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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,

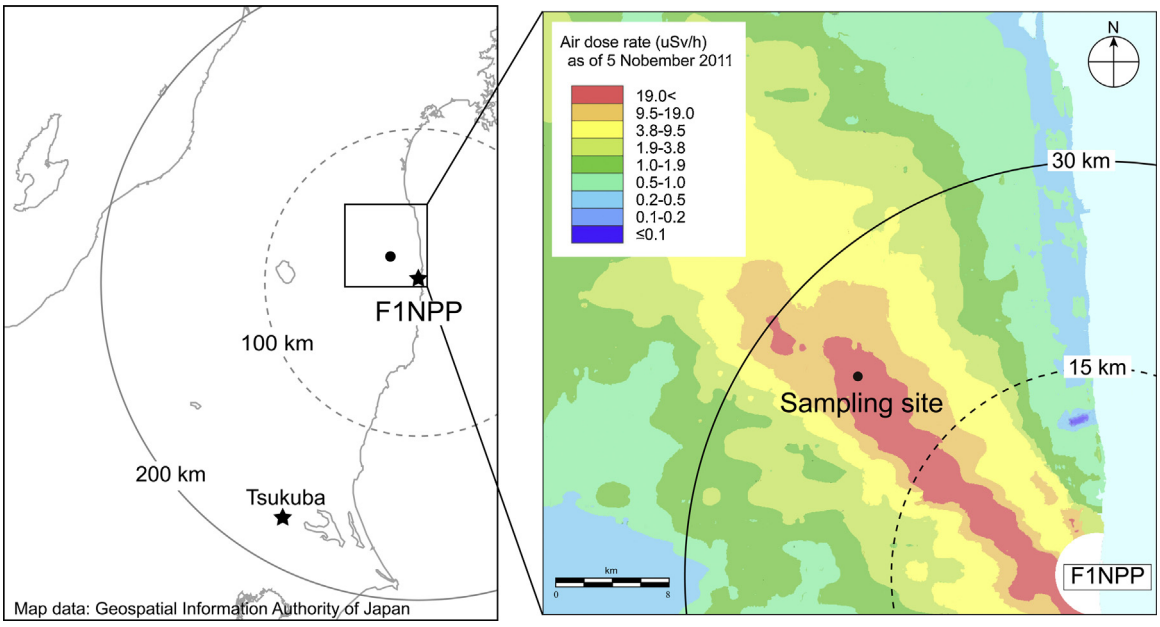

Fig. 1. Left: Map showing the location of the Fukushima Daiichi Nuclear Power Plant (F1NPP) on the east coast of Japan and the sampling site (solid dot), located ~20 km northwest of the FNDPP. Right: Map of the area northwest of the F1NPP (the location of the mapped area is indicated by the black square in the left panel), showing the air dose rate on 5 November 2011 (as determined by MEXT) and the location of the soil sampling site (black dot); the sample was collected from the most highly contaminated zone, i.e., the so-called difficult-to-return zone.

Olympus, Tokyo, Japan) and a micromanipulator (Quick Pro, Micro Support Co., Ltd., Shizuoka, Japan) equipped with tungsten needles.

3. Results

3.1. Particle figures and radioactivity

The sampling location (shown in Fig. 1) received radionuclide deposits on 15 March 2011, mainly via wet processes (Kinoshita et al., 2011; Tsuruta et al., 2014). The air dose rate at the time of sample collection was 22.3 μSv/h, measured 1 m above the ground surface. The bulk density of soil was 1.24 g/cm^3, which is within the range of the values reported for soils in Fukushima (Kato et al., 2012; Honda et al., 2015; Matsunaka et al., 2016). Radionuclides in the soil sample were measured by gamma spectrometry using a high-purity germanium semiconductor detector. On the basis of these measurements and a correction for radioactive decay, we calculated that the amount of ^{134}Cs deposited from the atmospheric plume(s) was 27.07 ± 0.06 MBq/m^2 (86,900 ± 200 Bq/kg) and that of ^{137}Cs was 27.02 ± 0.01 MBq/m^2 (86,720 ± 40 Bq/kg). The activity ratio of ^{134}Cs/^{137}Cs was 1.00 (decay corrected as of 11 March 2011) which was consistent with the results of a large-scale ground survey performed by the Japanese government (Saito et al., 2015).

