Difference in the Water Solubility of Radiocesium between Fukushima and Chernobyl Areas

The adsorption of radiocesium (RCs) on particulate matter (PM) in aquatic environments is important to understand its mobility and bioavailability. We have found that the distribution of RCs in the aqueous phase relative to PM in the Pripyat River (Chernobyl) is very high compared with that in the Kuchibuto River (Fukushima) as a consequence of differences in the Cs affinity for PM in the two rivers based on Cs speciation and organic matter-clay mineral interaction. Our results included the following: (i) extended X-ray absorption fine structure (EXAFS) spectroscopy showed that the contribution of outer-sphere (OS) complex of Cs on PM is larger in Chernobyl than in Fukushima and (ii) scanning transmission X-ray microscopy (STXM) revealed a larger association of humic substances and clay minerals in Chernobyl partly due to high Ca^{2+} in the Pripyat River.

A large amount of radiocesium (RCs) dispersed by the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident and deposited in the Fukushima area has migrated through river water on land [1]. In aquatic environments such as in river water, the biggest concern is the accumulation of RCs in fish through the food web because of the importance of fish in the human diet [2]. Such accumulation in the food web begins with the uptake of RCs by aquatic microorganisms such as planktons, and the amount of intake is controlled by the dissolved fraction of RCs. Thus, the solid-water partition of RCs in river water is of particular importance in aquatic environments.

Various studies have shown that RCs in river water in the Fukushima area are highly insoluble in terms of solid-water partition: more than 70% of RCs (i.e., dissolved fraction < 30%) are strongly adsorbed on PM in river water primarily due to the high affinity of Cs for phyllosilicate minerals consisting of vermiculite-like structures [3, 4]. This characteristic is reasonably interpreted by the formation of inner-sphere (IS) complex of Cs into the phyllosilicates abundant in the Fukushima area, as shown by EXAFS at the Cs L_{III}-edge measured at BL-9A [spectra (iv), (v), and (vi) in Fig. 1]. Its appearance is caused by the presence of oxygen and silicon in the siloxane layer as the second neighboring atoms. The PM in river water contained such phyllosilicates, resulting in the increase of the adsorbed fraction in the Kuchibuto River in Fukushima.

By contrast, the opposite case was observed in the Pripyat River in the Dnieper River reservoir system near Chernobyl in relation to the Chernobyl Nuclear Power Plant (CNPP) accident in 1986. Within two years after the accident, more than 80% of RCs existed in dissolved form in the Pripyat River [2, 5]. The contrasting behavior of RCs between Chernobyl and Fukushima is important in interpreting the subsequent migration of RCs and their incorporation into the food web. Thus, we investigated the origin of different solid-water partitions of RCs in the Pripyat and Kuchibuto Rivers in the Chernobyl and Fukushima areas, respectively [6]. A plausible explanation for the difference is the mineral composition of the PM related to the adsorption-desorption reactions. A further possible explanation is the effect of natural organic matter (NOM) which is abundant in the Chernobyl area supplied from peat wetland, the predominant soil type in this area. It has been suggested that RCs in the organic soil area are more mobile in the Pripyat catchment area, since NOM lowers the affinity of RCs for soil particles. This fact is consistent with our findings obtained by EXAFS (Fig. 1), which demonstrated that the NOM largely inhibits Cs adsorption to PM from the Pripyat River [7]. The EXAFS results revealed that IS complex is not the dominant species for the original PM (spectrum (ii)), but the IS complex fraction increased after the removal of NOM from PM by treatment with H_2O_2 (spectrum (iii)). This fact showed that the PM has the potential to form IS species, but the presence of NOM inhibits its formation and increases the OS complex, which leads to the large distribution of RCs in the aqueous phase in Chernobyl.

