Title

Indoor and outdoor radionuclide distribution in houses after the Fukushima Dai-ichi Nuclear Power Plant accident

Highlight

- The radionuclide distribution inside and outside houses 8.5 years after deposition was investigated.
- The contamination levels varied depending on material and location.
- Our study shows these uncertainty factors that should be considered in dose assessment.

Abstract

An enormous amount of radionuclides was released into the atmosphere following the Fukushima Dai-ichi Nuclear Power Plant accident, with part of it penetrating houses. Information on radionuclide distribution inside and outside houses is essential for assessing external dose rate of resident. To investigate the radionuclide distribution, we selected two houses in Fukushima Prefecture unoccupied since the accident, and collected both soil samples around the houses and their house material samples of floor, inner wall, ceiling, outer wall, and roof, in 2019. Radioactivity of ^{137}Cs in the samples was measured using a high-purity germanium detector. We found that the surface contamination densities of ^{137}Cs of house materials relative to the deposition density was roughly in the order of roof > floor ~ glass wool in roof space ~ outer wall > inner wall > ceiling. The relative surface contamination densities of ^{137}Cs varied depending on the surface orientation, the surface material, and location such as sampling height, indicating that those factors should be considered in the dose assessment for residents.

26 Keywords

27 Fukushima Dai-ichi Nuclear Power Plant, Radionuclide distribution, Radiocesium

1. Introduction

An enormous amount of radionuclides was released into the atmosphere following the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident. Part of it was transported to residential areas and deposited on various surfaces outside houses (outer walls and roofs) and their surrounding areas (ground) via dry and wet deposition. Additionally, radionuclides penetrated houses through openings (e.g., windows, doors, and ventilation systems) and deposited on surfaces of floors, inner walls, and ceilings (Hirouchi et al., 2017). Evacuated people planning to return their houses in the affected areas are concerned about indoor dose rate from the penetrated radionuclides. Indoor dose rate is assessed from the radioactivity concentration on the ground and surfaces in residential areas, using a model simulating dose rate in residential areas (Meckbach and Jacob, 1988; Furuta and Takahashi, 2014). The models require radioactivity concentration on the ground and each surface in residential areas or ratio of the concentration on each surface to that on the ground, as a parameter to represent the indoor and outdoor radionuclide distribution. Therefore, it is important to determine the contamination level on surfaces in residential areas and the ground for indoor dose assessments of evacuated people planning to return their houses and decontamination workers who will clean the houses. Additionally, to investigate the generality of the indoor and outdoor radionuclide distribution, it is necessary to investigate the factors that affect radionuclide distribution.

Various surveys were conducted after the Chernobyl and FDNPP accidents on the level of contamination by radionuclides of indoor and outdoor surfaces of residences (Nichoison and Hedgecock, 1991; Allott et al., 1992, 1994; Roed, 1997; Andersson et al., 2002; Yoshida-Ohuchi et al., 2016; Yoshimura et al., 2017; Shinohara and Yoshida-Ohuchi, 2018). Radiation dose rate can be systematically assessed considering the contribution of both indoor and outdoor surfaces by clarifying the relationship of the contamination levels among various indoor and outdoor surfaces including the ground. However, the previous studies targeted the

