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Title

Identification of penetration path and deposition distribution of radionuclides in houses by experiments and numerical model

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Abstract

In order to lift of an evacuation order in evacuation areas and return residents to their homes, human dose assessments are required. However, it is difficult to exactly assess indoor external dose rate because the indoor distribution and infiltration pathways of radionuclides are unclear. This paper describes indoor and outdoor dose rates measured in eight houses in the difficult-to-return area in Fukushima Prefecture and identifies the distribution and main infiltration pathway of radionuclides in houses. In addition, it describes dose rates calculated with a Monte Carlo photon transport code to aid a thorough understanding of the measurements. The measurements and calculations indicate that radionuclides mainly infiltrate through visible openings such as vents, windows, and doors, and then deposit near these visible openings; however, they hardly infiltrate through sockets and air conditioning outlets. The measurements on rough surfaces such as bookshelves implies that radionuclides discharged from the Fukushima-Daiichi nuclear power plant did not deposit locally on rough surfaces.

Highlights

- Indoor and outdoor dose rates were measured in the difficult-to-return area.
- Dose rates were calculated with the Monte Carlo photon transport code EGS5.
- Main penetration pathways and indoor deposition of radionuclides are discussed.

Keywords

Dose rate measurement, Penetration, Deposition distribution, EGS5

1. Introduction

Enormous amounts of radionuclides were deposited over large areas of Japan by the Fukushima-Daiichi accident in 2011. In consequence, an evacuation order was issued for large areas of Fukushima Prefecture. The areas where the annual cumulative dose was expected to be 20–50 mSv or exceed 50 mSv were defined as the restricted residence area and the difficult-to-return area, respectively; these are areas from which people remain evacuated (IAEA, 2015). Due to recent decontamination activities, the area in which residents can return to their homes has gradually increased (Reconstruction Agency, 2016).

In order to lift of an evacuation order in both zones in preparation for the lifting of the evacuation order and restricted residence area and return residents to their homes, human dose assessments are required. In particular, indoor external dose assessment is important because residents may remain indoors for long periods. The International Atomic Energy Agency (IAEA)-TECDOC-225 (IAEA, 1979) and TECDOC-1662 (IAEA, 2000) suggested that the representative dose reduction factor of a wooden house is 0.4. However, according to measurements after the Fukushima-Daiichi accident, there were some houses where the dose reduction factor exceeded 0.4 (Yoshida et al., 2014). Yoshida et al. (2014) pointed out the possibility of radioactive contamination on indoor surfaces as one of the reasons for this. Furuta and Takahashi (2014, 2015) calculated indoor dose rate distributions with a Monte Carlo photon transport code to evaluate the influence of radionuclides deposited on outdoor ground. However, their calculations did not consider the influence of radionuclides on indoor surfaces. Upon a radioactive plume passing over a house, a portion of the radionuclides can infiltrate into the house through openings such as windows, doors, and vents, and can then

deposit near the openings as well as on indoor surfaces. Hence, the indoor external dose should be assessed by taking into account the influence of radionuclides deposited on indoor surfaces as well as those on outdoor ground. However, the indoor external dose rate cannot be assessed accurately because the infiltration pathways and indoor distribution of radionuclides are unclear.

Therefore, this study aims to accurately assess the doses to people living in areas contaminated by the Fukushima-Daiichi accident. In this study, outdoor and indoor dose rates at the center of rooms and near openings were measured in houses in the difficult-to-return area. The indoor dose rates were calculated with a Monte Carlo photon transport code, with the following aims: i) to investigate the difference between indoor and outdoor dose rates, ii) to examine the indoor distribution and main infiltration pathways of radionuclides, and iii) to explore the contribution of various contaminated surfaces to indoor dose rate measurements.

2. Materials and Methods

2.1 Experimental methods

In October and November, 2015, ambient dose equivalents (hereinafter dose rates) were measured by a NaI(Tl) scintillation survey meter (Hitachi Aloka Medical Ltd., TCS-171B) in eight houses (referred to as Houses A–G) in Futaba town and Ohkuma town, which are located in the difficult-to-return area in Fukushima Prefecture. The measurements were carried out as follows: Dose 1 was measured at a height of 1 m from ground level at the four corners outside a house, Dose 2 was measured at a height of 1 m from floor level in the center of a room, and Dose 3 was measured at a height of 5 cm from the indoor surfaces of the floor, wall,

and ceiling.