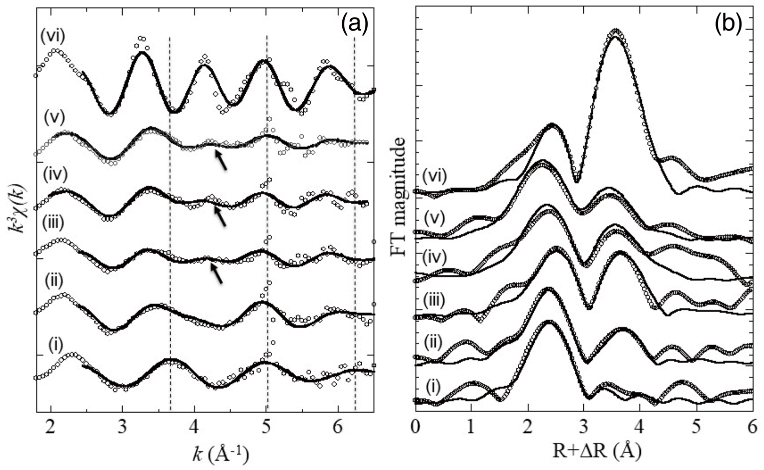

Figure 1: Cs L_{III}-edge EXAFS in (a) k and (b) R spaces. (i): hydrated Cs^+ in water; (vi): Cs adsorbed on vermiculite; (ii) and (iii): Cs adsorbed on particulate matter in the Pripyat River before and after the removal of NOM, respectively; (iv) and (v): Cs adsorbed on PM in the Kuchibuto River before and after the removal of NOM, respectively.

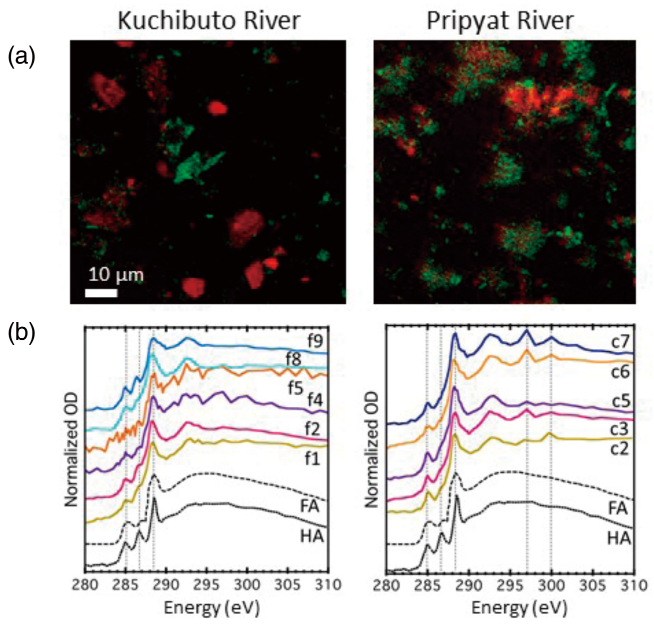

Figure 2: Results of STXM analyses. (a) Distributions of Al and C in PM in the Kuchibuto and Pripyat Rivers. (b) Carbon K-edge XANES spectra at various carbon-rich spots in PM. Spectra of fulvic (FA; Suwannee River (SR) fulvic acid) and humic acids (HA; SR humic acid) are shown for reference.

In this study, the association of phyllosilicates and NOM was also confirmed by STXM developed at BL-13A to obtain more direct evidence of the effect of NOM. As a result, it was found that the distribution of NOM represented by the C signal in PM is strongly correlated with Al-bearing particles collected from the Pripyat River, demonstrating the strong affinity between NOM and clay minerals in the Pripyat River, but this is not the case in Fukushima [(a) in Fig. 2]. Carbon K-edge XANES also showed that the NOM is a humic substance [(b) in Fig. 2]. The much higher abundance of Ca^{2+} ion in the Pripyat River than in the Kuchibuto River can facilitate the coagulation of phyllosilicates and humic substances. The difference can be interpreted by the difference in geology among the two regions: limestone, which is one of the main geologies of this area, can supply a large amount of Ca^{2+} into the river water, while weathered granite is the main lithology in the Fukushima area.

This study based on various X-ray spectroscopic methods showed that the water solubility of RCs in Fukushima is much lower than that in Chernobyl due to the inhibition effect of adsorption of RCs into phyllosilicates, which originates from the difference in the soil and rock types in both areas. The lower water solubility by the larger affinity of Cs for PM in the Fukushima area controls the migration of RCs in the area and further circulation into the ecosystem.

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BEAMLINES

BL-9A and BL-13A

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