Mineralogy Dependent Dissolution of Inhaled Uranium in Simulated Lung Fluids in Uranium Mine Lands, New Mexico

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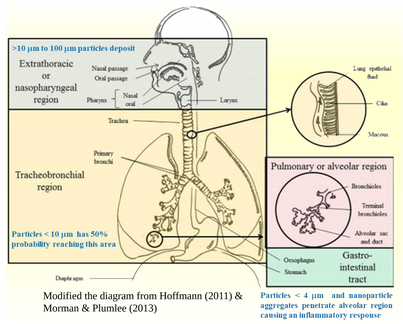
Abstract

The smaller dust particles (PM_{10} or less) can pass through the human respiratory tract, ultimately reaching the lungs. In places where mining occurred, these dusts were long considered health problem due to contamination with heavy metals such as uranium. These metals upon exposure, could either accumulate inside lungs causing irritation and radiation damage or dissolve in lung fluids thereby enter the blood stream, as complexed species. Upon entering blood stream they may excrete or stay complexed with other biological components where they may alter the natural body fluid compositions. The uranium complexation in human biological systems may yield different health conditions depending on several factors, including the mineralogy of the uranium in the source material. In this study, leaching of uranium from (1) dust samples collected around Jackpile mine area, (2) fine-grain sediments from St. Anthony Mine and, a (3) U_3O_8 standard was investigated in two different simulated lung fluids (SLF). The two SLFs mimic two different lung conditions: Gambel's solution (GS) simulates the upper lung conditions with which inhaled dust first interacts, while Artificial Lysosomal Fluid (ALF) mimics the lung conditions at phagocytosis, a defensive mechanism against foreign inhaled bodies. Our results indicate that the dissolution of uranium in dust in these two different SLFs depends not only on the fluid pH and composition but also on the uranium mineralogy of the source material and on the mode of sediment transport. Dust transported via wind demonstrates higher dissolution in GS while dusts and sediment collected around mine pits are more soluble in ALF. The ability of uranium to complex with the organic and inorganic ligands in these lung fluids, for an example, uranium containing clay leach out in the presence of amino acids and ammine complexes, may alter the composition, thereby disturbing body functions. Apart from the experimental studies, geochemical simulations have done using PHREEQC 3.3.8. for these two lung fluid systems with single uranium mineral phase demonstrate differences in extent of uranium dissolution depending on the parent material. Therefore, uranium mineralogy may play an important role in leaching inside the lungs and in subsequent complexation, potentially influencing any resultant health impact.

Introduction

Human exposure to toxic elements (heavy metals, volatile organic compounds, nanoparticles) in the environment can occur via ingestion, inhalation and dermal pathways where the exposure to these contaminated media constitutes a risk for human health. Air pollution is globally considered as a major environmental concern. Particulate matter (PM) is an air pollutant which becomes inhalable as their particle size decreases. As finer fraction can penetrate to the deeper lungs their clearance from the lungs also become difficult. Thus, these dust particles are closely associates with human toxic effects. Areas where mining taken place in the past, has the environment contaminated with heavy metals from groundwater to the windblown dust in the troposphere.

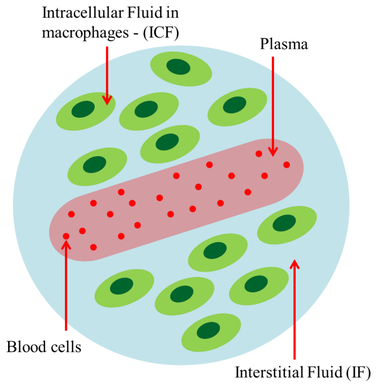


Modified the diagram from Hoffmann (2011) & Morman & Plumlee (2013)

These PM once inhaled may trap in the two compartments; the neutral extracellular environment in the interstitiam of the lungs and acidic environment in the macrophages. During their residency inside the lungs, the metals in these PM chemically interact with the biological components such as anions, proteins, lipids in our body. Thus, these metals may form metal-ligand complexes some of which may be toxic to humans. In current study, we focused on understanding uranium mobility in lungs to better understand its toxicity towards mine workers and residents of these then-mining lands.

Uranium complexes are known to bind with DNA triggering whole slew of protein replication errors, some of may cause cancers independent from uranium radioactive damage. It also induces oxidative stress which leads to a genomic instability. Some studies suggest the death macrophages when exposed to uranyl nitrates. Therefore, uranium induces different toxicity levels depending on its concentration and phase.

Simulated Lung Fluids (SLFs)

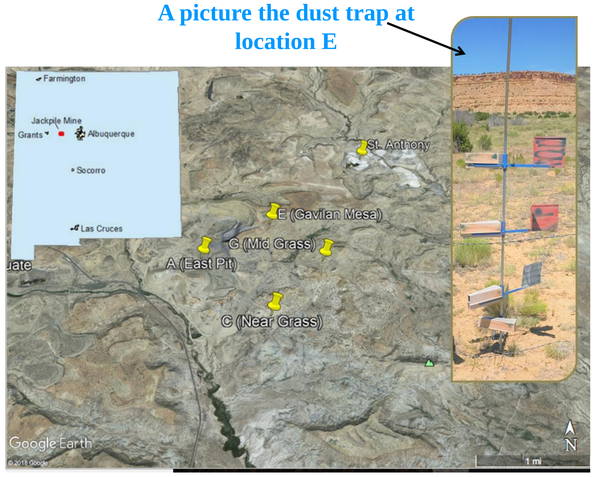


The IF is simulated using Gambel's Solution (GS) and ICF is simulated with Artificial Lysosomal Fluid (ALF).

