

Plutonium desorption from mineral surfaces caused by environmental concentrations of hydrogen peroxide

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Knowledge of Pu adsorption and desorption interactions with mineral phases is crucial for understanding its environmental mobility. Here we demonstrate that environmental concentrations of H_2O_2 can affect the stability of Pu adsorbed to goethite, montmorillonite and quartz across a wide range of pH values. In batch experiments where Pu(IV) was adsorbed to goethite for 21 days at pH 4, 6, and 8, the addition of 5 – 500 μM H_2O_2 resulted in significant Pu desorption. At pH 6 and 8 this desorption was transient with re-adsorption of the Pu to goethite after 30 days. At pH 4, no Pu re-adsorption was observed. Experiments with both quartz and montmorillonite at 5 μM H_2O_2 desorbed far less Pu than in the goethite experiments highlighting the contribution of Fe redox couples in controlling Pu desorption at low H_2O_2 concentrations. Plutonium(IV) adsorbed to quartz, and subsequently spiked with 500 μM H_2O_2 also resulted in significant desorption of Pu, demonstrating the complexity of the desorption process. Our results provide the first evidence of H_2O_2 catalyzed desorption of Pu(IV) from mineral surfaces. We suggest that this reaction pathway coupled with environmental levels of hydrogen peroxide may contribute to Pu mobility in the environment.

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