosome which contains the highly acidic environment required to destroy the foreign bodies.^{4,5}

Characterization of the Samples

BET Surface Area Analysis of the Samples

Surface areas of samples were measured in a seven-point N₂-Brunauer-Emmet-Teller (BET) isotherm using a Quantachrome Autosorb-1 surface area analyzer. Samples were outgassed overnight (~24 h) at a temperature of 105°C prior to the BET analysis.

EPA-3052b Digestion Method⁶

To determine the elemental concentrations of each dust sample an acid digestion procedure was followed. A 0.2 \pm 0.01 g subsample was weighed from each sample and placed in an individual digestion tube. Samples were sieved through 500 μ m sieve to remove organic debris prior to analysis. Three mL of trace-metal-grade hydrofluoric acid and 9mL of trace-metal-grade nitric acid were added to each digestion tube, and each tube was capped and placed in a holder. A preset microwave routine (Milestone EthosUP) included a 25 minute ramp to 180°C, after which the oven held that temperature for 10 minutes, consistent with the EPA 3052b digestion method.

Pre-concentration and XRD Analysis of the Uranium Minerals of the Dust Samples

As the total %U of these samples are lower than 1% (the usual detection limit of the XRD analysis), a pre-concentration procedure was carried out. The dust samples were first sieved through a 500 μ m US standard sieve to remove debris. The uranium minerals in the samples are coatings around the quartz grains. Therefore, while sieving, the dust was lightly scratched using a porcelain pestle to scratch out the uranium minerals. Additional sieving was carried out using 120 μ m, 45 μ m and 20 μ m US standard sieves. The finest fraction collected was analyzed by XRD. Then, the spectra were compared with 15 different common uranium minerals in New Mexico along with common major minerals (i.e. quartz, kaolinite, microcline, dolomite, calcite, rutile). The presence of uranium minerals was confirmed only when their intensities and d – spacing were matched with respective standard patterns with at least five major peaks.

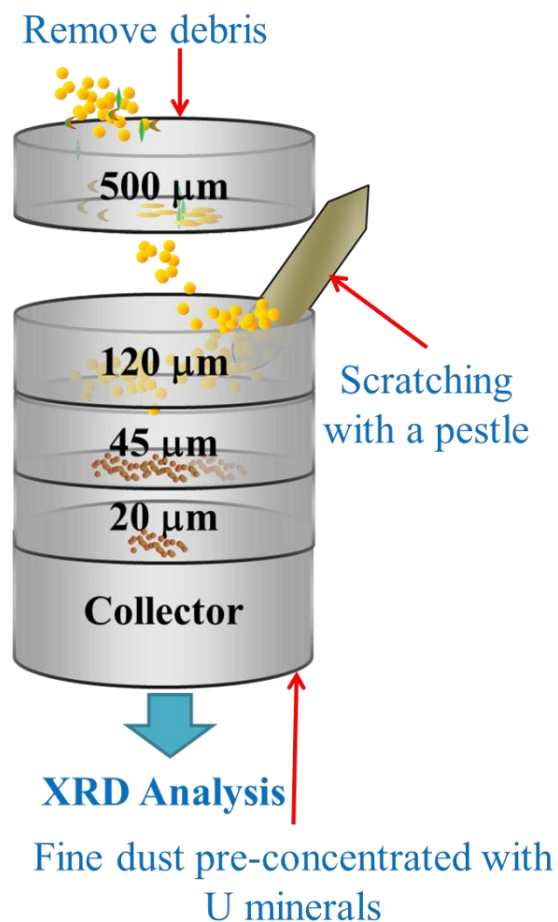


Figure S2: Systematic diagram illustrating the pre-concentration procedure of uranium minerals

Particle Size Analysis

Scanning Electron Microscope (SEM) images of the dust samples were analyzed using the software package ImageJ to obtain the particle sizes. The dust samples were first sonicated in isopropyl alcohol for 30 minutes to prevent aggregation and to obtain a well dispersed suspension. After air drying, the samples were sputter coated with Platinum and imaged with a NOVA-Nano-SEM-450. The obtained images were used to calculate the particle sizes.