53 contamination either indoor or outdoor surfaces and did not provide sufficient data to assess
54 radiation doses from all contaminations. Furthermore, the difference in contamination level
55 among indoor surfaces was not surveyed because of the application limits of the wipe test used
56 for sample collection (Yoshida-Ohuchi et al., 2016). In the comparison of survey results after
57 the accidents in Fukushima (Yoshimura et al., 2017) and Europe (Andersson et al., 2002), the
58 contamination levels measured on roofs relative to the ground were different between them.
59 However, because information to explain the cause of the difference is insufficient, it is not
60 possible to determine whether the ratio of the contamination levels on roofs to the ground
61 should be changed depending on the country and roof material. To clarify this difference, more
62 information is needed on the contamination levels of indoor and outdoor surfaces in Fukushima.
63 This study aimed to investigate the indoor and outdoor radionuclide distribution for
64 dose assessments and clarify the reasons behind their variability. With this aim, we determined
65 the radioactivity of the soil and house material such as floor, inner wall, ceiling, outer wall, and
66 roof collected in Fukushima in 8.5 years after deposition by directly analyzing them with a
67 high-purity germanium detector.
68
69 2. Materials and Methods
70 Samples of surface materials were collected in two wooden houses (referred to as
71 House A and House B) with two stories in Okuma Town on October 30, 2019, that is 8.5 years
72 after the contamination incident. The FDNPP is located at the seashore of this town at a distance
73 of ~3 km, which was hence for the most part highly contaminated by radionuclide after the
74 accident. The geometric mean ground deposition density of ^{137}Cs in this town was found to be
75 1.23 MBq m^{-2} on March 15, 2011 (MEXT, 2011). Notably, the houses selected in this study
76 were located in a highly contaminated area in Okuma Town and unoccupied after the accident;
77 the inside and outside areas of the target houses had been neither cleaned nor decontaminated,

and almost no people entered these areas after the FDNPP accident. All windows, doors, and walls of these houses were closed and unbroken throughout the 8.5 years in visual confirmation. These facts imply that the radionuclides deposited onto indoor surfaces during the passage of the radioactive plume is hardly disturbed by humans or animals after the evacuation of the residents. Moreover, indoor contamination hardly originated from resuspensions penetrating through relatively large openings, such as broken windows, from outdoor soil.

We collected samples of the floor, inner wall, ceiling, outer wall, and roof of both houses using a handsaw and knife. The tatami mat, wood, paper, glass wool, clay tile, and gypsum were collected as common materials. Additionally, the clay wall was collected in House A, and the carpet and slate roof were collected in House B. The ceiling of the second-floor was covered with paper on the room side and glass wool on the roof space side. For the ceiling material on the second-floor, we cut out only the paper with a knife and rip out the glass wool in the roof space. The glass wool was classified as a floor material in the roof space in this study. The indoor sampling point was preselected to limit access and reduce floor contamination by workers performing the measurements.

To determine the contamination distribution on the outer walls at a certain height, samples were obtained at the heights of 20–60 cm, 60–100 cm, 100–140 cm, and 140–180 cm above the ground. In the roof materials, samples were collected around the top and bottom roof edges. The first-floor and second-floor roof materials were collected in House A and House B, respectively. Notably, the first-floor roof materials were collected from an area that was quite far from the area of the second-floor roof cover to avoid the influence of redeposition of radionuclides from the second-floor roofs due to run-off. One sample was collected from each sampling point without no repetition.

Soil samples outside the target houses were collected at the depth of ground surface less than 1 cm, 0–5 cm, 5–10 cm, 10–15 cm, and 15–20 cm using a 100-mL soil core sampler

103 (DIK-1801, Daiki Rika Kogyo Co., Ltd.; length = 51 mm, φ = 50 cm). Although the second
104 layer did not have the surface layer (< 1cm), it was designated as 0–5 cm layer in this paper.
105 The ^{134}Cs and ^{137}Cs activities in all collected samples were measured to evaluate the
106 surface contamination density (Bq cm^{-2}). In this study, since radionuclides can penetrate the
107 samples, the surface contamination density included not only the radionuclides on the surface
108 but also the radionuclides that penetrated the sample. The samples made of paper or glass wool
109 were folded into small pieces to measure their activity using a 100-mL plastic container (U-8
110 container, Shinto Chemical Co., LTD., Bunkyo-ku, Tokyo). The bulk samples, consisting of
111 wood, tatami mat, clay, gypsum, and slate, were directly ground into powder before packing in
112 a U-8 container. The ^{134}Cs and ^{137}Cs activities in the house materials and soil was measured
113 using a high-purity germanium detector (GEM25P-70, ORTEC). The measured radioactivity
114 was decay-corrected to the sampling date (as of October 30, 2019).
115
116 3. Results and discussion
117 3.1. Relative surface contamination density
118 The results of surface contamination density of various house material and outside soil
119 samples are shown in Table S.1 in supplementary material. To assess the external dose rate of
120 people, it is necessary to determine the distribution of radionuclides on various surfaces. The
121 distribution has been mostly provided in terms of relative surface contamination density to the
122 ground deposition density in previous studies (Roed, 1997; Andersson et al., 2002; Yoshimura
123 et al., 2017). According to these studies, the surface contamination density relative to the ground
124 deposition density (R_m) can be defined as follows:

