Biogeochemical mechanisms controlling Pu on environmental timescales

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Mechanisms that influence plutonium (Pu) migration include redox transformations, colloid-facilitated transport, sorption/desorption rates, solubility, interactions with organic matter and bacteria. Humans have produced approximately 2,700 metric tons of Pu worldwide. It is estimated that 1% (or 27,000 kg) has been released into the environment where subsequently low-level transport in the surface and subsurface has occurred. Due to its long half-life (^{239}Pu $t_{1/2}$ ~24,000 yrs) and high toxicity, Pu will persist in the environment for a long time. Efforts to predict its behavior have been undertaken where a more robust conceptual model is emerging. We have preformed a series of laboratory experiments investigating the behavior of Pu in the presence of clay minerals. One year sorption of Pu(V) on montmorillonite showed a continued uptake, and only after months was the extent of Pu(V) sorption similar to Pu(IV)—documenting slow surface-mediated reduction of Pu(V) to Pu(IV). Pu desorption from montmorillonite was investigated as a function of time, and pH. At pH 8, significantly less Pu was desorbed from samples pre-sorbed for 6 months compared to 12-day sorption, consistent with a surface aging of Pu. A third series of experiments explored the reduction of Pu(V) to Pu(IV) on clay minerals with varying structural Fe. At pH 8, the rate of reduction of Pu(V) to Pu(IV) was strongly Fe dependent; yet at long-time scales the extent of sorpiton was independent of Fe. Lastly, desorption experiments were preformed on hydrothermally altered nuclear glass with trace Pu. In the 140ºC colloid sample, smectite was formed and the desorption rates match that of montmorillonite above. In contrast, the 200ºC colloids samples consisting of smectite and zeolites, exhibited lower desorption rates suggesting higher temperatures may lead to a more stable structural assoication with Pu. These experiments highlight the need for long-term studies to understand Pu behvaior on environmental timescales (decades) and elucidate the biogeochemical mechanisms that ultimately control its sorption rate and extent as well as its stability in the environment.