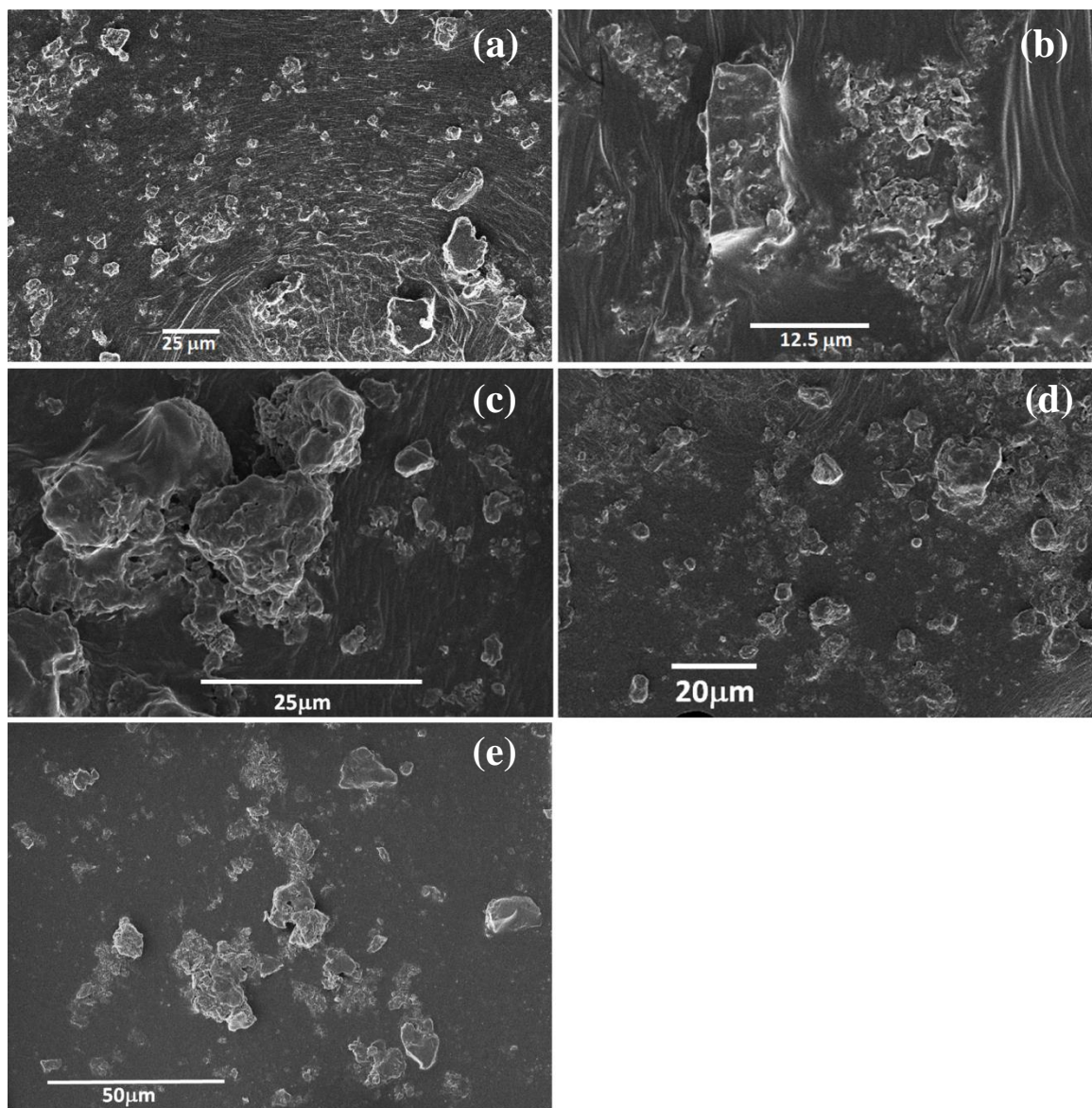


Figure S3: SEM images of the samples of (a) Site A, (b) Site C, (c) Site E, (d) Site G, (e) St. Anthony

Table S2: Particle Sizes of the Samples and their Respective Number of PM₁₀, and PM₄ Particles as a Percentage of Total Number of Particles Analyzed

Sample	# of particles analyzed	Average length (μm)	PM ₁₀	PM ₄
St. Anthony	424	4.6±4.0	90%	57%
A	520	4.7±4.6	88%	61%
C	421	3.4±4.6	92%	75%
E	418	4.0±4.8	89%	74%
G	380	3.9±3.3	94%	69%

Given below, **Figure S4** represents the particle size distribution as a function of size.

