Effect of Cesium-bearing Microparticles to the Solid-Liquid Distribution of Cesium in Rivers

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Introduction: The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident occurred on 11th March 2011 and a lot of radionuclides were emitted into the environment from the reactors (Yoshida and Takahashi, 2012). About 1.3×10^{16} Bq of Cesium-137 (Cs-137) was deposited to the ground by dry and wet depositions (Chino et al., 2011). According to Konoplev et al. (2016), solid-liquid distribution coefficient K_d values in Fukushima rivers are by 1-2 orders of magnitude higher than those in Chernobyl and it is possibly because the presence of Cs-bearing microparticles and higher Radiocecium Interception Potential (RIP) values of soils in Fukushima than those in Chernobyl. Cesium-bearing microparticle is a glassy, water-insoluble particle (Adachi et al., 2013). Cesium and many other elements from the reactor were found in the microparticles (Abe et al., 2014). The calculation of isotope ratio of Cs shows that the particles are from unit 2 or 3 (Nishihara et al., 2012). In this paper, the microparticles in suspended particles on the filters in river waters will be shown, focusing on the effect of the microparticles to the K_d values in the rivers. If there are the microparticles on the filter, apparent K_d value will be higher than intrinsic K_d value related to the adsorption-desorption reaction to the clay minerals.

Method: Suspended particles (3-63 μm) were collected from some rivers in Fukushima Prefecture by filtering water (60-90 L) in 2014 - 2016. After the filters were dried, total radioactivity of Cs-137 on the filters was measured by high purity Germanium semiconductor detector (HPGe). Planar distribution of radiation of filter was measured by autoradiography with Imaging Plates (IPs). After microparticle with high radioactivity was found on the filter, it was separated from other particles by water and NaI scintillation counter. Our method can separate microparticles more quickly and efficiently than previous methods (Adachi et al., 2013). Also, the microparticles can be more readily found by Scanning Electron Microscope (SEM) without using glue in the separation processes. The microparticles were measured by SEM coupled with Energy Dispersive x-ray Spectroscopy (EDS) to determine that the particles were similar to the particles reported in Adachi et al. (2013) etc. In the end, radioactivity of Cs-137 in the microparticle was measured by HPGe.

Result: Some hot spots were found on filters of Kuchibuto River collected in 2014 and 2015 by autoradiography. No hot spots were found on the filters from other rivers. The results of SEM-EDS showed that these hot particles and reported Cs-bearing microparticles were almost similar because they consist of elements such as Si, Cs, Fe, and Zn. Radioactivities of Cs-137 in the microparticles were about 0.8-3.0 Bq. Their isotope ratios of Cs (Cs-134/Cs-137 around 1.1) showed that the microparticles were from unit 2 or 3 of FDNPP. On filters collected on 3rd May 2014, 30% of Cs-137 on the filters was from the microparticles, while 46% of Cs-137 on filters was from the microparticles for the samples of 11th November 2015. If K_d value is calculated using these filters, the value will be overestimated compared with intrinsic K_d value that assumes adsorption-desorption reactions on clay minerals etc. The filters after 1-3 years from the accident and from different places should be measured successively. These results may explain the phenomenon that K_d values in rivers in Fukushima are higher than those in Chernobyl. The effect of the Cs-bearing microparticles must be considered when K_d values of Cs in Fukushima are discussed.

Keywords: Fukushima, Cesium, Cesium-bearing particle, nuclear power plant accident, radioactivity, Solid-liquid distribution