To examine the indoor distribution and main infiltration pathway of radionuclides, Dose 3 was measured at the center of the floor and ceiling and on the surfaces around openings (which are defined as windows, doors, ventilations, air conditioning outlets, and sockets in this paper). It is reported that the inflow of air from such openings is large (Murakami and Yoshino, 1983). Dose 3 around the openings was measured on surfaces about 10 cm and more than 50 cm from each opening.

It is reported that the indoor deposition rate of radionuclides depends on the surface-area-to-volume ratio of a room (Fogh et al., 1997). In this study, Dose 3 was also measured on the surfaces of a bookshelf filled with books and near the bookshelf to examine the surface area dependence of the deposition amount. During the measurement of Dose 3, the detector was covered with a 1.5-cm-thick cylindrical Pb collimator to eliminate the influence of radiations from outside the target area.

2.2 Computational methods

To explore the contribution of various contaminated surfaces to the indoor dose rates, dose rates were calculated with the Monte Carlo photon transport code EGS5 (Hirayama et al., 2004). We constructed a model house of 12 m × 8 m × 5.5 m (Fig. 1) with a first floor and second floor. The thickness of the walls, roof, and floorboards was assumed to be 4 cm based on Furuta and Takahashi (2015). The density and elemental composition of the walls, roof, floor, and soil were based on Furuta and Takahashi (2014, 2015). Cs-137 was assumed to be distributed uniformly ($= 1 \text{ Bq m}^{-2}$) at the following locations: p(i) on the outdoor

ground surface, except for under the house; p(ii) in the ground, except for under the house, with a depth

profile of concentration C described by an exponential function:

$$C = C_0 \exp\left(-\frac{d}{\beta}\right),$$

where C_0 is the concentration on the surface, d is depth (g cm^{-2}), and β is often called the relaxation mass

depth (g cm^{-2}) and was set to 3 g cm^{-2} based on Matsuda et al. (2015); p(iii) on the outdoor wall; p(iv) on the

roof; p(v) on the ground floor indoor wall; and p(vi) on the first floor floorboards. The detector was set at the

measurement point shown in Fig. 1. Additionally, the Pb collimator was set to reproduce the measurements.

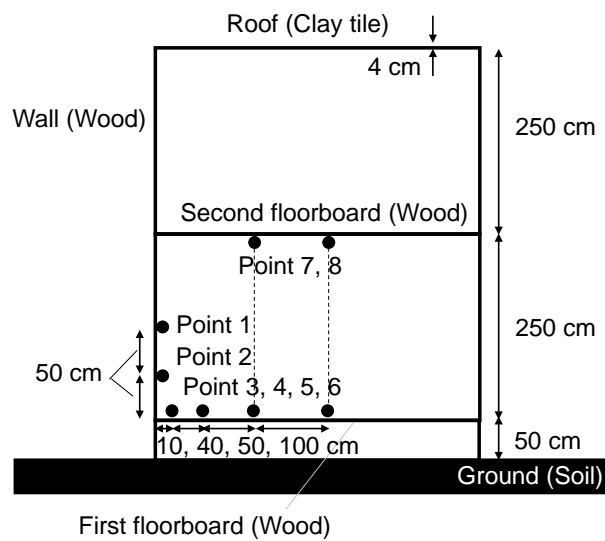


Fig. 1. Layout of the model house and dose rate evaluation points.

3. Results and discussion

3.1 Indoor and outdoor dose rates

This section reports the investigation of the difference between Doses 1 and 2 (shown in Table 1). Dose 1 was measured at the point closest to the measurement point of Dose 2. Dose 1 values differed among the houses due to differences in the deposition amount of radionuclides, and ranged from 0.83 to 15.96 $\mu\text{Sv h}^{-1}$. It is apparent that Dose 1 is governed by artificial radiation because the values are much higher than the natural radiation level, which is less than 0.06 $\mu\text{Sv h}^{-1}$ in this area (Minato, 2006). Therefore, the discussion in this paper is not considered the influence of natural radiation. Dose 2 differed among the houses and ranged from 0.47 to 8.31 $\mu\text{Sv h}^{-1}$; inside the houses, the values of Dose 2 were higher at the same locations where Dose 1 was higher.