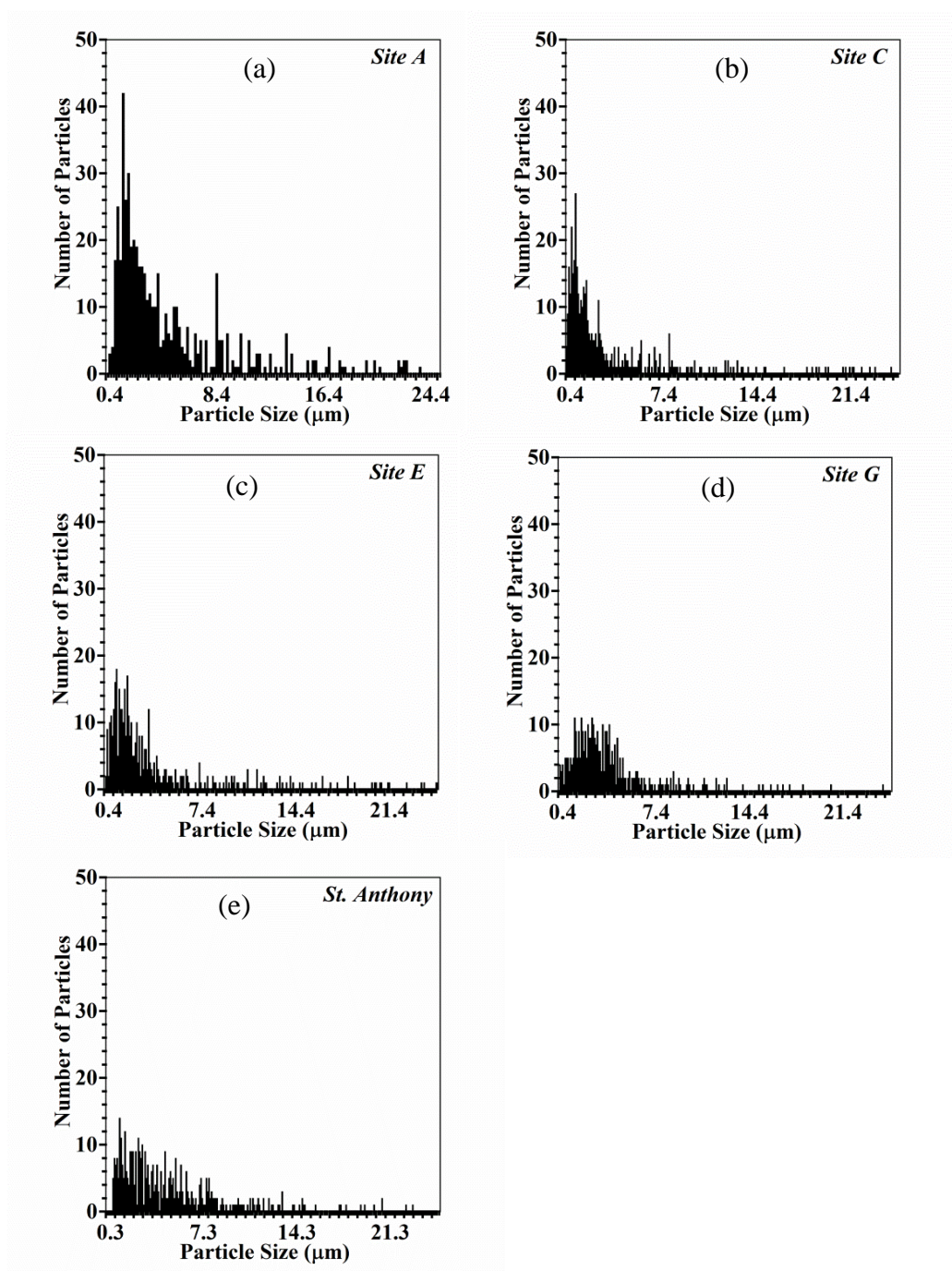


Figure S4: Particle size distribution of the samples from (a) Site A, (b) Site C, (c) Site E, (d) Site G, and (e) St. Anthony. Larger standard deviation of the average length indicates a larger particle size distribution.

