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Short communication

First successful isolation of radioactive particles from soil near the Fukushima Daiichi Nuclear Power Plant

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ABSTRACT

The Fukushima Daiichi Nuclear Power Plant (F1NPP) accident in 2011 released radionuclides into the atmosphere in both aerosol and gaseous form. Subsequent studies of contamination in the environment have focused on the bulk radioactivity in samples. Comparatively little is known about the relative contribution and patterns of soluble versus particulate deposition of the radionuclides.

We investigated a sample of heavily contaminated surface soil from a site 20 km northwest of the F1NPP and isolated four radioactive particles from the surrounding soil. These particles had a maximum particle area equivalent diameter of 6.4 μm and a maximum ^{137}Cs radioactivity of 67.5 ± 0.1 Bq per particle. They were larger than the particles identified in aerosol samples shortly after the accident at a location 170 km southwest of the F1NPP. Two of the particles were spherical and two were fragmental.

Silicates were a major component of the Fukushima radioactive particles. These characteristics clearly differ from the so-called hot particles observed at the Chernobyl accident in 1986. Clarifying the physical and chemical properties of the radionuclides released from the F1NPP accident is important for assessing the potential long-term impacts to humans.

1. Introduction

The Fukushima Daiichi Nuclear Power Plant (F1NPP) accident, caused by tsunami damage following the Great East Japan Earthquake on 11 March 2011, released enormous amounts of radioactive material into the atmosphere. Radiocaesium (Radioactive Cs) was one of the most abundant radionuclides released into the environment during the F1NPP accident, with an estimated radioactive Cs release on the order of 9–36 PBq (Terada et al., 2012; Stohl et al., 2012; Katata et al., 2015; Winiarek et al., 2012). Understanding the physical and chemical properties of the emitted radioactive caesium is important to accurately evaluate the possible impacts to human health and to assess the long-term distributions of these particles after deposition in residential areas, agricultural fields, mountains, and aquatic environments. In the early stages of the F1NPP accident, radioactive Cs-bearling particles 2–10 μm in diameter was dominant (Doi et al., 2013; Miyamoto et al., 2014). In addition, radioactive Cs were transported from catchment to downstream as suspended matters (Chartin et al., 2013; Lepage et al., 2016). And, it was stored in river basin as sediment (Kitamura et al., 2014). However, the chemical forms of radioactive Cs were unclear at that time.

To investigate the chemical forms of radioactive Cs, Adachi et al. (2013) isolated radioactive water-insoluble microspheres from atmospheric samples collected on 14 and 15 March in Tsukuba, which is approximately 170 km southwest of the F1NPP. The radioactive particles were isolated from the aerosol samples using a scanning electron microscope equipped with an energy dispersive X-ray spectrometer (SEM–EDS). The particles contained Cs, O, Fe, and Zn and were several micrometres in diameter; the ^{137}Cs activity of the particles was approximately 2–3 Bq per particle. These particles were collected from the plume that was released on 14 and 15 March, but they were not observed in aerosol samples from other plumes, such as one that occurred 20–21 March 2011. Abe et al. (2014) used state-of-the-art synchrotron X-ray spectroscopy to examine the chemistry of the Cs-bearing particles isolated by Adachi et al. (2013). They concluded that the particles: (1) contained elements derived from nuclear fission processes and from nuclear reactor and fuel materials; (2) were amorphous; (3) were highly oxidized; and (4) consisted of glassy

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spherules formed from a molten mixture of nuclear fuel and reactor material. Yamaguchi et al. (2016) recently found similar Cs-bearing particles from deposits on non-woven fabric cloth and on a needle of Japanese cedar in Fukushima and showed their internal compositions using a focused ion beam and a transmission electron microscope. In contrast, Kaneyasu et al. (2012) showed that radioactive Cs was hosted in sulfate aerosols, as determined by a particle size analysis of the radioactive material and measurements of non-radioactive aerosol chemical components in April–May 2011. Kaneyasu et al. (2012) argued that the Cs-bearing sulfate aerosols were submicron in size and soluble in water; such particles would behave differently than the Cs-bearing particles reported by Adachi et al. (2013). Therefore, it is thought that the physical and chemical forms of the radioactive Cs emitted during the F1NPP accident, as well as their release times, varied considerably. In particular, soluble radioactive Cs has been assumed to distribute rather uniformly in the dose calculation in human body. However, the presence of high concentrated insoluble radioactive Cs-bearing particle in the early F1NPP plume could result in underestimation of internal exposure in human body who had been exposed to such plume. For instance, effective dose coefficients for inhalation become the larger if radioactive Cs is the less soluble (e.g. ICRP Publication 119).