$$R_m = \frac{A_{s,l,m}}{A_g}, \quad (2)$$

125 where $A_{s,l,m}$ is surface contamination density on the sample surface s made of material m at the
126 location l, and A_g is the deposition density on the open undisturbed ground. The subscript s

indicates the floor, inner wall, ceiling, outer wall, and roof surfaces; moreover, the subscript *l* indicates different locations (first floor, second floor, roof space, height above the ground, top side, and bottom side); finally, the subscript *m* indicates different materials (tatami mat, carpet, wood, glass wool, clay, paper, gypsum, clay tile, and slate).

Since it is well known that radiocesium migrates into the soil over time after deposition (e.g. Matsuda et al., 2015; Yoshimura et al., 2020), and our data also show the migration (Table S1), the data for all collected soil core samples (0–20 cm including the ground surface) were used to estimate the deposition density on the ground. Obtained ground ^{137}Cs deposition density at House A and B was 2.26 and 0.87 MBq m^{-2}, respectively, and moderately agreed to 1.01 MBq m^{-2} (decay corrected to October 30, 2019) obtained by MEXT (2011).

The distribution of the ^{137}Cs relative surface contamination density inside and outside two examined houses is summarized in Fig. 1. The results provide information on both indoor and outdoor relative surface contamination densities on house materials, although variability of data was not investigated due to the sampling limitations. Since the relative surface contamination densities of ^{134}Cs were quite similar with those of ^{137}Cs, we show results of only ^{137}Cs from here.

3.2. Indoor distribution

As shown in Fig. 1, the indoor relative surface contamination densities were 3×10^{-3}–7×10^{-2} on the floor, 6×10^{-5}–4×10^{-4} on the wall, and 7×10^{-5}–3×10^{-4} on the ceiling. The indoor surface contamination density values reflected the following relationship: floor in a room of carpet > floor in a room of wood and tatami mat > floor in a roof space of glass wool > inner wall of clay >~ inner wall of paper > ceiling of paper.

The surface contamination densities on different indoor surfaces can be compared based on Fig. 2, which shows the surface contamination density on the indoor samples relative

to those on the wooden floor at the first floor of the houses. The deposition density ratios of the inner wall, ceiling, and roof space to the wooden floor were 0.002–0.06, 0.004–0.01, and 0.4–0.7, respectively. This relationship indicates that the surface contamination density differed depending on the surface orientation because dry deposition and resuspension for particles have gravitational settling component. Furthermore, the surface contamination density differed depending on the surface roughness because carpet and clay wall with its rougher surface would increase local turbulence. Yoshida-Ohuchi et al. (2016) reported that radionuclide contamination reflected the following relationship: floor in a room > floor in a roof space > vertical column. Moreover, the deposition density ratio of the roof space floor to the room floor was 0.5–1. The relationship to the orientation and deposition density ratio of the roof space to the room floor was consistent between our results and those reported by Yoshida-Ohuchi et al. (2016).