Table S3: Specific Surface Area Measurements and Total U% of the Analyzed Samples

Sample	Source of the samples	7 points N ₂ BET surface area (m ² /g)	%U
U ₃ O ₈	National Bureau of Standards, Assay: 99.9%	0.46±0.04	85
St. Anthony Sediment	St. Anthony Mine	1.61±0.08	0.87
Site A	Jackpile Mine	2.10±0.09	0.23
Site E	Jackpile Mine	14.5±1.0	0.18
Site C	Jackpile Mine	0.77±0.14	0.14
Site G	Jackpile Mine	1.77±0.59	0.23

Langmuir Type Model

The Langmuir model fitting was done using inbuilt Langmuir Function (LangmuirEXT1) of OriginPro 8.

$$y = \frac{abx^{1-c}}{1 + bx^{1-c}}$$

where, **y** is the amount of total dissolved uranium, **x** is the reaction time, **a** is the maximum amount of total dissolved uranium, **b** is an equilibrium constant associated with the surface dissolution, and **c** is a coefficient used to obtained the best fit (usually kept at 0).

Table S4: Averaged Rates of U Dissolution for the First 3 hours and Percentage of U Dissolved in each SLF after 24 hour Exposure

Sample	Averaged rates of U dissolution for 1 st 3 hours (µg L ⁻¹ m ⁻² h ⁻¹)		%U dissolved in SLF upon 24 hour exposure	
	GS	ALF	GS	ALF
U ₃ O ₈	3.9E ⁴ ±1120	1.05E ⁶ ±12101	0.743	16.162
St Anthony	104±3	233±6	0.700	1.801
Site A	5.0±0.1	4.6±0.2	0.036	0.043
Site C	1274±21	2017±36	11.086	7.873
Site E	3.2±0.2	2.0±0.2	0.297	0.173
Site G	63±1	184±5	0.810	1.367

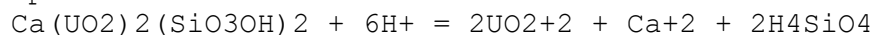
PHREEQC 3.3.8 Input (MINTEQ Database) of the Two Simulated Lung Fluids

- The inbuilt MINTEQ database has used as the basic database. The other solid and aqueous species were imported from the other inbuilt databases (LLNL.dat, MINTEQ.v4.dat) within the model.