IP autoradiography images of the soil sample showed that contamination by radionuclides was relatively uniform, although many spot-type contaminations were apparent (Fig. 2a). We isolated four radioactive particles from such spot-type contamination positions in the sample (Fig. 2b). Compared to the analysis of radioactive Cs adsorbed on clay minerals, the present radioactive particles were detected within ~10 min by using IP, so that the specific activity seems stronger than those found by Mukai et al. The sizes and morphologies of the particles varied: the maximum area equivalent diameter was 6.4 μm (particle B), and the particles were either spherule or fragmental in shape. The radionuclides ^{134}Cs and ^{137}Cs were detected by gamma spectroscopy in all of the particles, with particle B showing the highest concentrations (Fig. 2c); namely 69.5 ± 0.5 Bq for ^{134}Cs and 67.1 ± 0.1 Bq for ^{137}Cs (values corrected for decay). Although the particles' total radioactive Cs activity was less than 1/1000 of the total unit soil activity (Bq/kg), there were many more radioactive particles in the sample (see Fig. 2a). The activity ratio of ^{134}Cs/^{137}Cs was 1.03 ± 0.01, which is consistent with values for the bulk soil sample. However, no other gamma emitters were detected in the particles (Fig. 2c). The relationship between particle volume and ^{137}Cs concentration is proportional (Fig. 3), suggesting that these particles have the same source.

3.2. Particle compositions

The elemental constituents of the particles were measured with an EDS (Fig. 4a). All particles showed X-ray spectra of Si, O, Fe, and Zn, as well as Cs. Although X-ray peaks for Al were observed in all particles, the peaks could not be separated from those of the background sample, and we therefore could not confirm its presence in the individual particles. An element map of particle B is shown in Fig. 4b. Uniform distributions of Cs, O, Fe, Zn, and Si were observed within the particles (Fig. 4b). Approximately 80% was composed of Si and O, within each particle followed by K, Fe, and Zn.

4. Discussion

The particles, collected 27 months after the accident, were larger and more radioactive than those analysed in previous studies (Adachi et al., 2013; Abe et al., 2014; Yamaguchi et al., 2016). In particular, particle B had approximately 20 times the volume of the particles observed in aerosol samples collected in Tsukuba (Adachi et al., 2013), which is located approximately 170 km southwest of the F1NPP. Because of the high dry deposition velocity of the present particles (~10 cm/s for a 6 μm particle;

