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Discovery of non-spherical heterogeneous radiocesium-bearing particles not derived from Unit 1 of the Fukushima Dai-ichi Nuclear Power Plant, in residences five years after the accident

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ABSTRACT

Non-spherical heterogeneous radiocesium-bearing particles were found on masks worn during cleaning work in residences near the Fukushima Dai-ichi Nuclear Power Plant (FDNPP), from which residents had evacuated. Three slightly larger (6.6–12 μm) non-spherical radiocesium-bearing particles were found in a residence in Futaba Town, a straight distance of 2.11 km west-northwest from the FDNPP. These were collected on October 25, 2016, 5 years and 7 months after the Fukushima Dai-ichi nuclear disaster and were presumed to originate from the Plant's Unit 2 based on the measured radioactivity ratio of $^{134}Cs/^{137}Cs$. The main elemental composition was similar to particles already reported in other studies. However, this is the first time that such particles had a clearly heterogeneous distribution.

1. Introduction

A huge amount of radioactive materials was introduced into the atmosphere by the Fukushima Dai-ichi nuclear disaster initiated by the tsunami following the 2011 Tohoku earthquake on March 11, 2011. Among these were the radiocesium isotopes ^{134}Cs and ^{137}Cs, which have still been deposited; the ambient dose rate has remained higher than before the disaster (Ministry of Education, Culture, Sports, Science and Technology, Japan, 2011). Soon after the disaster, the Japanese government designated a series of evacuation and protection zones around the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) (Fukushima Prefectural Government, 2015). In 2012, these areas were reorganized into three categories depending on the level of contamination. As shown in Fig. 1, a large part of Namie Town, most of Futaba Town, nearly half of Okuma Town, and a part of Tomioka Town were classified as “Difficult-to-return zone”, where the annual cumulative dose exceeds 50 mSv and the annual cumulative dose may not fall below 20 mSv after 5 years.

Little is known about the chemical and physical properties of the radioactive materials released into the environment by the Fukushima Dai-ichi nuclear disaster. Some studies have addressed radiocesium-bearing particles. Adachi et al. (2013) found spherical, water-insoluble micro-particles containing Fe, Zn, and Cs; these mainly reached the ground by dry deposition. Abe et al. (2014) used synchrotron radiation X-ray micro-beam analyses to reveal that the radiocesium-bearing particles may also contain Rb, Zr, Mo, Sn, Sb, Te, Ba, and U. Although the radiocesium-bearing particles can be spherical with a diameter of about 2 μm, larger non-spherical particles with a diameter of 6.4 μm have been discovered in soil samples collected from a site 20 km northwest of the FDNPP (Satou et al., 2016). Furuki et al. (2017) reported the discovery of Cs-rich radiocesium-bearing particles in soil and gravel collected 2–4 km from the FDNPP.

In another study, indoor contamination of dispersed radiocesium was investigated within ninety-five residential houses in Iitate Village, Odaka District, and the towns of Futaba, Okuma, and Tomioka from July 2013 to January 2015 (Yoshida-Ohuchi et al.,

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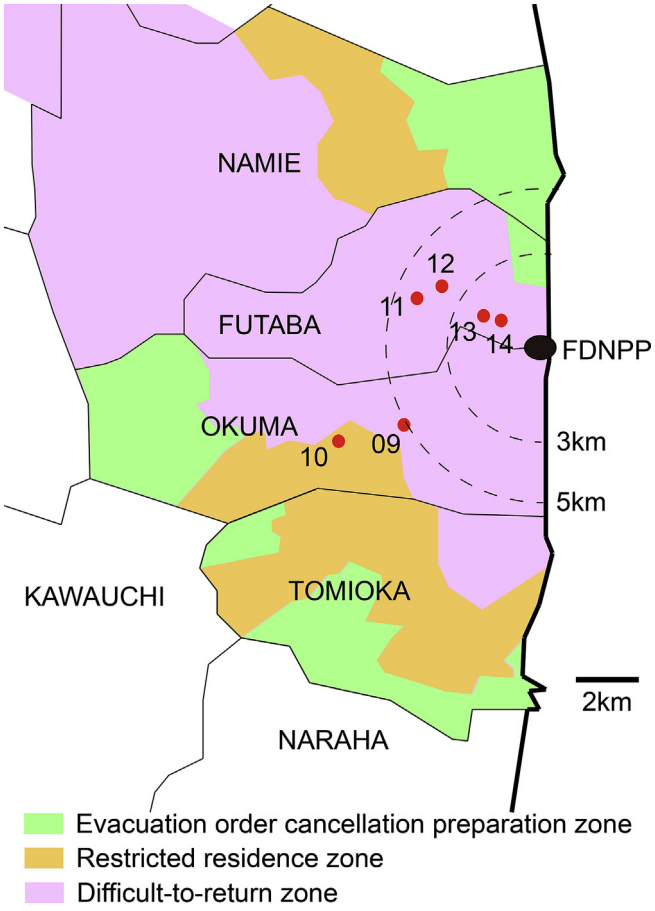

Fig. 1. Map of residence locations and evacuation areas (July 12, 2016). Dotted concentric circles indicate 3 km and 5 km distance from FDNPP.