Composition (g·L^{-1})	Gamble	ALF
NaCl	6.779	3.21
Na_2HPO_4		0.071
$NaHCO_3$	2.268	
Trisodium citrate dihydrate	0.055	0.077
NH_4Cl	0.535	
Glycine	0.375	0.059
NaH_2PO_4	1.872	
L-cysteine	0.121	
NaOH		6.0
Citric acid		20.8
$CaCl_2{\cdot}2H_2O$	0.026	0.128
Na_2SO_4		0.039
$MgCl_2{\cdot}6H_2O$		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

Sample Locations & Collection

A picture the dust trap at location E

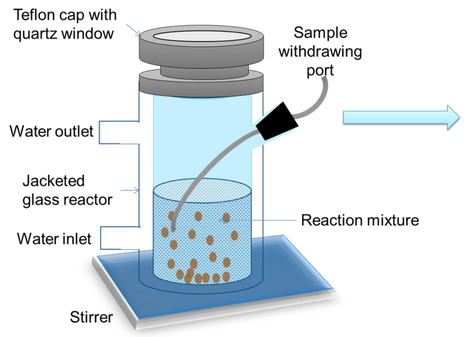


Dust and sediments were collected around Jackpile Mine and St. Anthony Mine. Dusts collected at1.5m above the surface has used in lung fluid study.

Characterization

Sample	7 points N_2 BET Surface Area (m^2/g)	%U
U_3O_8	0.50±0.04	99
St. Anthony Sediment	1.6±0.1	0.9
Site A	2.1±0.1	0.23
Site E	14.5±1.0	0.18
Site C	0.8±0.1	0.14
Site G	1.8±0.6	0.23

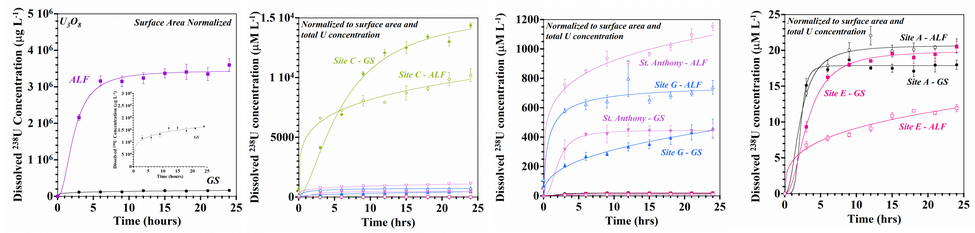
Dissolution of Uranium in Simulated Lung Fluids



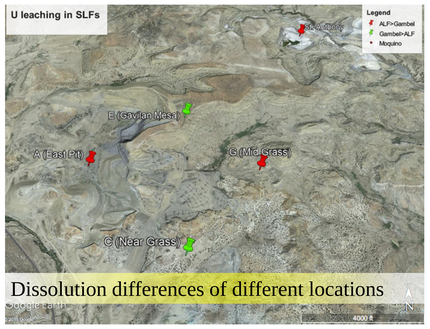
ICP – MS Analysis

Maintained at 37 °C

Results & Discussion



Sample	Initial pseudo-first order rates of U dissolution (μg L^{-1}m^{-2}h^{-1})		%U dissolved in SLF upon 24 hour exposure	
	GS	ALF	GS	ALF
U_3O_8	3.9E^4±1120	1.05E^6±12101	0.7	16.2
St Anthony	104±3	233±6	0.7	1.8
Site A	5.0±0.1	4.6±0.2	0.036	0.043
Site C	1274±21	2017±36	11.0	8.0
Site E	3.2±0.2	2.0±0.2	0.3	0.2
Site G	63±1	184±5	1.0	1.4



Dissolution differences of different locations

Due to low pH and high anionic ligand concentration, ALF is expected to show higher U dissolutions than in GS. However, dusts from **site C** and **E** dissolve more in GS than ALF, with site C dust samples having higher leaching percentages.

Possible factors affecting dissolution differences

1. Mineralogical differences of both uranium containing minerals and other existing minerals

XRD analysis indicate microcline and kaolinite rich chemistry in the samples from site A, G and St. Anthony while no microcline or kaolinite was identified in either of site E and C samples.

2. Extent of weathering and modes of transport

Uranium mineralogy and the dissolution differences in SLFs – PHREEQC 3.3.8

Mineral/ Uranium phase	U Ox State	Max U dissolution (M) ALF	Gambel	ALF/Gambel
U_3O_8	5, 6	3.101E-02	2.999E-02	1.0337
Uraninite (UO_2)	4	1.000E-02	9.997E-03	1.0003
Carnotite ($K_2O.2UO_3.V_2O_5.nH_2O$)	6	4.044E-04	6.513E-03	0.0621
Tyuyamunite ($CaO.2UO_3.V_2O_5.nH_2O$)	6	4.052E-04	7.207E-03	0.0562
UO_2HPO_4	4	1.000E-02	9.998E-03	1.0002
Uranocircite ($((UO_2)_2(PO_4)_2)$)	4	4.456E-03	1.595E-02	0.2794
Uramphite ($(NH_4)(UO_2)(PO_4)3H_2O$)	4	2.000E-02	1.999E-02	1.0005

Some uranium bearing minerals are theoretically more soluble in GS than ALF. Therefore, the differences seen in bench experiments might have influenced by the sample mineralogy.

-Calculated using a modified Minteq database

Conclusion

Uranium in dust around mine lands may be inhalable. Upon inhalation, these dust can solubilize in deep lung fluids where their extents of dissolution depends on number of factors. One such is the mineralogical properties of inhaled dust. Some uranium phases may dissolve in interstitial conditions of lung while some will dissolve during phagocytosis. In either way, these uranium bearing dust posses the risk of entering blood stream of individuals exposed as mobile species that bear the ability to bind and complex with biological components such as DNA, protein.

Acknowledgement

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