The contamination density is considered to be affected by the initial deposition as well as the redistribution due to resuspension after the deposition. Concerning the deposition velocity, it is known to be the particle size dependent factor. Shortly after the FDNPP accident (April and May 2011), the activity median aerodynamic diameter was reported to be ~0.5 μm in Tsukuba, at ~100 km from the FDNPP (Kaneyasu et al., 2012). Notably, particles in the size range of 0.1–1 μm are more likely to penetrate houses compared to other particles (Liu and Nazaroff, 2001, 2003; Thatcher et al., 2003). The deposition velocity of particles, which penetrated the houses in our study, should be comparable to that of 0.5-μm particles. In theoretical calculations, the ratios between the deposition velocities of 0.5-μm particles on the wall and ceiling in relation to that on the floor corresponded to ~0.2 and 0.02, respectively; such values generally decreased for larger particles (Lange, 1995; Lai and Nazaroff, 2000). In the experiment by Lange (1995), the deposition velocity of 0.5-μm indium particles on the wall and ceiling was ~0.17 and 0.02, respectively, of that on the floor (Fig. 2). Although the deposition velocity is

not directly comparable to the contamination density long time after the deposition, the deposition velocities were decreased in the order of floor, wall and ceiling, which is consistent to our contamination density data (Fig. 2).

The surface contamination density on the carpet was approximately three times higher than that on tatami mats and wooden flooring, while that on the clay walls was twice higher than on the wallpaper located in the first floor (Fig. 2), although there was only one sample for each material. Hence, the surface contamination density varied depending on the materials, even when both surface orientations and rooms were the same. This is because the rate of deposition on the surfaces and that of resuspension from them differed among the surface materials. In fact, the deposition velocity is known to depend on the electrostatic field and surface roughness, which vary among materials (Lai, 2002). Theoretical calculations indicated that particles with sizes <1 μm are affected by both of these parameters and are more likely to deposit, while those with sizes >1 μm are hardly affected (Otani et al., 1989; Opiolka et al., 1994; Lai and Nazaroff, 2000; Andersson et al., 2004). As described in the previous section, since most particles penetrating houses presumably have sizes <1 μm (Liu and Nazaroff, 2001, 2003; Thatcher et al., 2003), the amount of deposition into indoor surfaces could be affected by difference in the electrostatic field and surface roughness among materials.

The resuspension of deposited particles can also differ among materials, because the resistance to resuspension is lower on smoother surfaces (Andersson et al., 2004; Braun et al., 2002). The house material considered in this study was collected 8.5 years after the initial deposition of particles; hence, due to natural ventilation, some of these particles were likely to have experienced resuspension. Therefore, different indoor deposition velocities and resuspension rates may explain the differences in surface contamination density observed among materials. The experiments by Andersson et al. (2004) indicated that the particles with sizes >1 μm labeled with dysprosium and indium resuspended more easily than those with sizes

<1 μm. Particles resuspended from the ceiling and wall can deposit on the floor because a larger particle is more affected by gravitational settling. As a result, the ratio of the surface contamination densities on the wall and ceiling to the floor can become smaller over time due to resuspension. The redistribution due to resuspension probably attributed to the fact that the ratio of the surface contamination densities on the wall and ceiling to the floor in this study was smaller than that of deposition velocity obtained by Lange (1995), as shown in Fig. 2.

3.3. Outdoor distribution

The relative surface contamination densities of ^{137}Cs for the outdoor samples are plotted against the elapsed time after the accidents in Fig. 3, together with those by previous studies in the literature (Roed, 1997; Andersson et al., 2002; Yoshimura et al., 2017). Our results show that the relative surface contamination densities were 2×10^{-3}–1×10^{-2} on the outer wall of gypsum and 4×10^{-3}–2×10^{-1} on the roof of clay tile and slate. Outdoor surface contamination density reflected the following relationship: roof of slate > roof of clay tile > outer wall of gypsum. Moreover, the surface contamination density on the outer wall of gypsum and roof of slate tended to be higher for the samples collected at lower heights.

The relative surface contamination densities of ^{137}Cs for outdoor samples of this study were compared to those measured in Europe in the case of wet deposition, because ^{137}Cs from the FDNPP accident mainly deposited with rainfall (Morino et al., 2013). According to Roed (1997), immediately after the deposition of particles derived from the Chernobyl accident, the relative surface contamination densities of radiocesium on the roofs and outer walls were 0.3–0.9 and 0.001–0.3 in the case of wet deposition, respectively. Two years after the deposition, the relative surface contamination densities of radiocesium on roofs and outer walls were 0.1–0.7 and 0.01–0.03, respectively; furthermore, 14.5 years after the deposition, those on roofs was 0.05–0.4 (Andersson et al., 2002). Yoshimura et al. (2017) reported that the relative surface

contamination densities of radiocesium originated from the FDNPP accident on roofs and outer walls in Fukushima Prefecture were ~0.02 and 0.01, respectively, 4 years after the deposition.