The radioactive solid particles observed at nuclear power plant accidents, nuclear tests, operations of nuclear fuel reprocessing factory, and other nuclear disasters (Salbu, 2011). For example, radioactive particles so-called hot-particles were observed around the Chernobyl Nuclear Power Plant site (Sandalls et al., 1993). Most of the radioactive particles were fine particles of uranium oxide. The Chernobyl accident happened phreatic explosion, and nuclear reaction resulting fire (Tcherkezian et al., 1994). Windscale accident in 1957 also happened core fire, which resulting in emission of radioactive particles from the stack to the atmosphere (Salbu et al., 1994). Emissions of radioactive particles were anticipated in the F1NPP accident (Salbu, 2011). However, a core fire and phreatic explosion did not occur in the F1NPP accident. The explosions of unit 1, 3, and 4 were hydrogen explosions (TEPCO, 2012). The unit 4 hydrogen explosion occurred in the reverse flow of hydrogen generated in unit 3. In addition, in the Cs-bearing particles isolated by Adachi et al. (2013) and Abe et al. (2014) only isotopes of radioactive Cs were detected by activity measurements. Other radionuclides (e.g. Co, Ru, Ag, Sn, Sb, and Am) were not detected. Thus, it is suspected that radioactive particles were released directly from the reactor, and further investigation of the Cs-bearing particles is necessary to reveal their chemical and physical features. It is also necessary to identify the differences between the Fukushima radioactive particles and those in other incidents to gain basic scientific information useful for efforts such as environmental and health impact assessment, decontamination, and decommission.

The Cs-bearing particles have not been found from aerosol samples from known plumes in Fukushima Prefecture, which is closer to the release site and the location of the most severe contamination. In contrast, studies applying autoradiography have shown high-intensity spots of autoradiographic images in samples of soil (Itoh et al., 2014; Niimura et al., 2015; Satou et al., 2015; Yanaga and Oishi, 2015), forest leaves, vegetables, and dust from inside a building 40 km south of the F1NPP (Kashimura et al., 2013; Shibata et al., 2013; Tanaka et al., 2013). These results imply the presence of radioactive particles. None of the studies, however, have confirmed the chemical and physical properties of Cs-bearing radioactive particles by isolating them.

Heavy contamination by radionuclides occurred to the area northwest of the F1NPP on the same day (Katata et al.,2015). Within 30 km-range from the F1NPP site is about the same range of much of the radioactive particles (so-called hot particles) were found in the case of the Chernobyl accident (Sandalls et al., 1993). The particles had different characteristics, depending on the sampling locations and the distance from the accident site (Salbu and Kerkling, 1998). Therefore, the radioactive particles obtained closer to the nuclear power plant would be expected to give even more information about the accident. Thus, we aim to investigate the radioactive particles' distribution in the northwest area of the F1NPP which has not been made hitherto. We addressed the following questions. First, did radioactive particles come from the F1NPP? Second, did spot type contaminations come from Cs-bearing particle?

2. Materials and methods

2.1. Sample collection

A soil sample was collected at a location approximately 20 km northwest of the F1NPP on 12 June 2013, twenty-seven months after the accident. We collected five surface samples of soil from a location at 37°33′30″N, 140°49′38″E (Fig. 1), inside the exclusion zone located northwest of the F1NPP. This zone is distinguished by high air dose rates, based on airborne monitoring by the Ministry of Education, Culture, Sports, Science and Technology (MEXT). The location was the highest air dose rate spot within our entry permission. This land, which was used as a paddy field before the accident, has been abandoned and vegetation has therefore regrown. We sampled the surface soil (0–5 cm) with a 5-cm-diameter core sampler (DIK-115B, Daiki-Rika, Saitama, Japan). The soil samples were bulked and homogenised in a plastic bag (Onda et al., 2015) and then transported to the laboratory and stored at ambient temperature.

2.2. Particle isolation and analysis

The radioactivity of the bulk soil sample was measured with a high-purity germanium (HPGe) semiconductor detector (GEM-type, Seiko EG&G, Tokyo, Japan). After the radionuclide determination, the soil sample was packed into a polyethylene bag for imaging plate (IP) autoradiography using a BAS IP MS 2025 (high-sensitivity model, GE Healthcare Japan, Tokyo, Japan) and a digital imager (BAS-1800II, Fujifilm, Tokyo, Japan). Because high concentrations of radioactive Cs were detected in the soil sample, a relatively short IP exposure time (5 min) was applied to the sample to detect only relatively strong radioactive emissions from particles; such emissions produce distinctive spots on the autoradiograph. Soil near high-strength spots detected by IP was separated from the surrounding soil for further analysis. The IP and separation procedures were repeated on increasingly smaller samples until relatively few soil particles remained. The selected soil particles were collected on sticky carbon tape and the existence of gamma emitter activity in the γ-spectrum, was demonstrated by using an HPGe detector.

We then cut the carbon tape into successively smaller pieces to isolate any radioactive particles from non-radioactive soil particles using a technique developed in previous studies (Adachi et al., 2013; Abe et al., 2014). Backscattered electron images are very useful in searching for radioactive particles containing elements with high atomic number, such as Fe, Zn, Pb, and Cs, from carbon tape (Salbu et al., 1994). Finally, we obtained small pieces of carbon tape containing radioactive particles and analysed these particles using SEM–EDS (SEM: SU 3500, Hitachi High-Technologies Co., Tokyo, Japan; EDS: X-max 50 mm, Horiba Ltd., Kyoto, Japan) to observe the particle morphology, measure the particle size, and determine the composition. In some cases, particles on carbon tape were transferred to Gecko Tape (Nitto Denko Co. Ltd., Japan), which is of high purity elemental carbon, using a microscope (BXiS series,