The ratio of Dose 2 to Dose 1 was different among houses and rooms and ranged from 0.19 to 0.76. Except for House F, the ratios of Dose 2 to Dose 1 were 0.4–0.6 in almost all rooms, and these values were similar to the ratios examined in Fukushima Prefecture by other authors (Yoshida et al., 2014; Matsuda et al., 2017). However, there were some houses where the difference in the ratio among the rooms was approximately three times that in House G. Furuta and Takahashi (2014) suggested that this ratio depends on factors such as distance from a wall, presence of windows, and outdoor environment. Yoshida et al. (2014) suggested that one reason why the ratios are different among houses is the presence of radionuclides in a room. It is likely that the differences in the ratio in the present study were also caused by this reason.

Table 1 Measured outdoor and indoor dose rates and the ratios of indoor to outdoor dose rate.

House	Dose rate ($\mu\text{Sv h}^{-1}$)		Dose 2/ Dose 1	House	Dose rate ($\mu\text{Sv h}^{-1}$)		Dose 2/ Dose 1
	Indoor (Dose 2)	Outdoor (Dose 1)			Indoor (Dose 2)	Outdoor (Dose 1)	
A	2.86	4.90	0.58	E	0.49	0.93	0.53
	3.10	4.90	0.63		0.48	1.08	0.45
	2.97	4.90	0.61		0.47	1.17	0.40
	3.49	8.35	0.42	F	1.17	5.08	0.23
	2.23	6.10	0.37		1.22	4.67	0.26
	2.25	6.10	0.37		0.92	4.67	0.20
	2.46	6.60	0.37		0.94	4.91	0.19
	2.16	4.91	0.44		1.30	4.67	0.28
2.59	4.91	0.53	0.95		3.14	0.30	
1.46	3.08	0.47	G	2.02	4.76	0.42	
1.82	5.08	0.36		2.10	4.76	0.44	
1.92	5.08	0.38		2.72	6.38	0.43	
1.53	4.67	0.33		2.61	6.57	0.40	
1.32	3.14	0.42		1.46	6.57	0.22	
1.52	3.14	0.48		1.50	6.57	0.23	
1.37	3.14	0.44		1.57	6.57	0.24	
2.20	3.14	0.70		1.53	3.21	0.48	
B	8.31	15.48	0.54	2.03	3.21	0.63	
	4.46	15.48	0.29	H	2.83	5.12	0.55
	7.79	15.29	0.51		3.60	5.93	0.61
	6.81	15.29	0.45		3.59	5.93	0.61
	6.32	15.29	0.41		3.30	6.77	0.49
	6.55	15.29	0.43		3.32	6.77	0.49
	5.44	15.65	0.35		3.52	8.31	0.42
	4.71	15.96	0.29		3.14	8.31	0.38
4.08	11.51	0.35					
C	0.63	0.83	0.75				
	0.57	0.83	0.68				
	0.72	0.95	0.76				
	0.78	1.47	0.53				
	0.78	1.26	0.62				
	0.71	1.26	0.56				
	0.67	1.26	0.53				
	0.68	1.26	0.54				
	0.62	1.13	0.55				
	0.65	1.34	0.48				
	0.65	1.34	0.48				
	0.63	1.34	0.47				
	0.57	1.34	0.42				

3.2 Dose rate distribution near openings

Values of Dose 3 at a point 10 and 50 cm distant from the openings [Dose 3 (10) and Dose 3 (50)]

are shown in Table 2. Dose 3 (10) was about 30% higher than Dose 3 (50), except at sockets and air

conditioning outlets. The ratio of Dose 3 (10) to Dose 3 (50) on the floor was about 15% higher than that on the wall. These characteristics were commonly found in all studied houses.

