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Demonstration of the Aged Effects of Sorbed Plutonium Complexes on Savannah River Site Sediments

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Relevant Background

There have been many studies in the past noting that the reversibility of sorption for actinide elements decreases as the aged complexes are formed (Kaplan et al., 2004; Morsania et al., 2004; Tamcik, et al., 2011). This leads to theoretical problems as equilibrium Kd values partition coefficients are assumed to be linear in order to verify that and are often used in transport and risk assessment models. In addition, further studies have suggested that geochemical constituents like natural organic matter (NOM) can play a role in mobilizing actinides (McCarthy, Czerwinski, et al., 1998; McCarthy, Serkiz, et al., 1998).

The sediments used in these experiments were core materials remaining from an 11 year lysimeter experiment completed at the Savannah River Site 21 years ago (McCarthy, Czerwinski, et al., 1998) and after aging for 32 years on the sediments. The selective extractions may be significantly different from those measured previously at the Savannah River Site for plutonium aged for 32 days in close sediment.

Objectives

The objective for the study was to determine the effect of aging Pu complexes on Savannah River Site sediment through desorption and selective iron extraction experiments using dual isotopes. In the presence of a variety of organic and inorganic ligands to demonstrate changes in freshly sorbed plutonium versus aged plutonium complexes.

Experimental Methods

Measurement of Desorption Kd's for Plutonium on Savannah River Site Sediment after 32 years in the Presence of a Variety of Ligands

Desorption Experiments

Desorption experiments were evaluated with batch desorption experiments over 7 and 28 days in the presence of a variety of organic and inorganic ligands. Kd values were compared with those measured previously at the Savannah River Site for plutonium aged for 32 days in close sediment.

Table 1: Summary of Results of Desorption Experiments

<table>
<thead>
<tr>
<th>Ligand</th>
<th>Kd (mL/g)</th>
<th>7 day</th>
<th>28 day</th>
</tr>
</thead>
<tbody>
<tr>
<td>NaCl, CaCl2, Na2PO4, or NaF at 10 mM</td>
<td>5100 ± 4000</td>
<td>6000 ± 5000</td>
<td></td>
</tr>
<tr>
<td>NaCl, CaCl2, Na2PO4, or NaF at 100 mM</td>
<td>5100 ± 4000</td>
<td>6000 ± 5000</td>
<td></td>
</tr>
<tr>
<td>NaCl, CaCl2, Na2PO4, or NaF at 1000 mM</td>
<td>5100 ± 4000</td>
<td>6000 ± 5000</td>
<td></td>
</tr>
<tr>
<td>NaCl, CaCl2, Na2PO4, or NaF at 10,000 mM</td>
<td>5100 ± 4000</td>
<td>6000 ± 5000</td>
<td></td>
</tr>
</tbody>
</table>

Selective Iron Extractions

Selective iron extractions were performed following a 3-day dual isotope sorption (239/240Pu and 238Pu) experiment. Samples were prepared in duplicate with each of the highest concentration of each ligands mentioned with one being analyzed for total iron and one for amorphous iron. Amorphous phases were analyzed for both iron and as described below for the determination of solids for extractions.

Table 1: Characteristics of SR Site Sediment Used in this Work

<table>
<thead>
<tr>
<th>Characteristics</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>5.1</td>
</tr>
<tr>
<td>Solids</td>
<td>110 g</td>
</tr>
<tr>
<td>Organic Carbon</td>
<td>9.0</td>
</tr>
<tr>
<td>Total Carbon</td>
<td>15.3</td>
</tr>
<tr>
<td>Nitrogen</td>
<td>0.05</td>
</tr>
<tr>
<td>C/N</td>
<td>299</td>
</tr>
</tbody>
</table>

Results: Batch Sorption/Desorption Experiments

Figure 2: Results of selective extractions for total iron and amorphous iron for 239/240Pu at 4 µg/g aged for 32 years on SRS sediment

Results: Selective Extractions

Figure 3: Results of selective extractions for total iron and amorphous iron for 239/240Pu at 4 µg/g aged for 32 years on SRS sediment

Results: Alpha Spectroscopy

Figure 5: Results of alpha spectroscopy of aqueous end 32 year aged plutonium complexes from Savannah River Site sediment

Summary and Implications

Desorption Experiment

Desorption Kd values were not statistically different for different ligands and different concentrations after plutonium complexes aged for 53 years on SRS sediments. Kd values were not corroborated in changes in concentration of ligands.

Selective Iron Extractions

Selective iron extractions demonstrated a significant fraction of plutonium associated with the iron phases. 8.2% of the 239/240Pu (14.8% with the amorphous iron oxides) and 5.4% with the crystalline iron oxides. 94.4% of the 239/240Pu being associated with the total fine fraction of the soil.

Further Investigation

Further investigation may be necessary to analyze by nssSIMS to confirm that plutonium is strongly associated with iron oxides within the sediments. nssSIMS analysis of the secondary ion mass sampled is analyzed by electron microscopy by mass spectrometry. The method will be capable of measuring iron and plutonium co-extracted down to ppb levels.

References


Acknowledgements

Thanks to Daniel Kaplan at the Savannah River Site for his aide in the formulation of the experimental protocol.