GS Solution

PHASES

Uranophane



log_k 17.49

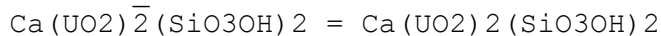
delta_h 0 kcal

-Vm 68.32 cm³/mol

SURFACE_MASTER_SPECIES

Uranophane Ca(UO₂)₂(SiO₃OH)₂

SURFACE_SPECIES

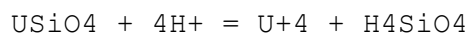


log_k 0

delta_h 0 kJ

PHASES

Coffinite



log_k -7.62

delta_h -14.548 kcal

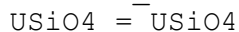
-Vm 68.32 cm³/mol

SURFACE_MASTER_SPECIES

Coffinite USiO₄

Quartz SiO₂

SURFACE_SPECIES



log_k 0

PHASES

Uranyl_carbonate



log_k 4.0395

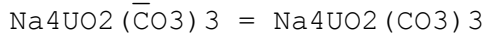
delta_h 0 kcal

-Vm 68.32 cm³/mol

SURFACE_MASTER_SPECIES

Uranyl_carbonate Na₄UO₂(CO₃)₃

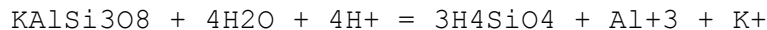
SURFACE_SPECIES



log_k 0

PHASES

Microcline



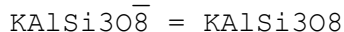
log_k 0.616

delta_h -12.309 kcal

SURFACE_MASTER_SPECIES

Microcline KAlSi₃O₈

SURFACE_SPECIES



log_k 0

SURFACE_SPECIES


```

SiO2 = SiO2
  log_k      -3.9993
  delta_h    32.949 kJ
SOLUTION_MASTER_SPECIES
  Lactate      Lactate-          0      90.08      90.08

SOLUTION_SPECIES
Lactate- = Lactate-
  log_k      0
Lactate- + H+ = LactateH
  log_k      3.86
  delta_h    -1361.9 kJ

SOLUTION_MASTER_SPECIES
  Pyruvate      Pyruvate-        0      88.06      88.06
SOLUTION_SPECIES
Pyruvate- = Pyruvate-
  log_k      0
Pyruvate- + H+ = PyruvateH
  log_k      2.5
GAS_PHASE 1
  -fixed_pressure
  -pressure 1
  -volume 1
  -temperature 37
  O2(g)      1
EQUILIBRIUM_PHASES 1
  Quartz      0 0
  Microcline  0 0
  Dolomite(disordered) 0 0
  Kaolinite   0 0
  Rutile      0 0
  Calcite     0 0
  Uraninite   0 0
  Coffinite   0 0
  Autunite    0 0
  Uranyl_carbonate 0 0
  Torbernite  0 0
  Tyuyamunite 0 0
  Carnotite   0 0
  Uranophane  0 0
  Schoepite   0 0
SOLUTION 2
  temp        37
  pH          7.3
  pe          4
  redox       pe
  units       mmol/l
  density     1
  Na          159.2424
  Cl          126.4344
  Alkalinity  27

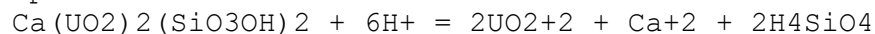
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Citrate	0.187
N(-3)	10.0019
Glycine	4.9953
P	15.6026
S(-2)	0.9987
Ca	0.1769
-water	0.1 # k

ALF Solution

PHASES

Uranophane



log_k 17.49

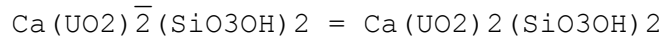
delta_h 0 kcal

-Vm 68.32 cm³/mol

SURFACE_MASTER_SPECIES

Uranophane Ca(UO₂)₂(SiO₃OH)₂

SURFACE_SPECIES

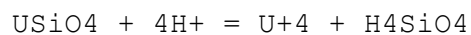


log_k 0

delta_h 0 kJ

PHASES

Coffinite



log_k -7.62

delta_h -14.548 kcal

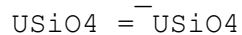
-Vm 68.32 cm³/mol

SURFACE_MASTER_SPECIES

Coffinite USiO₄

Quartz SiO₂

SURFACE_SPECIES



log_k 0

PHASES

Uranyl_carbonate



log_k 4.0395

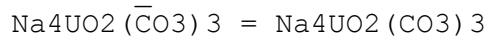
delta_h 0 kcal

-Vm 68.32 cm³/mol

SURFACE_MASTER_SPECIES

Uranyl_carbonate Na₄UO₂(CO₃)₃

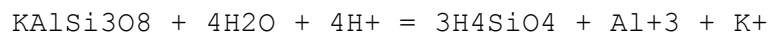
SURFACE_SPECIES



log_k 0

PHASES

Microcline



log_k 0.616

delta_h -12.309 kcal

SURFACE_MASTER_SPECIES

Microcline KAlSi₃O₈

SURFACE_SPECIES

```

KAlSi3O8 = KAlSi3O8
  log_k      0
SURFACE_SPECIES
SiO2 = SiO2
  log_k      -3.9993
  delta_h    32.949 kJ
SOLUTION_MASTER_SPECIES
  Lactate      Lactate-      0      90.08      90.08