2016). This study revealed that in houses in Okuma, Futaba, and Tomioka, closer to the FDNPP than those in Odaka District and Iitate Village, surface contamination was inversely proportional to the square of the distance between a house and the FDNPP.

The ventilation rate of a typical Japanese wooden house is high. Other damage, such as cracks from the 2011 Tohoku earthquake, could also increase ventilation. Radiocesium-bearing particles, which invaded residences with the inflow of outside air after the residents evacuated, will cause internal exposure to evacuees who return to their homes. The effective dose by inhalation varies depending on the size and chemical form of the particle even if the radioactivity level is the same. As Abe et al. (2014) reported, some of these radiocesium-bearing particles contain U, so these are also assumed to contain not only nuclear fuel materials such as ^{235}U or ^{238}U but also descendant nuclei. Therefore, it is unknown how much exposure the evacuees will face upon resuming their lives.

The radiocesium-bearing particles derived from the Fukushima Dai-ichi nuclear disaster have previously only been isolated from high-volume air sampler filters or soil samples; no report has yet documented their discovery from indoor samples. In Japan, wearing a mask is commonly used as a countermeasure to inhalation of house dust. In a previous study, we showed that wearing a nonwoven fabric mask could reduce inhalation exposure to dispersed radiocesium from the Fukushima Dai-ichi nuclear disaster (Higaki and Hirota, 2013). Such masks have also been used to reveal as the main source of radiocesium adhered to masks (Higaki et al., 2014). In the current study, we measured the radioactivity adhered to nonwoven fabric masks used while conducting indoor cleaning of residences in order to investigate the presence of radiocesium-bearing particles and to estimate the potential inhalation dose of returning evacuees in order to develop countermeasures against future inhalation.

2. Material and methods

2.1. Sampling

Vigorous housecleaning activities were conducted by two to three people within six residential houses in Futaba and Okuma, Fukushima Prefecture from September 29 to December 26, 2016. A total of six houses in Okuma Town (ID_09 and ID_10) and Futaba Town (ID_11 to ID_14), where residents had evacuated, were cleaned up. Fig. 1 shows a map of the residences. The housecleaning activities were done like as those in; using a vacuum cleaner, clearing dust from a wall, ceiling, lintel, and furniture, and sweeping all rooms with every door and window kept closed. A nonwoven fabric mask was worn throughout these activities; 14 total masks were used. All masks were packed into individual polyethylene bags after use to prevent cross-contamination.

2.2. Mask analysis

Each mask was packed into a plastic cylindrical vessel (47 mm diameter, 6 cm height) to a 1.7 cm height and analyzed by a high-resolution gamma spectrometry system that included a high-purity germanium detector (CANBERRA Industries Inc., GX4018). The detector was shielded with 10 cm lead blocks, along with 2 cm copper and 0.5 cm acrylic plates to decrease the background contribution. The counting time of every sample was 50,000 to 100,000 s. Radioactivity standard solutions of ^{134}Cs (Japan Radioisotope Association, CZ-010) and ^{137}Cs (Japan Radioisotope Association, CS-005) were added to unused mask for quantitative measurement. The radioactivity of the standard solution of both ^{134}Cs and ^{137}Cs were 236 Bq as of March 11, 2011. Before analysis by the gamma spectrometry system, the masks were exposed for 7 days with an imaging plate (BAS MS 2040, Fuji-film, Japan), confirming that radiocesium was heterogeneously distributed on all masks.

2.3. Separation and measurement of radiocesium-bearing particles

High radioactivity particles assumed to be radiocesium-bearing particles were separated from the masks using a technique such as relatively short-time exposure with an imaging plate. Details of the separation method are described elsewhere (Kurihara et al., 2017). After separation, the high radioactivity particles were analyzed using SEM–EDS (SEM: Hitachi S-4500; EDS: Kevex Sigm) to observe particle morphology, measure particle size, and determine particle composition. Each particle was quantified with a high-purity germanium detector with a counting time of 30,000 to 350,000 s. Radioactivity standard solutions of ^{134}Cs (0.182 Bq as of November 25, 2016, Japan Radioisotope Association, CZ-010) and ^{137}Cs (1.40 Bq as of November 25, 2016, Japan Radioisotope Association, CS-005) were dropped on a 1-mm square filter paper. These radioactivity standard solutions have been calibrated by Japan calibration service system (JCSS). The detection limits of ^{134}Cs and ^{137}Cs were 0.0120 Bq and 0.0135 Bq each with a counting time of 350,000 s. It was possible to identify that a particle was released from each nuclear given unit by using the ^{134}Cs/^{137}Cs radioactivity ratio decay-corrected at the time of the Fukushima Dai-ichi nuclear disaster (March 11, 2011) compared with the ORIGEN code by Nishihara et al. (2012).