The 1.5-cm-thick Pb collimator used in this study could not completely eliminate the contribution of radiation from outside the target area. Therefore, radiations from radionuclides deposited on the outdoor ground, outdoor walls, and roofs, as well as on the indoor walls and floor, can contribute to that measured by the detector. In this study, measurement points on walls near the openings were beneath the openings; i.e., the measurement point of Dose 3 (10) was higher than that of Dose 3 (50). It is expected that Dose 3 (50) is higher than Dose 3 (10) because the distance between the detector and radionuclides deposited on the outdoor ground and indoor floor is shorter. In reality, however, Dose 3 (50) was lower than Dose 3 (10), except at sockets and air conditioning outlets. This result implies that radionuclides infiltrated into the room through the openings and deposited on surfaces near those openings.

For the floor measurements, the measurement points distant from the openings [Dose 3 (50)] were further from indoor walls than those near the openings [Dose 3 (10)]; therefore, the contribution of radionuclides deposited on the outdoor ground was lessened. In reality, Dose 3 (50) was again less than Dose 3 (10). The fact that the ratio of Dose 3 (10) to Dose 3 (50) on the floor was higher than that on the wall can be attributed to the influence of both deposition near the openings and distance between the detector and the radionuclides.

This paragraph describes the reason why the ratio of Dose 3 (10) to Dose 3 (50) was approximately 1 for sockets and air conditioning outlets. The pathway through a socket is more complicated than that

through other openings. It is likely that radionuclides hardly infiltrated a room through sockets, because radionuclides can deposit on wall surfaces along their path of travel. In additions, it is highly probable that radionuclides hardly deposited on walls near air conditioning outlets, because air conditioning units do not generally exchange air directly between indoor and outdoor environments.

Table 2 Measurements of dose rate ($\mu\text{Sv h}^{-1}$) near openings.

House	Distance from opening		Dose 3 (10)/ Dose 3 (50)	Surface (Wall or Floor)	Opening	House	Distance from opening		Dose 3 (10)/ Dose 3 (50)	Surface (Wall or Floor)	Opening
	10 cm [Dose 3 (10)]	50 cm [Dose 3 (50)]					10 cm [Dose 3 (10)]	50 cm [Dose 3 (50)]			
A	0.60	0.48	1.25	Floor	Door	E	0.15	0.13	1.15	Wall	Vent
	0.59	0.56	1.05	Floor	Window		0.24	0.23	1.04	Wall	Window
	0.53	0.36	1.46	Floor	Window		0.20	0.18	1.09	Wall	Window
	0.75	0.50	1.51	Floor	Window		0.20	0.16	1.22	Wall	Window
	0.94	0.60	1.58	Floor	Window		0.20	0.18	1.15	Wall	Window
	0.78	0.67	1.17	Floor	Window		0.14	0.15	0.96	Wall	Socket
B	0.75	0.71	1.06	Wall	Vent	0.13	0.10	1.26	Floor	Door	
	0.63	0.48	1.31	Wall	Window	0.42	0.39	1.07	Wall	Vent	
	0.58	0.49	1.18	Wall	Window	0.32	0.31	1.01	Wall	AC*	
	0.27	0.24	1.13	Floor	Door	0.40	0.30	1.33	Floor	Window	
	0.77	0.52	1.49	Floor	Window	0.31	0.22	1.43	Floor	Window	
C	1.95	1.59	1.23	Wall	Vent	0.60	0.39	1.53	Floor	Window	
	2.64	2.55	1.04	Wall	Window	0.29	0.17	1.69	Floor	Door	
	2.58	2.44	1.05	Wall	Window	0.52	0.47	1.09	Wall	Vent	
	2.54	2.59	0.98	Wall	Socket	0.52	0.44	1.18	Wall	Window	
	2.57	2.59	0.99	Wall	Socket	0.91	0.80	1.14	Wall	Window	
	2.02	0.97	2.08	Floor	Door	0.65	0.68	0.96	Wall	AC*	
	1.95	1.10	1.77	Floor	Window	0.30	0.31	0.97	Wall	Socket	
1.61	1.07	1.50	Floor	Window	0.36	0.32	1.15	Floor	Door		
D	0.32	0.33	0.97	Wall	Vent	0.95	0.52	1.82	Floor	Window	
	0.30	0.27	1.13	Wall	Vent	0.51	0.29	1.75	Floor	Window	
	0.27	0.19	1.42	Wall	Window	0.90	0.60	1.50	Floor	Window	
	0.26	0.20	1.30	Wall	Window	0.74	0.59	1.26	Floor	Window	
	0.14	0.15	0.93	Wall	Socket	1.17	1.11	1.05	Wall	Vent	
	0.26	0.20	1.30	Floor	Window	0.88	0.84	1.05	Wall	Window	
	0.22	0.19	1.14	Floor	Window	1.31	1.09	1.20	Wall	Window	
						1.04	1.02	1.02	Wall	Window	
					0.44	0.37	1.19	Floor	Door		
					0.99	0.87	1.15	Floor	Window		
					0.64	0.46	1.40	Floor	Window		
					1.18	0.73	1.63	Floor	Window		
					0.66	0.38	1.74	Floor	Door		

