

## Evaluation of the migration of radiocesium based on chemical speciation

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Chemical form of radiocesium is fundamental information for evaluation of its migration in the environment. After the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, we analyzed <sup>137</sup>Cs in aerosols, rock, soil, leaves, river suspended sediment and river water collected in Fukushima. Here, we review the migration of radiocesium in the environment based on our up-to-date data.

Many particles with high radioactivity were found in aerosols collected in March, 2011, where 50% to 90% of radiocesium was water-soluble. This means that radiocesium was still present mostly in a water-soluble fraction of aerosols before deposition and just after deposition on the ground. However, it was found that little amount of radiocesium was contained in a soluble fraction in soil and weathered rock samples by leaching experiments with water at various pH conditions. Possibly, such a soluble fraction of radiocesium was strongly fixed on rock and soil particles after dissolution in water (e.g. rainfall) on the ground. At the moment, chemical species of radiocesium would have changed from soluble to insoluble form. This strong fixation of radiocesium in soils can be explained by formation of inner-sphere complex in phyllosilicate minerals of clay minerals, which was confirmed by extended X-ray absorption fine structure (EXAFS) analysis. Field-scale observation reflected well the strong adsorption of radiocesium because most of the radiocesium stayed within 5 cm from the surface in soil layers.

In particular, in river and ocean systems, whether radiocesium is particulate or dissolved form is closely related to uptake by organisms and incorporation into food chain in ecosystems. We have monitored radiocesium concentrations in the Abukuma River system since summer in 2011. Total <sup>137</sup>Cs concentration in river water including both dissolved and particulate fractions decreased drastically from summer to winter in 2011, and then gradually decreased with time except at heavy rainfall events. From the strong fixation of radiocesium on soil particles, it was expected that radiocesium was predominant in particulate matter in river systems. More than 70% of radiocesium was particulate form, where the contribution of silt size (3 ~ 63 μm) fraction was the largest. However, radiocesium in dissolved fraction suggested an increase at estuary. This implies desorption of radiocesium from particulate matter because of an increase in salinity.

We made adsorption experiments to determine distribution coefficient,  $K_d$ , between fluvial sediment and river water, and further desorption experiments to examine the reversibility of adsorption-desorption process.  $K_d$  values determined by adsorption and desorption experiments were consistent, indicating that radiocesium adsorption was a reversible process. In addition, when artificial seawater was used for desorption experiment, the resulting  $K_d$  value was lower than that obtained using river water. This clearly demonstrated the influence of ionic strength on adsorption-desorption process through competition of cesium ions with other ions (e.g.,  $K^+$ ,  $Na^+$  and  $Ca^{2+}$ ), which is consistent with the field observation as noted above. Furthermore, we applied generalized adsorption model (GAM) to predict the distribution of radiocesium between particulate matter and water in the Abukuma River system. As a result, it was demonstrated that GAM can predict the apparent  $K_d$  values calculated from <sup>137</sup>Cs concentrations in fluvial sediment and river water as well as lower  $K_d$  values at estuary.

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