SOLUTION_SPECIES
Lactate- = Lactate-
  log_k      0
Lactate- + H+ = LactateH
  log_k      3.86
  delta_h    -1361.9 kJ

SOLUTION_MASTER_SPECIES
  Pyruvate      Pyruvate-      0      88.06      88.06
SOLUTION_SPECIES
Pyruvate- = Pyruvate-
  log_k      0
Pyruvate- + H+ = PyruvateH
  log_k      2.5
GAS_PHASE 1
  -fixed_pressure
  -pressure 1
  -volume 1
  -temperature 37
  O2(g)      1
EQUILIBRIUM_PHASES 1
  Quartz      0 0
  Microcline  0 0
  Dolomite(disordered) 0 0
  Kaolinite   0 0
  Rutile      0 0
  Calcite     0 0
  Uraninite   0 0
  Coffinite   0 0
  Autunite    0 0
  Uranyl_carbonate 0 0
  Torbernite  0 0
  Tyuyamunite 0 0
  Carnotite   0 0
  Uranophane  0 0
  Schoepite   0 0
SOLUTION 2
  temp      37
  pH         4.5
  pe         4
  redox      pe
  units      mmol/l
  density    1

```

Ca	0.8707
Citrate	99.2444
Cl	57.1991
Glycine	0.7859
Lactate	0.7585
Mg	0.2459
Na	211.0301
P	0.807
Pyruvate	1.5636
S(6)	0.2746
Tartarate	0.3912
-water	0.1 # kg

Table S5: The Initial Mineral Molar Ratios of the each Site used in PHREEQC3.3.8; Intensities were Based on the Observed Intensities in XRD Analysis

Bulk mineralogy	Uranium mineralogy	St. Anthony Sediment	Site A	Site C	Site E	Site G
Quartz		99	99	99	99	99
Dolomite		1	1	1	1	1
Kaolinite		1	1	0	0	1
Microcline		0	1	0.01165	0.012	1
Calcite		0	0	1	1	1
Rutile		1	0	0	0	0
	Uraninite	0.01	0	0.06	0.02	0.002
	Coffinite	0.01	0	0.0065	0.005	0.0025
	Andersonite	0	0	0.015	0.00012	0.0005
	Autunite	0.08	0.01	0.012	0.01	0.005
	Torbentite	0	0.01	0	0.005	0.0005
	Tyuyamunite	0	0	0	0	0.00001
	Carnotite	0	0	0.005	0.001	0
	Uranophane	0.001	0	0	0	0
	Schoepfite	0.1	0	0	0	0

Table S6: Modeled Equilibrium Concentrations of Dissolved Uranium for Different Uranium Minerals in each SLF using PHREEQC3.3.8. The dissolution was Modeled by Introducing only One Uranium Containing Mineral at a Time.

Mineral	Dissolved U concentration (M)		
	ALF	GS	GS/ALF