* AC means an Air Conditioning outlet.

3.3 Deposition of radionuclides on a household items having a large specific surface area

In this section, the influence of household items having a large specific surface area, such as a bookshelf, on deposition amount is described. Dose 3 values for a bookshelf surface and a wall surface near the bookshelf are shown in Table 3. The ratio of these values was approximately 1. Indoor deposition occurs in two stages: the convection of radionuclides with airflow from the center of a room to a surface, and transport across the boundary layer by Brownian diffusion (Hussein et al., 2014; Liu and Nazaroff, 2003). Liu and Nazaroff (2003) suggested that significant enhancement in particle deposition is not expected in the presence of a rough surface for particle diameters less than 0.1 μm and larger than 0.4 μm . The activity size distributions of ^{134}Cs and ^{137}Cs , as measured by Kaneyasu et al. (2012) in Tsukuba since April 28, 2011, had a distinct maximum in the 0.49–0.7 μm range. Based on these facts, it is considered that particulate radioactivity discharged from the Fukushima-Daiichi accident did not deposit locally on rough surfaces such as bookshelves; this is consistent with our measurements.

Table 3 Measurements of Dose 3 ($\mu\text{Sv h}^{-1}$) on a bookshelf surface and a wall surface in close proximity.

Bookshelf	Wall	Bookshelf/Wall
0.16	0.16	1.00
0.21	0.21	1.00
0.37	0.38	0.97
0.85	1.02	0.83

3.4 Comparison between dose rates on the floor and ceiling

Dose 3 values for the floor and ceiling at the center of a room are shown in Table 4. Dose 3 on the ceiling was about 20% higher than that on the floor. The deposition rate onto a floor is ten times larger than that onto a ceiling because of gravity deposition (Sehmel, 1973). Therefore, it is expected that the amount of radionuclides deposited onto a floor is larger than that onto a ceiling. However, our measurements were inconsistent with this expectation. The contribution of radionuclides deposited on the outdoor ground is greater at higher measurements near the center of a room because artificial radionuclides due to the accident are hardly penetrating under the floor and the detectable area is large. In addition, the contribution of radionuclides deposited onto the roof is greater at higher measurements, although the measurements on roofs were not performed. It is likely that Dose 3 on the ceiling was higher than that on the floor because of these reasons. The amount of radionuclides on the ceiling and floor cannot be determined by dose rate measurement alone unless these contributions are eliminated.

Table 4 Measurements of Dose 3 on the floor and ceiling at the center of a room.

House	Dose rate ($\mu\text{Sv h}^{-1}$)		Floor/Ceiling
	Floor	Ceiling	
A	0.70	1.08	0.65
B	0.35	0.78	0.45
C	0.59	1.63	0.36
D	0.14	0.21	0.67
E	0.14	0.17	0.82
F	0.19	0.31	0.61

3.5 Dose rate calculated with a Monte Carlo electron-photon transport code

The calculation results are shown in Table 5. For measurements on the wall (Points 1 and 2), the largest contribution to dose rate was from the radionuclides on the outdoor ground (if radionuclides did not infiltrate the ground). For measurements on the floor (Points 3–6), the largest value was that on the indoor floor. For measurements on the ceiling (Points 7 and 8), the contribution from all locations was almost equal.

