



Fukushima: Highly Radioactive Cesium-Bearing Microparticles Released into Japan's Natural Environment. Impacts on Human Health

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Abstract

Microparticles containing substantial amounts of radiocesium collected from the ground in Fukushima were investigated mainly by transmission electron microscopy (TEM) and X-ray microanalysis with scanning TEM (STEM). Particles of around 2 μm in diameter are basically silicate glass containing Fe and Zn as transition metals, Cs, Rb and K as alkali ions, and Sn as substantial elements. These elements are homogeneously distributed in the glass except Cs which has a concentration gradient, increasing from center to surface. Nano-sized crystallites such as copper- zinc- and molybdenum sulfide, and silver telluride were found inside the microparticles, which probably resulted from the segregation of the silicate and sulfide (telluride) during molten-stage. An alkali-depleted layer of ca. 0.2 μm thick exists at the outer side of the particle collected from cedar leaves 8 months after the nuclear accident, suggesting gradual leaching of radiocesium from the microparticles in the natural environment.

Introduction

Although almost five years have passed since the accident of Fukushima Daiichi Nuclear Power Plant (FDNPP), radioactive contamination in the surrounding area is still a serious problem in Japan. The largest radionuclide deposition event occurred on March 15–16 and the second largest on 21–23, 2011. Wet deposition was a major source of radiocesium contamination of terrestrial environment¹, while contribution of dry deposition was larger near the FDNPP². In order to understand and predict the fate of radioactive materials contaminating the terrestrial environment, it is important to clarify the physicochemical properties of the deposited materials. From previous cases of radionuclide release, it is known that the chemical species of released radiocesium is monovalent cation (Cs⁺) which is soluble³. Deposition of radiocesium as insoluble particles has also been pointed out. Autoradiography analyses using imaging plate (IP) showed spots of particulate materials on plant tissues collected from Fukushima^{4,5,6}. On the aerosol filter collected from March 14–15, 2011 in Tsukuba, 170 km south-southwest of FDNPP, Adachi *et al.*⁷ discovered spherical

particulate radiocesium of 2.0–2.6 µm in diameter, with particles insoluble in water having a glass-like structure⁸. These microparticles contain several fission products of U-235 other than radiocesium, and Fe and Zn which are also used in nuclear reactors⁸. Hence, they were considered to be released directly from nuclear reactors.

Kaneyasu *et al.*⁹ suggested that vaporized radiocesium was transported with sulfate aerosol in the air, dissolved to cloud droplets and fell as rain. On the aerosol filter collected on March 20–21, 2011, rainy days in Tsukuba, the majority of radiocesium was in water-soluble form⁷. Such water-soluble radiocesium that reached the ground surface as a solute was fixed to soils, especially to clay minerals¹⁰. In the terrestrial environment, the majority of radiocesium is present in solid form regardless of the initial form of deposition. However, compared to clay minerals originally contaminated by soluble radiocesium in soil, the solid radiocesium, which was initially deposited as radioactive microparticles, had stronger radioactivity. Although the contribution or percentage of such radioactive microparticles in the contamination level of Fukushima has not been evaluated, its influence on human health may be serious in terms of its intense radioactivity. Moreover, the structural detail of the microparticles may give insights into the state of the broken reactor and fuel debris.

In the present study, we investigated radioactive microparticles, similar to those reported by Adachi *et al.*⁷, but collected from the ground, by observing their internal structure with transmission electron microscopic (TEM) techniques.

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