Autunite	1.028E-02	1.626E-02	1.58
Carnotite	4.044E-04	6.513E-03	16.11
Schoephte	2.170E-02	1.252E-02	0.58
Torbernite	6.078E-02	1.574E-02	0.26
Tyuyamunite	4.052E-04	7.207E-03	17.79
Uraninite	7.074E-01	7.081E-01	1.01
Coffinite	8.166E-01	7.792E-01	0.95
Uranophane	4.627E-02	1.186E-02	0.26

Table S7: Modeled Equilibrium Concentrations of Dissolved Uranium for each Site in each SLF Using PHREEQC3.3.8

Site	Dissolved U concentration (M)		
	ALF	GS	GS/ALF
Site A	1.654E-01	3.498E-02	0.211
Site C	9.307E-01	1.128E+00	1.212
Site E	2.499E-01	2.919E-01	1.168
Site G	7.551E-02	3.426E-02	0.454
St. Anthony	5.940E-02	1.174E-02	0.198

Table S8: Percentage of each Uranium Mineral Dissolved in the Two SLFs and the Percentage Contribution of each Mineral to the Extent of Total Dissolved Uranium Concentration at Equilibrium. The Percentage of each Mineral Dissolved in each Site was obtained from the PHREEQC3.3.8. The Percentage Contributions from each Mineral at Equilibrium were obtained using Mass Balance Equations, Constructed using Modeled Data with Experimentally obtained Extents of Dissolutions.

Site	Major uranium minerals identified	% of the mineral dissolved (amount in the solution phase at equilibrium)		% Contribution to the total dissolved U concentration at equilibrium	
		ALF	GS	ALF	GS
St. Anthony	Uraninite	100	100	8	10
	Coffinite	100	100	8	10
	Autunite	0.02	-1*	0.02	0
	Uranophane	10	10	0.2	0.2
	Schoepite	14	10	81.98	79.8
Site A	Autunite	-14*	4	0	30
	Torbernite	100	12	100	70
Site C	Uraninite	100	100	66	51
	Coffinite	100	100	7	6
	Andersonite	100	100	16	13
	Autunite	-61*	100	0	21
	Carnotite	100	100	11	9
Site E	Uraninite	100	100	56.96	53.34
	Coffinite	100	100	14.24	13.33
	Andersonite	92	0	0.31	0
	Autunite	-50*	10	0	5.33
	Torbernite	100	100	28.48	26.67
	Carnotite	0.02	25	0.001	1.32
Site G	Uraninite	100	100	26	33
	Coffinite	100	100	33	42
	Andersonite	100	100	7	8
	Autunite	16	-10*	21	0
