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Radiocesium contamination in house dust within evacuation areas close to the Fukushima Daiichi nuclear power plant

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ABSTRACT

Outdoor decontamination efforts have been ongoing since the Fukushima Daiichi nuclear power plant (FDNPP) accident; however, little is known about indoor contamination. Therefore, house dust was sampled based on particle size in 21 wooden buildings (19 residential houses and 2 community centers) within the evacuation area close to the FDNPP, Fukushima Prefecture, Japan. Activities of radiocesium (^{137}Cs) per gram of house dust increased with decreasing particle size (mean: 6.1×10^3, 2.6×10^3, 1.6×10^3, 7.5×10^2, 5.0×10^2, and 4.6×10^2 Bq/g for < 4–20, 20–63, 63–180, 180–500, 500–1000, and 1000–2000 μm, respectively). The ^{137}Cs concentrations in house dust were inversely related to the square of distance from the FDNPP for < 4–1000 μm particles. From the results of the multiple linear regression analysis, distance from the FDNPP and direction from the FDNPP (northwest) were significantly related to the radioactivity of house dust. It was found that 19%, 33%, and 48% of ^{137}Cs in house dust were extracted in water, 1 M HCl, and not extracted, respectively. Considering the bioaccessibility and assuming a 20 mg/day daily intake of house dust, the daily doses would be 7.2 Bq/day (mean) and 18 Bq/day (95th percent quantile). These results provide valuable insight into indoor radioactive Cs contamination in the area around the FDNPP and possible oral exposure to indoor radioactive Cs after returning home.

1. Introduction

On 11 March 2011, the Great East Japan Earthquake and subsequent tsunami devastated the northeastern coast of Japan and damaged the Fukushima Daiichi nuclear power plant (FDNPP; TEPCO, 2012). Between 12 and 20 March, radioactive materials were released from the FDNPP because of hydrogen explosions, damage to the plant infrastructure, and containment vessel venting (TEPCO, 2011, 2012). The released radioactive materials were transported by wind and contaminated the ground, plants, and buildings via dry and wet deposition as the radioactive plume passed over the area.

Following the Chernobyl accident, several studies considered the radioactivities of house dust. In Helsinki, Finland, the radioactivities of house dust on the floor were at the same levels as outdoor soil because residents transport outdoor soil into the indoors (mechanical transport), while the radioactivities of house dust on and behind furniture were higher than those of floor dust and soil because outdoor aerosols penetrated to the indoors and were deposited onto the floor (Kulmala et al., 1988). In Northwest England, the ^{137}Cs radioactivities were found to be outdoor > entrance mat > hall carpet > lounge carpet because outdoor soil was mechanically transported into the indoor environment via the bottoms of shoes (Allott et al., 1992, 1994).

There are currently no publications highlighting the radioactivities of house dust after the Fukushima accident. Although, mechanical transport and aerosol infiltration and deposition were the sources of indoor radiocesium in European studies, mechanical transport could be negligible in Fukushima because these houses were evacuated after the accident and because Japanese customs sees people remove their shoes when entering a house. Furniture and indoor surfaces of buildings within residential houses in the evacuation areas were reportedly contaminated with radioactive cesium (Cs) (Yoshida-Ohuchi et al., 2016) derived directly from the FDNPP or the resuspension of radioactive material adsorbed on soil.

Decontamination was performed in residential areas of the evacuation zone, except for difficult-to-return zones, until the end of March 2017 and the evacuation instructions issued throughout these areas were cancelled in March and April 2017 (Nuclear Emergency Response Headquarters, 2016), allowing residents to return to their homes. After

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the Chernobyl accident, indoor surface cleaning was recommended to reduce dose in Europe because indoor contamination may be significant (Andersson et al., 2008; Andersson, 2009). However, at Fukushima, decontamination was only conducted outdoors; indoor environments remain largely as they were at the time of the accident, with little known about indoor Cs contamination.

The ICRP recommends that the additional effective dose for public should not exceed 1 mSv in a year; this is based on the annual effective dose from natural radiation sources, which is approximately 1 mSv, and reflects the low variation in mortality rate between 1 and 5 mSv/year continuous exposure (ICRP, 1990a, 2007). The Japanese limit of radiocesium (sum of ^{134}Cs and ^{137}Cs) for intake of general food (including radiostrontium and radioplutonium) is 100 Bq/kg, equivalent to 0.9 mSv/year assuming 374 kg/year of food intake (% of annual food intake) and 1.81×10^{-5} mSv/Bq of the effective dose coefficient (MHLW, 2013). The maximum permitted levels of ^{134}Cs and ^{137}Cs in EU for import food from third countries was 600 Bq/kg except for milk and infant food, which is 370 Bq/kg, equivalent to 1.0 mSv/year assuming 10% of daily diet is contaminated (EU, 2011). After Fukushima accident, the maximum permitted levels of ^{134}Cs and ^{137}Cs in EU for imports import food from Japan was 500 Bq/kg except for milk, dairy products, infant/young-children food, and liquid foodstuffs, which is 200 Bq/kg because > 10% of daily diet could be contaminated (EU, 2011). The committed effective does from natural radioactive elements for Japanese due to ingestion is reported to be 0.80 mSv/y mainly contributed by ^{210}Po in seafood (Ota et al., 2009).