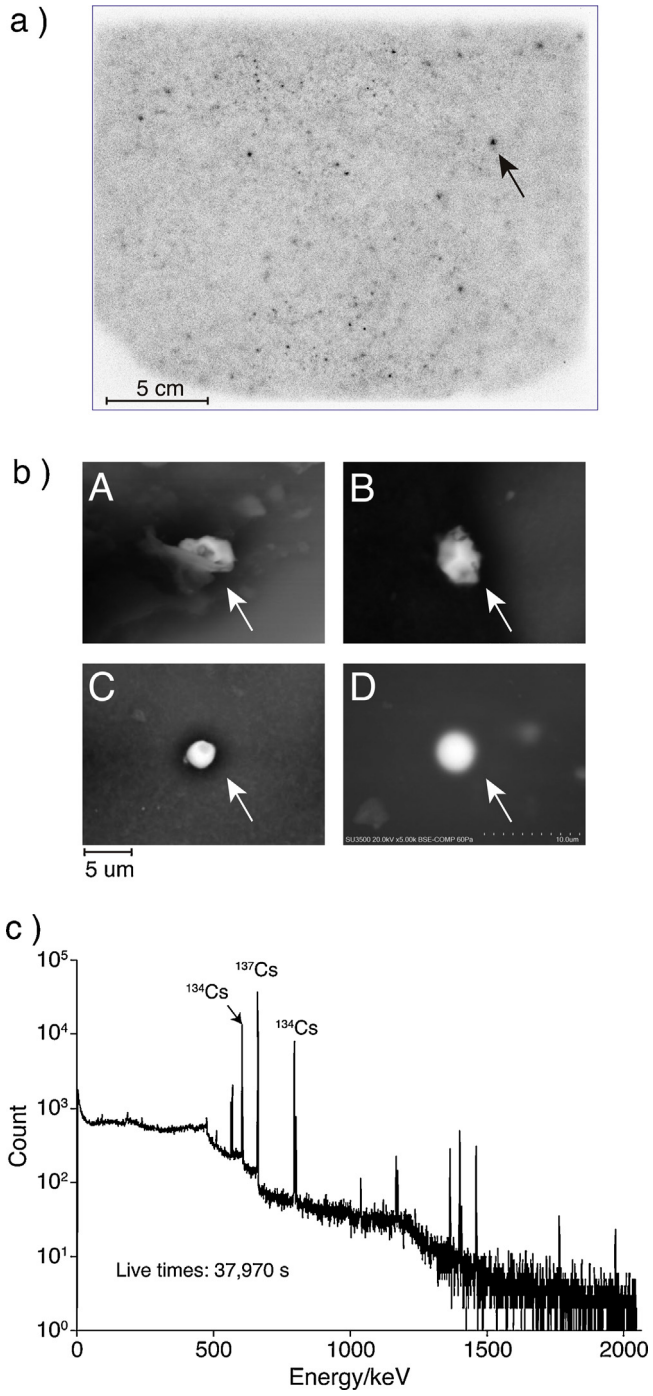

Fig. 2. Distribution of radioactive spots of different types in the soil sample and SEM images of the isolated radioactive particles. (a) Autoradiography image obtained by the imaging plate (IP) technique. The square frame represents the imaging plate and the grey colour distributed over the entire soil sample indicates that the soil was uniformly contaminated with radioactive materials. Black spots indicate aggregates of radioactive atoms, which occur throughout the soil sample. The black arrow indicates particle B. (b) SEM images of four radioactive particles (arrows) isolated from the soil sample. The particles are either spherules or fragmental in shape. Area equivalent diameters of particle A, B, C, and D are 4.5, 6.4, 4.0 and 3.0 μm, respectively. (c) Gamma ray spectrum of particle B.

Seinfeld and Pandis, 2006), it is possible that particles of the size collected in this study deposited rapidly and were therefore confined to areas near the source.

The specific radioactivity of ^{137}Cs within the present radioactive particles is consistent with those in the previous studies from the F1NPP by Adachi et al. (2013) and Abe et al. (2014) ($=2\times10^{16}\times[\text{volume}]^{1.43}$; Fig. 3). By contrast, the specific radioactivity of hot particles observed in the Chernobyl accident ($=4\times10^{7}\times[\text{volume}]^{0.74}$; Tcherkezian et al., 1994) is smaller than present particles from the F1NNP, indicating that the radioactive particles from the F1NNP accident have higher radioactive Cs concentration within the particle.

In addition, the diameters of the present radioactive particles range from 2 to 6.7 μm, which is consistent with the particle size distribution of aerosol containing the F1NPP ^{137}Cs (Doi et al., 2013; Miyamoto et al., 2014) and are within the range of PM_{10} and $PM_{2.5}$ (Davidson et al., 2005), which are inhalable to human body.

Particles C and D were spherules, suggesting that they formed from liquefied materials (Adachi et al., 2013). During the F1NPP accident the containment vessel pressure was observed to suddenly drop in each of the reactor units, suggesting that rapid cooling could have occurred following an adiabatic explosion during the exhaust process. Such rapid cooling offers a simple mechanism for the liquid-to-solid micro-particle conversion. In contrast, particles A and B were fragmental, suggesting that they were derived from larger fragments that had fractured in the soil during transportation or release.

The particles we collected contained Si, O, Fe, Zn, and Cs, which is consistent with the composition of previously reported particles (Adachi et al., 2013; Abe et al., 2014), although those studies could not confirm the presence of Si in the particles because Si was present in the substrate materials used in the previous analyses. We avoided this limitation by using a Gecko Tape substrate (Maeno and Nakayama, 2009). The result is consistent with the result by Yamaguchi et al. (2016) who showed the presence of Si using a transmission electron microscopy and EDS. Possible sources for the Si include silicate products that were used for the insulation in/around the reactor unit and around steam lines in the containment building of the F1NPP (Nuclear Safety Commission, Japan, 2006) as well as in the concrete of the containment vessel (Abe et al., 2014). We developed this novel point by vigorously surveying the material used for the reactor core and peripherals as well as rich in silicate.

The water solubility of these particles has important implications for human health, environmental impact, transportation, and deposition (Adachi et al., 2013). In the Chernobyl accident, highly radioactive particles were observed (Sandalls et al., 1993; Salbu et al., 1994), and they gradually collapsed and became soluble in water as time elapsed (Kashparov et al., 2004). The dissolution processes from radioactive particles contributed soluble radionuclides to the terrestrial environment. The dissolution rate depends on the soil pH (Kashparov et al., 2004). The radioactive particles in the soil sample remained stable or quasi-stable in the environment for 27 months, in a location that had experienced more than 3000 mm of precipitation during that time (Japan Meteorological Agency, 2015). Thus, the particles appear to exhibit low solubility in water and are stable in the ambient conditions of the environment where they were deposited. We could argue that, although the particles may have lost some components or been totally or partially oxidised, and although the molecular composition may have changed, these particles have remained in the ambient environment while maintaining their shape. Thus, long-term water solubility is recommended to be investigated to determine the corrosion rate of these Cs-bearing particles in the soil environment.