As shown in Fig. 3, the relative surface contamination densities on roofs and outer walls had a large uncertainty of approximately an order of magnitude. This broad range of relative surface contamination densities might have been caused by the translocation of radionuclides and differences in protection against rain and retention capacity for radionuclides and rainwater among materials. Andersson et al. (2002) reported that the variation in surface contamination density on the outer walls was due to different conditions during deposition, such as differences in rain protection and soil splash between locations. Nichoison and Hedgecock (1991) pointed out that differences in surface contamination density on roofs can be attributed to the translocation of radionuclides after the deposition due to run-off from the higher to the lower parts of roof. Roed (1997) indicated that the amount of run-off from roofs can be influenced by the surface material and roof angle, which affect the amount of rainfall accumulated prior to the run-off, one of the measures of rainwater retention capacity. Yoshimura et al. (2017) mentioned that the surface contamination density may depend on the different radiocesium retention capacities between porous and non-porous materials. The results of these studies are consistent with our findings: the surface contamination densities on the lower outdoor samples or porous material of slate were higher than those on higher surfaces or on non-porous material of the clay tile.

To discuss the difference in surface contamination densities between materials, selective strong fixation sites specifically for cesium cation are also an important factor. There were strong cesium fixation sites in some construction materials (Andersson, 2009). Fired clay materials, such as clay tile, might or might not have such fixation sites because the clay material changes structure on firing at more than 1000°C, although some clays have some fixation sites. The difference in surface contamination densities between materials may be caused by the

252 difference in amount of strong cesium fixation sites, which were not investigated in this study.

253 Notably, the results we obtained from measurements conducted 8.5 years after the
254 initial particle deposition were largely consistent with those obtained 4 years after the initial
255 deposition in Japan (Yoshimura et al., 2017). Moreover, the relative surface contamination
256 densities on roofs in Japan tended to be lower than in Europe (Roed, 1997; Andersson et al.,
257 2002), although the relative surface contamination densities on the outer walls were very similar
258 in the two countries. One possible reason for the different relative surface contamination
259 densities measured on roofs between Japan and Europe may be the difference in the ratio of the
260 surface contamination density on the roof to that on the ground immediately after the first or
261 major deposition (referred to as the roof/ground initial deposition ratio). Roed (1997) focused
262 on the relationship between the deposition amount of radionuclides and amount of run-off
263 rainwater from roofs and suggested that the amount of run-off rainwater can be expressed as a
264 function of the total rainfall and rainwater retention capacity. According to Roed (1997), cement
265 tiles, red tiles, and eternit has high rainwater retention capacities, while silicone-treated eternit
266 have no retention capacity. In Europe, one week after the deposition, the roof samples of
267 corrugated eternit, red clay tiles, and concrete tiles had relative surface contamination densities
268 >0.5, while those of silicon-treated eternity had ~0.2 (Andersson et al., 2002). These results are
269 consistent with the trend of rainwater retention capacity obtained by Roed (1997). Hence, the
270 difference in the total rainfall in countries and rainwater retention capacity among materials
271 may cause a difference in the radioactivity of run-off water and result in the difference in the
272 roof/ground initial deposition ratio. However, this is insufficient to explain the difference in
273 relative surface contamination density observed between roofs in Japan and Europe: there are
274 insufficient data regarding the relative surface contamination density in Japan in the first 2 years
275 after the accident. Therefore, further investigation is needed on temporal changes in relative
276 surface contamination density of materials in Japan, especially during the first 2 years after an

277 accident.