	Torbernite	100	100	13	17
	Tyuyamunite	0.005	0.00012	0.00001	0.01

* A negative number of the amount of moles dissolved indicates the particular mineral precipitates under the conditions considered.

The pH of the Solutions throughout the Dissolution Experiments

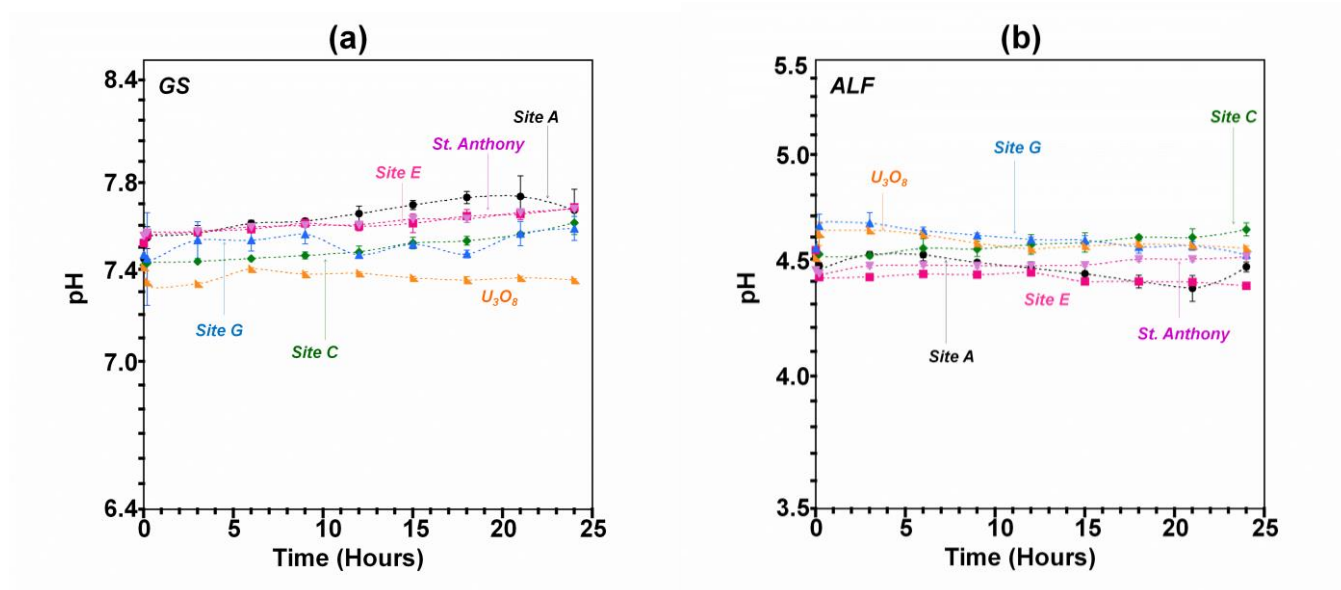


Figure S5: The variation of pH of the solutions (a) GS (b) ALF through the dissolution experiment

Detection of the UO_2^{2+} in Reacted Simulated Lung Fluids

Uranyl-Curcumin-Triton-X System was prepared according to the method described by Zhu et al.³ The calibration standards were prepared using 400 μ M stock uranyl acetate solution prepared in GS and ALF matrices instead of MilliQ water matrix. However, the UV-VIS absorption of two stock solutions made on SLFs was compared with a MilliQ water based stock solution using Evolution 200 UV – Visible Spectrometer to confirm that there is no significant interference from the matrix. The calibration plots are provided in the **Figure S6**.

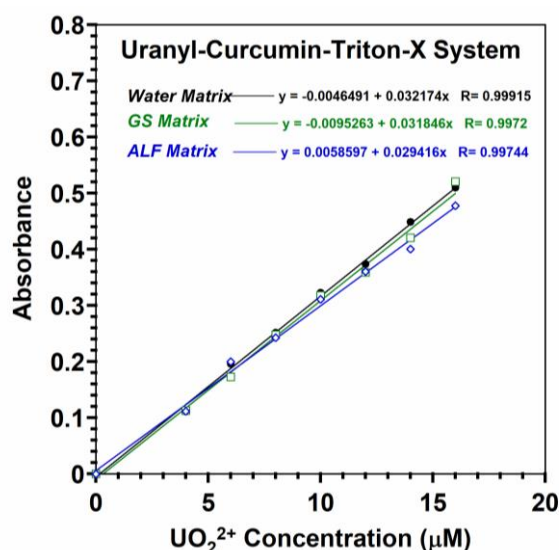


Figure S6: The calibration curve for Uranyl-Curcumin-Triton-X system, UV/ VIS absorption at 430 nm, standard solutions were prepared in MilliQ water, GS and ALF.

Colorimetric analysis qualitatively determined (orange coloration) all the samples contained UO_2^{2+} in the solutions.

Persistence through the Seasons

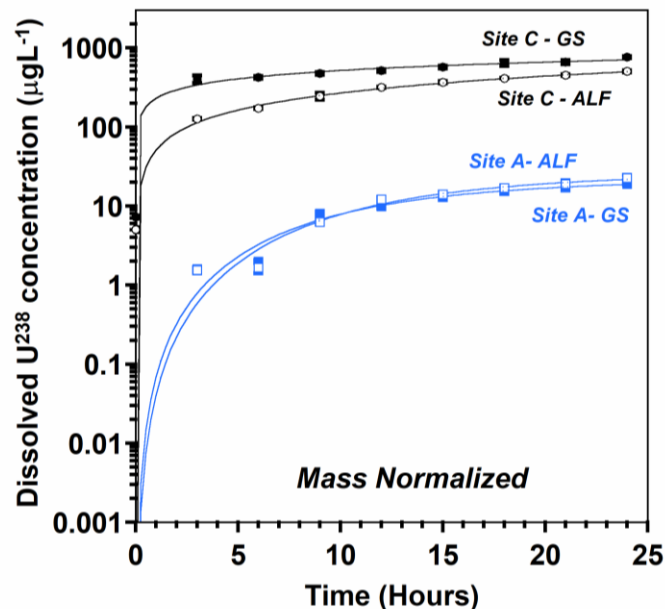


Figure S7: The dissolution of total uranium (TDU) from the two Sites A and C collected during the winter 2017, in the two SLFs as a function of time. Data has been normalized to their respective surface areas (Site A = $2.1 \pm 0.3 \text{ m}^2\text{g}^{-1}$, Site C = $0.90 \pm 0.1 \text{ m}^2\text{g}^{-1}$) and are fitted to Langmuir type model and presented with log scaled Y Axis (Total Dissolved U Concentration $\mu\text{g L}^{-1}$)

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