Discovery of radiocesium-bearing microparticles from ocean samples emitted from the Fukushima Daiichi Nuclear Power Plant accident

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Introduction: A large amount of radioactive Cs was emitted into the environment by the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident. Adachi et al. (2013) first reported radiocesium-bearing microparticles (CsMPs) from aerosol filters. Subsequent researches showed that the CsMP is SiO$_2$ glass with Cs, Cl, K, Fe, and Zn mainly contained in the particle. Diameter of CsMP is ~1-10 μm and $^{137}$Cs radioactivity is ~0.5 to 10$^2$ Bq. It has been suggested that the CsMP was mainly emitted from Unit 2 or Unit 3 of FDNPP based on the $^{134}$Cs/$^{137}$Cs activity ratio in the samples. Miura et al. (2018) reported CsMPs from the suspended particles in river water and their effect on $K_d$ value, which suggested CsMPs may exist in the ocean transported through rivers. Kubo et al. (2018) and Ikenoue et al. (2018) reported hot spots in the ocean samples by autoradiography but they did not separate CsMPs from these spots. In this presentation, we first report CsMPs separated from marine suspended particles, sinking particles, and sediments in coastal area of Fukushima and compare them with CsMPs from the terrestrial samples.

Method: We collected suspended particles (2011, 2013, 2015), sinking particles (2014), sediment cores (2011) from coastal area of Fukushima. By a wet separation method (Miura et al., 2018), we separated CsMPs from these samples. After measurement of radioactivity with a high-purity germanium semiconductor detector, scanning electron microscope (SEM) and energy dispersive X-ray spectroscopy (EDS) analyses were performed for separated CsMPs. Using autoradiography, we calculated $^{137}$Cs activity of unseparated hot spots over 0.1 Bq, which may be CsMPs.

Results and discussion: We separated 5 CsMPs from marine samples. The results of SEM-EDS analyses showed that these CsMPs have almost similar characteristics to the reported CsMPs because they mainly consist of Si, Cs, Fe, and Zn. Their $^{134}$Cs/$^{137}$Cs showed that the CsMPs were from Unit 2 or 3 of FDNPP. $^{137}$Cs radioactivity per volume is also similar to reported CsMPs from Unit 2 or 3. In this presentation, we will show the effect of CsMPs on $K_d$ values. CsMPs in the ocean samples will make apparent $K_d$ values be higher than intrinsic $K_d$ values related to the adsorption-
desorption reaction to the clay minerals, which may explain the large variation of Cs concentration in marine samples.