278

279 4. Conclusions

280 The indoor and outdoor radioactivity distribution in houses unoccupied 8.5 years after
281 the accident was investigated in Fukushima Prefecture. The surface contamination densities on
282 the floor, inner wall, ceiling, outer wall, and roof relative to the ground were 3×10^{-3}–7×10^{-2},
283 6×10^{-5}–4×10^{-4}, 7×10^{-5}–3×10^{-4}, 2×10^{-3}–1×10^{-2}, and 4×10^{-3}–2×10^{-1}, respectively. The relative
284 surface contamination densities on the horizontal surfaces were higher than those on the vertical
285 surfaces. Moreover, they were particularly high on rough and porous surfaces presumably
286 because of differences in the deposition velocity, resuspension rate, and radionuclide retention
287 capacity. These results provide essential information on the distribution of indoor and outdoor
288 radioactivity and factors affecting the radionuclide distribution, including direction, location,
289 and surface material, which are needed to assess the dose rate of the resident. The relative
290 surface contamination densities on outdoor samples were similar to those previously measured
291 in the Japanese site, while those on roofs in Japan tended to be lower than those measured in
292 Europe, probably because of the different roof/ground initial deposition ratio. To understand
293 the differences between both areas, further investigation is needed on its temporal changes,
294 especially those that occurred during the early time period after an accident. This investigation
295 provides essential information for the dose rate assessment for people returning to the evacuated
296 houses and cleaning workers.

297

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384 Fig. 1. Distribution of the relative surface contamination density of ^{137}Cs on the house
385 materials. The different materials analyzed are indicated between brackets.

386

387 Fig.2. Surface contamination density of ^{137}Cs for indoor samples relative to that on the
388 wooden floor at first floor and deposition velocity of 0.5-μm indium particles on indoor surfaces
389 relative to that on the floor. The surface contamination density was obtained in this study, while
390 the deposition velocity was obtained by Lange (1995). The uncertainty based on measurement
391 error was less than 10%.

