Adsorption and Desorption of Plutonium on Na-montmorillonite over a Ten Order of Magnitude Range in Concentration

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Plutonium (Pu) release to the environment through nuclear weapon development and the nuclear fuel cycle is an unfortunate legacy of the nuclear age. In part due to public health concerns over the risk of Pu contamination of drinking water, predicting the behavior of Pu in both surface and sub-surface waters is a topic of continued interest. It has been suggested that the affinity of Pu for mineral surfaces is significantly greater at low concentrations due to the presence of minor amounts of "high affinity" sorption sites (surface defects, heterogeneity, etc.). Hence the current practice of extrapolating the behavior of Pu at environmental concentrations (10^{-12} to 10^{-16} M), from the results of laboratory experiments which are typically performed at concentrations $>10^{-10}$ M, may not be valid.

Here we present results from batch adsorption experiments performed with Na-montmorillonite in pH 8 background electrolyte spiked with Pu(V) and Pu(IV) at initial concentrations ranging from 10^{-6} – 10^{-16} M. After 30 day's equilibration, Pu concentrations in the supernatant were determined using a variety of analytical techniques: liquid scintillation counting and quadrupole ICP-MS, NuPlasma HR MC-ICP-MS, and accelerator mass spectrometry. The resulting sorption isotherm was found to be largely linear indicating a similar Pu adsorption affinity for montmorillonite over ten orders of magnitude Pu concentration range (10^{-6} to 10^{-16} M). Importantly, these results suggest that, under the conditions of the experimental system, Pu(V) adsorption behavior at typical environmental concentrations (femtomolar) is not significantly different to that at typical laboratory concentrations (nanomolar to micromolar). While Pu affinity for the montmorillonite surface appears to be linear over this concentration range, desorption kinetics may still play an important role in the environmental mobility of Pu. Accordingly, we are investigating the stability of Pu associated with clay surfaces in a series of desorption experiments using flow-cells. The results of these experiments suggest that significant amounts of Pu adsorbed on the surface of clay particles can be desorbed. Modeling of the data is providing insight into the mechanisms controlling the observed environmental colloid-facilitated transport of plutonium. Quantification of desorption kinetics is critical to parameterizing colloid-facilited transport of Pu in the environment.