Decontamination of radioactive Cs from living areas are emerging issue in the area and large amount of contaminated soil and materials are being generated continuously. Volume reduction of the contaminants is pressing issue. In contrast, understanding of chemical form of radioactive Cs is important to remove radioactive Cs from contaminant. We have demonstrated

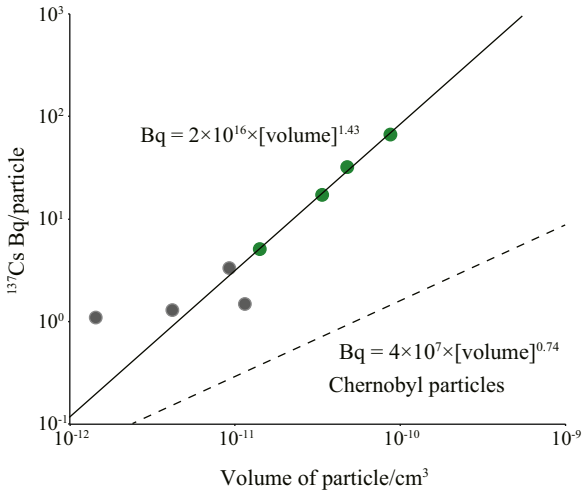

Fig. 3. Relationship between the volume of particles and the radioactivity of ^{137}Cs. The dashed line shows specific radioactivity of hot particles in the Chernobyl nuclear accident in 1986. Data were reconstructed from Tcherkezian et al. (1994).

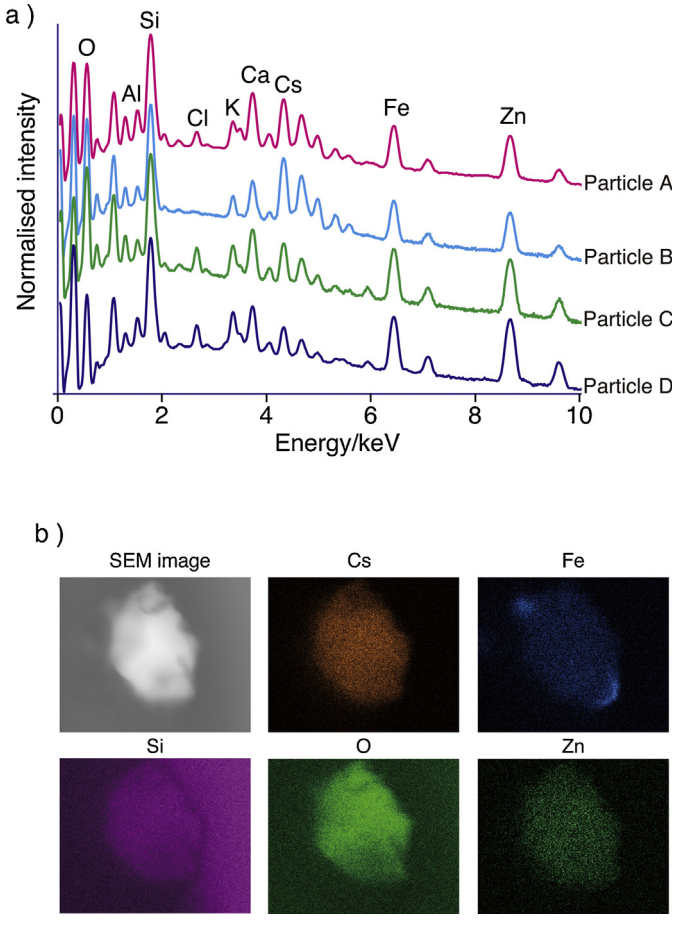

Fig. 4. EDS spectra and elemental mapping images. (a) Characteristic X-ray spectra from four radioactive particles. All particles exhibit signals characteristic of Si, O, Fe, and Zn, as well as Cs; Ca and K were also detected. (b) Elemental mapping of Cs, Si, O, Fe, and Zn in the particles. Silicone was present in the glass substrate. The Fe signal may represent material located beneath the particle.

that one of the existing forms of radioactive Cs has silicate glass state on this study and such knowledge will be helpful to the decontamination processes.

5. Conclusions

The radioactive particles we collected from the polluted soil were larger and more active than Cs-bearing particles analysed by Adachi et al. (2013), Abe et al. (2014) and Yamaguchi et al. (2016), but their constituents were similar to, those of the reported aerosol particles. In addition, the present study demonstrated that silicates are one of the major components in the radioactive particle, and that the particle characteristics were different from those of so-called hot particles observed at the Chernobyl accident in 1986. We suggest that the present radioactive particles in the soil were discharged to the atmosphere during the same F1NPP emission event on 14 and 15 March 2011 with the source of previously reported Cs-bearing solid particles. Our results provide information on the temperature of materials ejected during the accident, some of which appear to have been molten.

Our findings are useful to understand environmental dynamics studies of radioactive materials from the Fukushima accident. In addition, our findings have implications for the removal of the radioactive materials from the living areas and understanding of process of the accident. We suggest that additional investigations be conducted to analyse more particles from various distances from the F1NPP to elucidate the particle size–distance relationship, to determine the processes of synthesis of the particles in the reactor cores and peripherals, and to evaluate the effect of weathering on the particles in the environment.

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