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393 Fig.3. Relative surface contamination density of ^{137}Cs on outdoor surfaces: (a) outer walls
394 and (b) roof. The dots and bars in the diagram indicate the mean and the range of values,
395 respectively. The data regarding European sites were previously reported by Roed (1997) and
396 Andersson et al. (2002). Some of the data regarding Japanese sites were previously reported by
397 Yoshimura et al. (2017).
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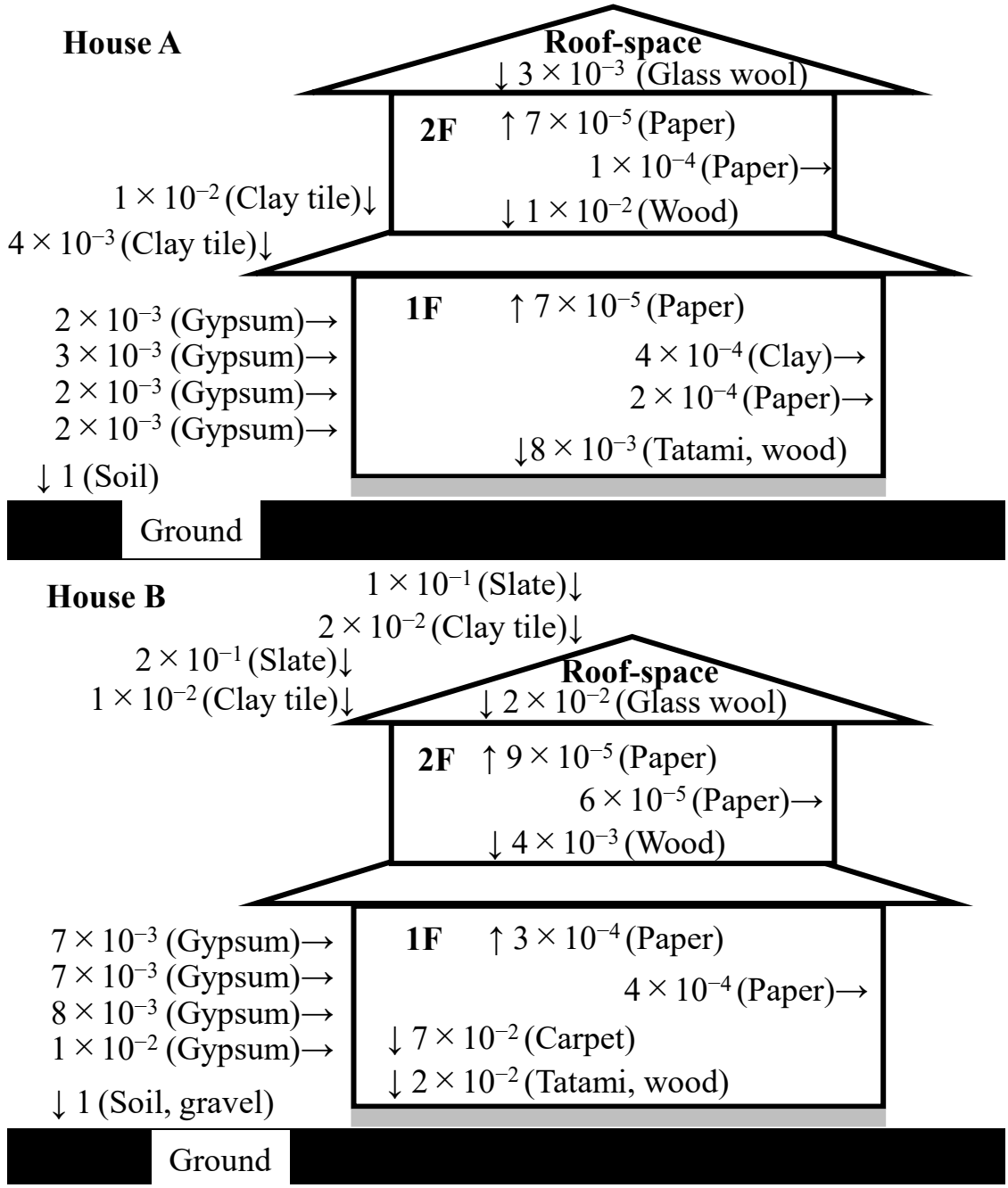

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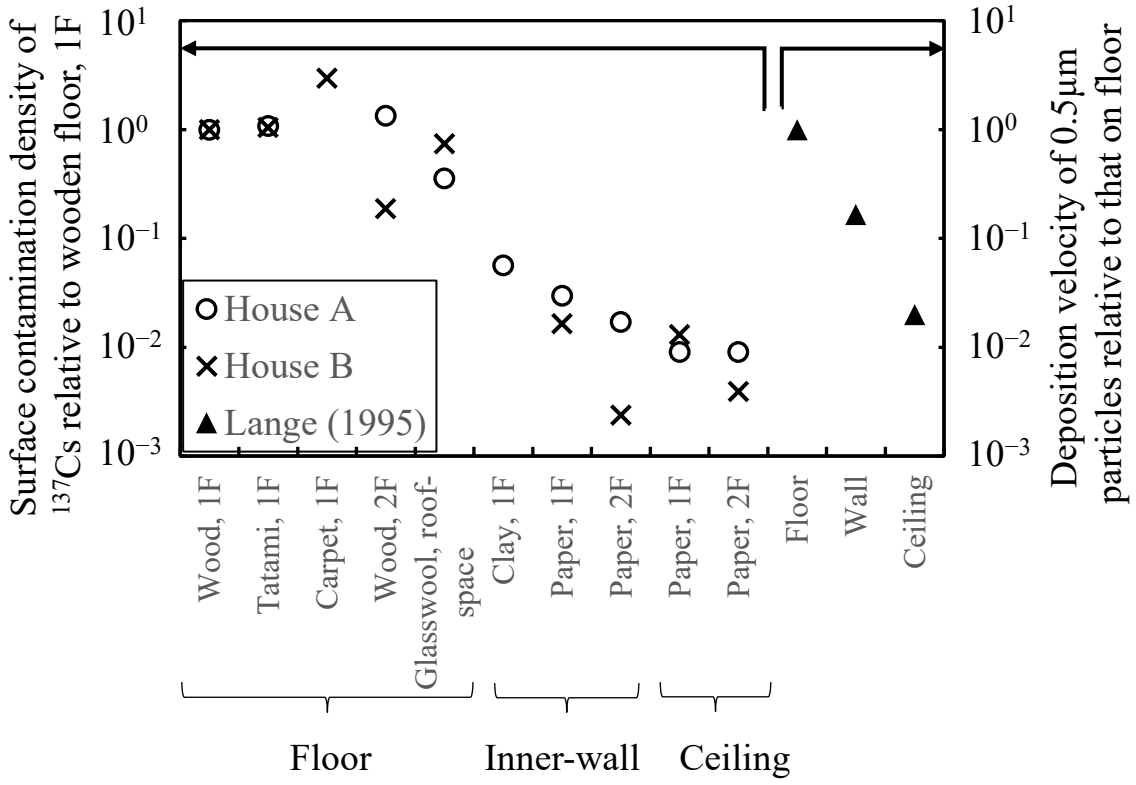