In the present paper, we described radiocesium contamination of house dust in 21 wooden buildings (19 residential houses and 2 community centers) in the evacuation area close to the FDNPP, and examined radiocesium activities in different sizes of house dust and extractability of radiocesium in house dust in water and in 1 M HCl solution. Finally, the possibility of Cs oral exposure via house dust after returning home.

2. Materials and methods

2.1. Survey area and period

Between April 2016 and January 2017, house dust was sampled by particle size during indoor cleaning of 21 wooden buildings (19 residential houses and 2 community centers) in the towns of Futaba and Okuma (Fukushima Prefecture), which are close to the FDNPP (1.60–6.92 km from the FDNPP; Fig. 1). Of these, 18 houses and 2 community centers were in difficult-to-return zones, and one house was in the habitation restriction zone. These buildings have all been uninhabited since the accident of March 2011. Subjects whose houses were not destroyed were recruited via the headmen of several wards.

2.2. House dust sampling

House dust was once sampled between 9:00 and 17:00 by particle size using a cyclone vacuum (DC61MH; Dyson) with multistage sieves (20, 63, 180, 500, 1000, and 2000 μm) and a 70-mm polytetrafluoroethylene filter (pore size 4 μm, Advantec PF040, Toyo Roshi Kaisha, Ltd., Japan), through a stainless-steel pipe with an electropolished interior (Fig. 2) after dusting the surface of furniture, lintels, and walls in the house. The house dust samples (20–63, 63–180, 180–500, 500–1000, and 1000–2000 μm) were stored in 2.5-mL polypropylene vials (outer diameter: 11 mm, height: 47 mm, wall thickness: 0.9 mm; Maruemu Co., Japan) after further sieving by shaking with a mixer (VTX-3000L; LMS Co. Ltd., Japan) and weighing with a balance (AUW120D; Shimadzu Co., Japan). Since a 4-μm filter can capture particles smaller than 4 μm with certain efficiency, hereinafter the smallest size fraction expressed is as < 4–20 μm.

In several houses, the house dust was collected separately on the first and second floors. For these houses, the Cs radioactivity of each house was calculated as the dust-weight-weighted average of each floor.

2.3. Separation depending on extractability

House dust samples (50 or 100 mg) were placed in polycarbonate ultrafiltration tubes (outer diameter: 29 mm, height: 110 mm, wall thickness: 1.0 mm; VIVASPIN 20-5K; General Electric Company, USA) with 2.5 or 5.0 mL of pure water and shaken at 200 rpm for 2 h using a reciprocal shaker (SR-1; TAITEC CORPORATION, Japan). Then, samples were centrifugally filtered at 12000 ×*g* for 60 min, the residue washed twice with 1 mL of pure water, and centrifugally filtered twice at 12000 ×g for 30 min. The triple-filtered solutions were combined, weighed with a balance, and stored in a polypropylene tube (outer diameter: 18 mm, height: 100 mm, wall thickness: 1.2 mm; Fisher Healthcare Ltd., UK). Then, 2.5 or 5.0 mL of 1 M HCl were added to the residue and the tubes were shaken at 200 rpm for 2 h based on the Japanese official method for soil content measurement (MOE, 2008a) for the evaluation of oral exposure. Next, the tubes were centrifugally filtered at 18,000 ×*g* for 90 min. The tubes were centrifugally filtered twice more at 18,000 ×*g* for 60 min after being washed twice with 1 mL of 1 M HCl. Finally, the triple-filtered solutions were combined,

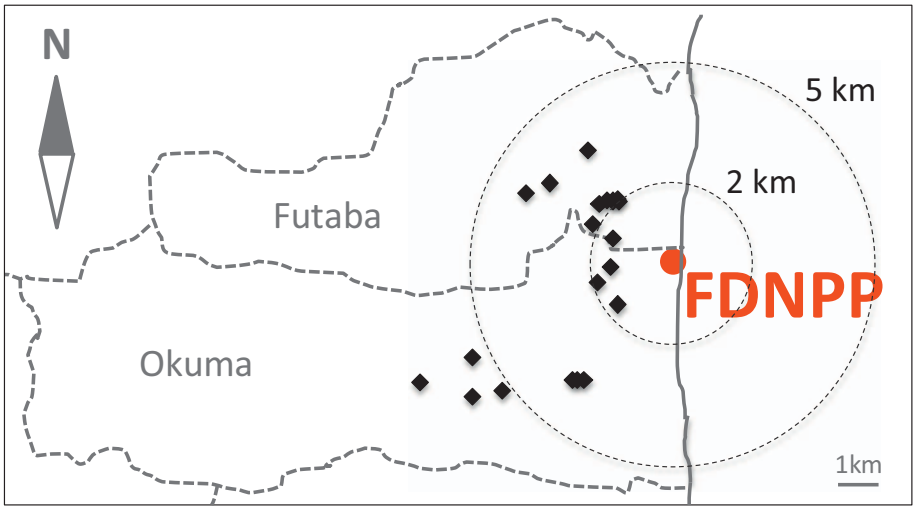

Fig. 1. Map showing location of the study area. Black diamonds denote the locations of buildings surveyed in the present study. The red circle denotes the location of the Fukushima Daiichi nuclear power plant (FDNPP). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)