

Chemical species of radiocesium in the contaminated soil, and contribution of non-mica minerals to tight association

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The accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP) occurred as a consequence of the massive earthquake and associated tsunami that struck the Tohoku and north Kanto regions of Japan on 11 March 2011. A series of hydrogen explosion was occurred from 13 March to 15 March at the units 1, 2, and 3. The release rate of ¹³⁷Cs on 15 March is estimated between 10¹² and 10¹⁵ Bq/h [1]. This fallout radiocesium were dispersed from FDNPP to the ocean [1,2] and land [1]. Some of the released radiocesium were deposited on the ground of the area located north-west direction from FDNPP. The spatial concentration distribution and depth profiles of radiocesium were measured to estimate a dose rate and the fate in the terrestrial environment.

For the estimation of migration of the fallout radiocesium, sorption behavior of the deposited radiocesium should be clarified. Cesium at low concentration is tightly sorbed on mica like minerals. This knowledge suggests that some amounts of fallout radiocesium are tightly sorbed with the contaminated soils when the soils contain mica like minerals. However, effects of the other minerals than mica like minerals on the sorption of radiocesium are scarcely known.

We have conducted the analyses of chemical species of radiocesium in the contaminated soils sampled in Fukushima by the desorption experiments using appropriate reagents solutions and size fractionation of the contaminated soils [3]. Three different soil samples were collected in Iitate, Fukushima on May, 2011. The soil samples were treated by a 1 mole L⁻¹ NH₄Cl solution and a 1 mole L⁻¹ CH₃COOH solution to dissolve mobile fraction of radiocesium. After the treatment the soil samples were analyzed by XRD to determine minerals and clay minerals in soil.

More than 65% of radiocesium were remained in the residual fraction of the soil samples after treatment of a 1 mole L⁻¹ NH₄Cl solution and a 1 mole L⁻¹ CH₃COOH solution, indicating that 35% of radiocesium was mobile and the residual fraction was tightly associated with the soil. The XRD analysis of the soil showed that two soil samples contained mica like minerals, and one soil sample did not. These results strongly suggest that in those Iitate soils the radiocesium was tightly associated not only with mica like minerals but also with non-mica like minerals.

In order to clarify the minerals that tightly associate radiocesium, we have conducted the sorption and desorption experiments of radiocesium on 17 minerals, and have found that some fractions of radiocesium were tightly associated with natural kaolinite and manganese

oxides other than illite [4]. There are several association sites in minerals for radiocesium. In natural environment, minerals are altered by weathering to generate the tight association sites in the altered minerals. Unfortunately the number of the tight association sites is very limited in the minerals of kaolinite and manganese oxides. Since the chemical concentration of radiocesium was extremely low, most of radiocesium are easily bound in the tight association sites (Fig. 2).

This finding of tight association of radiocesium on the non-mica minerals inform us the migration behaviour of radiocesium in environments.

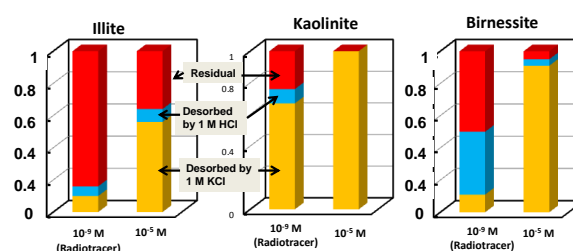


Fig.1 Desorbed fraction of radiocesium from minerals at different cesium concentrations. When Cs concentration is very low of 10⁻⁹ M, higher amounts of Cs was resided after the treatment with desorption reagents solution.

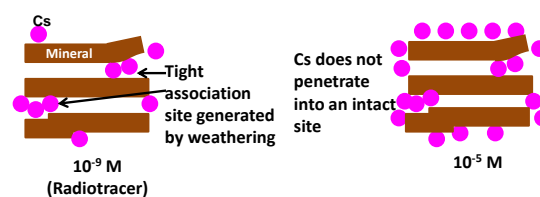


Fig.2 Schematic diagram of adsorption of radiocesium on weathered minerals. Tight association sites for radiocesium were generated by weathering. Even though the amounts of the tight association sites were limited, higher amounts of radiocesium was adsorbed on the tight association sites because of low concentration of radiotracer.

References

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