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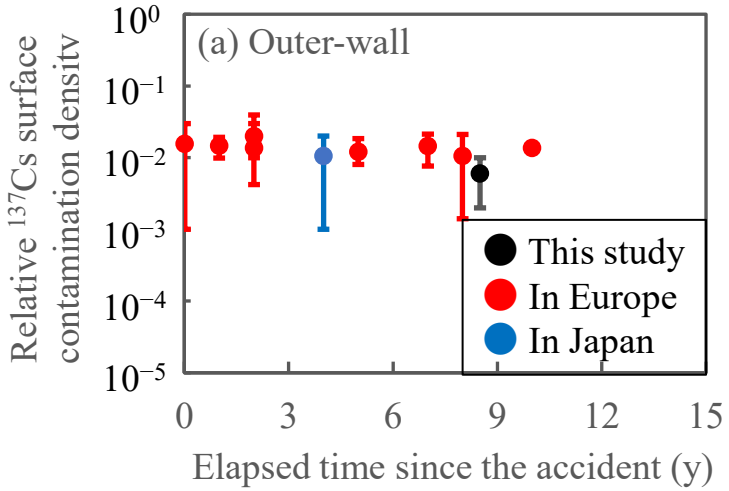

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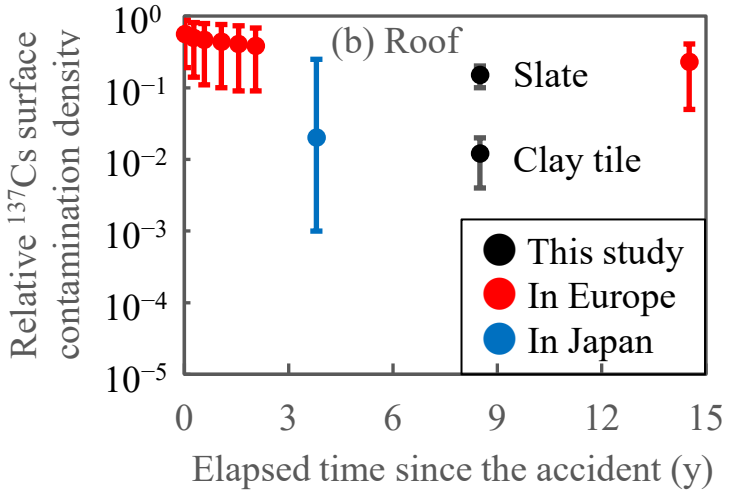

411

412 Fig.3. Relative surface contamination density of ^{137}Cs on outdoor surfaces: (a) outer walls
413 and (b) roof. The dots and bars in the diagram indicate the mean and the range of values,
414 respectively. The data regarding European sites were previously reported by Roed (1997) and
415 Andersson et al. (2002). Some of the data regarding Japanese sites were previously reported by
416 Yoshimura et al. (2017).