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Investigation of the River Particulate Matters in Fukushima and Chernobyl Related to Cesium Behavior

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The behavior of the radioactive cesium (radio-Cs), which was emitted by nuclear power plant accident, was different between in the Abukuma River (Fukushima) and in the Pripyat River (Chernobyl). Previous studies showed that dissolved Cs was dominant in the Pripyat River (approximately 70%). Conversely, Cs in the particulate matters (PMs) was dominant in the Abukuma River (dissolved Cs is approximately 30%). this difference of radio-Cs behavior was related to the blocking effect by the coating of natural organic matter (NOM) on clay minerals in the aquatic environments. However, the mechanism of the blocking effect by NOM has not been investigated for environmental samples yet in terms of morphological image and characterization.

Scanning transmission X-ray microscopy (STXM), X-ray microscopy using soft X-ray region, is very confortable to confirm the blocking effect by NOM. However, STXM had not been installed into synchrotron radiation facilities in Japan until 2012. Therefore, compact STXM (cSTXM) was newly designed and developed at BL-13A in KEK-Photon Factory (PF) until 2014. Its size, similar to laptop (A4 paper size), is much smaller than conventional STXM (Bruker's STXM). Piezo electronic driven stages were installed to all axis stages in the cSTXM. This system can be fast and precise sufficient to control the nano-scale imaging. As to spatial resolution using 30 nm outer most zone width FZP, we can attain resolution about 37 nm at sample measurement position. All detection was conducted by counting system using avalanche photo diode (APD) or photo multiplier (PMT). Soft X-ray region allow us to measure the light elements such as carbon, nitrogen, and oxygen.

As to samples, suspended water was collected from in the Abukuma River and the Pripyat River, which was filtered initially by 3.0 μm membrane filter and subsequently by 0.45 μm membrane filter using a pressurized pumping system. Finally, fine PMs ($>$0.45 μm) were obtained by this filtration. Before STXM measurement, these PMs were dispersed into water by supersonic wave for 5 min. The water droplet including the PMs was dropped on a 50-nm-thick Si3N4 membrane and air-dried. In addition to the sampling, the concentration of dissolved organic carbon (DOC) was measured in these rivers. In this study, we analyzed these PMs obtained from the two rivers using scanning transmission X-ray microscope (STXM) to investigate (i) the chemical distribution images of NOM (mainly composed by carbon) and clay minerals (mainly composed by aluminum) and (ii) the functional group images of carbon. In addition, (iii) the characteristic of NOM was measured by STXM-near edge X-ray absorption fine structure (NEXAFS) extracted from target area of NOM adsorbed on clay minerals.

Two distribution images (carbon and aluminum) showed that clay minerals were covered with NOM in the Fukushima and Chernobyl samples. By functional group mapping of carbon, it was found that distribution of each functional group was similar among different functional groups (e.g. aromatic, phenolic, carboxylic) in the PMs obtained from the two rivers. NEXAFS spectra of carbon were also similar to all samples, suggesting that humic substances (mainly composed of humic and fulvic acid) are the main cause of the blocking effect in these rivers. Based on these results, it is suggested that the blocking effect could occur in the Abukuma and Pripyat Rivers, which is caused by humic substances covering with clay minerals. Therefore, it is concluded that the degree of the blocking effect by NOM related to radio-Cs behavior was dependent on the concentration of DOC in the river (in the River Abukuma: 0.9-1.3 mg/L, in the River Pripyat: 18 mg/L). In addition to the studies on the rivers, investigation results for PMs obtained from Fukushima pond (DOC value is between the Abukuma and Pripyat Rivers) will be explained in this presentation.

Keywords: Natural organic matters, river particulate matters, Cs behavior, Scanning transmission X-ray microscopy (STXM), Fukushima, Chernobyl