



Cesium adsorption and desorption in Clay Minerals: Effects of Natural Organic Matter and pH

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Cesium (Cs) released into the environment poses significant environmental concerns and remediation challenges and a majority of Cs in the environment have remained within clay minerals of the surface soils due to the strong adsorption affinity of Cs. Different clay minerals have different bonding sites, resulting in various adsorption mechanisms. In addition, natural organic matter (NOM) can interact with clay minerals. In this work, three clay minerals (illite, vermiculite, montmorillonite) are used to investigate the adsorption and desorption of Cs over a range of Cs concentrations with and without humic acid (HA). For desorption, sodium chloride, ammonium chloride, and acetate acid are sequentially used. Comparison of Cs adsorption shows that vermiculite has much stronger affinity than other clay minerals, which can be attributed to multiple adsorption sites including interlayer site, type II site, and planar site with higher sorption capacities. Montmorillonite has slightly higher than illite, probably due to higher bounding water content. The impact of HA concentration on Cs adsorption was clay mineral-dependent and depending upon initial Cs concentrations. For low Cs concentrations, the HA effect on the adsorption for all three clay minerals was not significant. For high Cs concentrations, a high HA concentration (400 mg/L) decreased the adsorption for Illite and Vermiculite.

For montmorillonite the desorbed Cs mass was much higher than illite and vermiculite, indicating that Cs mostly sorbed the outer complex and partly bounding water. For illite, HA increased the desorption rate, indicating that a slight portion of Cs may be sorbed into HA matrix. For vermiculite, the HA addition slightly decreased the desorption rate. The Cs desorption in vermiculite was extremely low. For high Cs concentration cases in illite and Montmorillonite, the sequential desorption results show higher desorption rates in the high Cs concentration than in the low concentration. These results may suggest that Cs tends to occupy more reversible sites rather than irreversible sites. For vermiculite, the results are not different much. Due to the strong affinity of Cs into vermiculite, regardless of the initial Cs dose the sequential desorption efficiency was very low. High sodium concentration increased the desorption from all three minerals. However, the desorption rate with NH_4Cl was higher for cases with a low sodium concentration. A total desorption rate combined with sodium and NH_4Cl was similar for most of cases, indicating that the sum of adsorbed Cs on weak and strong exchangeable sites are similar. Key observations from batch testing were also confirmed by extended X-ray absorption fine structure spectroscopy (EXAFS) and TEM imaging. The expandability of clay minerals and effect of HA addition on Cs adsorption and desorption are highlighted to address the efficiency of Cs removal schemes from contaminated soils.

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