Cesium occurrence and the stability in Fukushima soils

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In Fukushima, radioactivity still remains in the surface environment, which derives from ^{137}Cs ($T_{1/2}$ = 30.07 y) and ^{134}Cs ($T_{1/2}$ = 2.062 y). The remaining radioactivity in the soils has been attributed to the strong binding of radioactive Cs to clay minerals. We have investigated the details of the Cs occurrence and chemical form combined with the mineralogy, chemical composition and microstructure, in order to understand the property of the Cs-bound clay minerals, the size dependence and the stability.

20 soil samples were collected from the highly contaminated zone elongated towards northwest from the Fukushima Daiichi power plant. The samples were sieved with 2 mm mesh. The powder X-ray diffraction (XRD) analysis was conducted on the untreated sample and elutriated (<1 micron) at untreated, etylene glycol treated and heated conditions. Sequential extraction was carried out to fractionate Cs to water soluble, ion exchangeable, carbonate bound, iron oxides bound, organic matter bound, and residue.

The XRD analysis revealed that major common minerals are composed of quartz, feldspar, and 1.4 nm-layered silicate. A major minerals in the elutriated samples (<1 micron) are sheet silicates with 1.4 nm and 1.0 nm d-spacing, which were subsequently characterized as smectite, chlorite, and muscovite (illite). In the size fractionation experiment, ~78% of ^{137}Cs was distributed in the <1 μm. The sequential extraction revealed that the radioactivity of each chemical fraction increases as the total radioactivity increases; however, ~90% of Cs is bound to the residue in the all soil samples, which represents unleachable Cs from interlayers of clay minerals. These results suggest an importance of fine clay minerals as a career of radioactive Cs. Although the size of the clay minerals is within the size range of colloid, the mobility of the submicron-sized clay minerals within soils must be minimum by aggregation and filtration effects, as ~98% of Cs still remains at the top ~5 cm in the vertical profile.