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Adsorption and Desorption of Cesium in Clay Minerals: Effects of Natural Organic Matter and pH

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Cesium (Cs) released into the environment (e.g., Fukushima accident) poses significant environmental concerns and remediation challenges. A majority of Cs in the environment have remained within the surface soils due to the strong adsorption affinity of Cs towards clay minerals. Different clay minerals have different bonding sites, resulting in various adsorption mechanisms at nanometer scale. For example, the illite commonly has a basal spacing of \sim 1.0 nm, but becomes wider to 1.4 nm once other cations exchange with K in the interlayer site. Cs adsorbs into these expanded wedged zone strongly, which can control its mobility in the environment. In addition, natural organic matter (NOM) in the surface soils can interact with clay minerals, which can modify the mechanisms of Cs adsorption on the clay minerals by blocking specific adsorption sites and/or providing Cs adsorption sites on NOM surface. In this work, three representative clay minerals (illite, vermiculite, montmorillonite) and humic acid (HA) are used to systematically investigate the adsorption and desorption behavior of Cs. We performed batch adsorption experiments over a range of Cs concentrations on three clay minerals with and without HA, followed by sequential desorption batch testing. We tested desorption efficiency as a function of initial adsorbed Cs concentration, HA content, sodium concentration, and pH. The sequential extraction results are compared to the structural changes in clay minerals, measured using extended X-ray absorption fine structure spectroscopy (EXAFS) and aberration-corrected (scanning) transmission electron microscopy (TEM) – energy dispersive X-ray spectroscopy (EDX). Hence, this work aims to identify the mechanisms of Cs fixation at the nanometer (or atomic-) scale as a function of the clay mineral properties (e.g. expandability, permanent surface charge) and varying organic matter content at different pH values and to enhance our atomic-scale mechanistic understanding of the clay mineral interactions with cesium in the presence of NOM. 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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