

Factors controlling radiocesium distribution in river sediments: Field and laboratory studies after the Fukushima Dai-ichi Nuclear Power Plant accident

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After the accident of the Fukushima Dai-ichi nuclear power plants, the distribution of radiocesium (RCs) in river system has been of important issue related to its transportation in environment. In this study, the distribution coefficient (K_d) of ¹³⁷Cs in river sediment sampled from the Abukuma and Kuchibuto Rivers was determined to explore particle-size dependence of RCs distribution combined with laboratorial estimations. The results showed that the K_d patterns of ¹³⁷Cs (= particle size dependence of K_d) were not only related to the particle size for larger particles (with small content of clay mineral), but also dependent on the content of clay mineral for smaller particles (with high content of clay mineral), sampling site, and especially cation concentration in the aqueous phase. In contrast, there was no obvious particle-size dependence in the K_d patterns of stable cesium (= ¹³³Cs).

In order to eliminate the contradiction of natural and laboratorial distributions of ¹³⁷Cs on sediments with various particle sizes, the adsorption species of Cs was explored by EXAFS. Based on the analysis recorded at BL-9A and BL-12C in Photon Factory, $k^3\chi(k)$ function and radial structure functions (RSFs) for Cs adsorbed on river sediments with various particle size were obtained. Here, CsNO₃ solution and vermiculite were also used as two end members, i.e., outer-sphere (OS) and inner-sphere (IS) complexes, because Fan et al. (2014) have confirmed almost pure IS complex for Cs adsorption on vermiculite. The $k^3\chi(k)$ function of Cs adsorbed on sediments exhibited some obvious differences in oscillatory frequencies and amplitude from CsNO₃ solution, which testified that a new local atomic structure of Cs after adsorbed on sediment. A small beat was observed at 4.2 Å⁻¹ in k space for Cs adsorbed sediments with different particle size, and very similar to vermiculite which has characteristic oscillation of IS complex. It is interesting that no obvious difference was observed between the particle sizes of 63-125 μm and 500-850 μm, which suggested that similar adsorption species were formed for Cs in sediments with different particle size.

Correspondingly, radial structural function showed that two shells were observed for Cs adsorbed on river sediments and vermiculite in the R ranges of 2.2-2.8 Å and 3.6-3.8 Å, respectively. In reality, the characteristic

shells can be attributed to OS and IS complexes, respectively (Qin et al., 2012; Fan et al., 2014). Similar intensities of OS and IS complex shells suggested the similar composition of OS and IS complexes as Cs species adsorbed on the sediments. In fact, the coordination ratio of CN_{IS}/CN_{OS} is more sensitive and reliable to the variation in the adsorption species of Cs. The largest CN_{IS}/CN_{OS} for Cs adsorption on vermiculite (~4.6) suggested that IS complex was dominant species. A smaller ratio of CN_{IS}/CN_{OS} was observed for Cs adsorption on sediment at 63-125 μm and 500-850 μm, respectively. The ratios of CN were around 1.0 and 2.0 for sites A and C, respectively. This fact suggested that both IS and OS complexes contributed to Cs species adsorbed on river sediments. More importantly, there is no obvious particle-size dependence for Cs species in sediments with different particle sizes, which is consistent with the distribution profile of stable cesium and of K_d without having clear particle-size dependence.

Our findings indicated that river sediment has high fixation ability to ¹³⁷Cs. However, the adsorbed ¹³⁷Cs can be extracted from sediments to water phase when salinity becomes high in water such as in seawater. In particular, the distribution patterns of ¹³⁷Cs at various particle sizes can suffer a slow equilibrium process of adsorption of ¹³⁷Cs during the transportation of river sediments, and should eventually behave similarly to stable cesium in the river system, if the contribution of cesium within 3D crystal structure of some minerals is minimal. Therefore, the particle size of sediments is a primarily important factor to RCs distribution at the early stage of its deposition, but finally Cs adsorbed on sediment will be equilibrated with the stable cesium during its transportation in the river system. The controlling factor such as the mineralogy in sediments will possibly make the distribution patterns of RCs similar to that of stable cesium at longer-term.

References

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