The next two paragraphs describe the comparison of the calculated dose rates at Points 1–6, in order to examine the difference between the measured Dose 3 (10) and Dose 3 (50). In comparing the calculated dose rates at Points 1 and 2, it is seen that the dose rate at the lower height was larger in the case of radionuclides on the ground surface [p(i)]. On the other hand, in the case of the radionuclides in the ground [p(ii)], the dose rate at the lower height was smaller. The difference between the measurement height dependence of dose rate in p(i) and (ii) is due to the attenuation effect of soil (Furuta and Takahashi, 2014). Even if the radionuclides infiltrate the ground deeper, the difference between the measured Dose 3 (10) and Dose 3 (50) of about 20% cannot be reproduced. In addition, according to Matsuda et al. (2015), there is a small possibility that β is larger than 3 g cm^{-2} . The contribution of the calculated dose rate from the radionuclides deposited on the roof and walls was almost independent of the measurement height and the contribution at the lower height from the radionuclide deposited on the indoor floor was larger. These calculation results suggest that the higher dose rates measured at points near the openings are probably caused by the presence of radionuclides near the openings.

In comparing the calculated dose rates at Points 3–6, the contribution from radionuclides deposited on the outdoor ground and wall was smaller, while that on the indoor floor was larger at the farther point from the indoor wall. In an assumption of the same activity concentrations at all locations (as shown in Table 5), the calculated total dose rate at the farther point from the wall was smaller; although at Points 3–6, the contribution of the calculated dose rates from the radionuclides on the indoor floor was largest. The relationship between dose rate and distance from the wall is consistent with the measurements. In addition, the ratio of the calculated dose rates at Points 3 and 4 was about 1.4, i.e., close to the ratio of measured Dose 3 (10) to Dose 3 (50). However, an assumption of the same activity concentrations at all locations is unrealistic because the amount of radionuclides deposited on a wall surface is smaller than that on the ground or floor (Sehmel, 1973; Roed, 1987). Given the differences in deposition rate to the surfaces, the contribution of the calculated dose rate from the radionuclides deposited on the wall should be smaller; therefore, the ratio of the calculated dose rates at Points 3 and 4 also becomes smaller. To reproduce the measurement ratio, the activity on the indoor floor should be smaller than that on the outdoor ground, and/or more radionuclides should be present near the openings.

This paragraph describes the comparison between the calculated dose rates at Points 5 and 7 and those at Points 6 and 8, in order to examine the difference between the measured dose rates on the ceiling and floor. At the higher point, the contribution of the calculated dose rates from the radionuclides on the indoor floor was smaller, while those on the outdoor ground, roof, and outdoor wall were larger. At Points 5 and 7 or

Points 6 and 8, the difference between the calculated dose rates from the radionuclides on the outdoor ground and on the roof ranged from 1.6 to 2.8 times greater. In the measurements, the difference between the dose rates on the floor and ceiling ranged from 1.2 to 2.8 times as shown in Table 4. As mentioned before, the activity on the indoor floor may be smaller than that on the outdoor ground. Additionally, in areas with large dose rates, artificial radionuclides govern the measured dose rates as mentioned in section 3.1. Given these assumptions, the calculations almost reproduce the measurements. This result reveals that the dependence of dose rate on the measurement height is caused mainly by the difference in contribution of radiation from the outdoor ground and/or the presence of radionuclides on a roof. According to some references, the presence of radionuclides on a roof or roof-spaces was confirmed by the comparison of measured dose rates from the first floors of one-story and two-story houses and dry smear tests (Matsuda et al., 2017; Yoshida et al., 2016). The results in this study are consistent with those in these references.

Table 5 Calculated dose rates (nSv h⁻¹). Activity concentration of Cs-137 was set to 1 Bq m⁻².

Point	on ground p(i)	in ground p(ii)	on outdoor wall p(iii)	on roof p(iv)	on indoor wall p(v)	on floor p(vi)
1	0.154	0.068	0.065	0.003	0.085	0.033
2	0.160	0.066	0.065	0.003	0.084	0.047
3	0.048	0.026	0.049	0.006	0.055	0.075
4	0.030	0.013	0.026	0.007	0.025	0.080
5	0.025	0.010	0.019	0.008	0.014	0.083
6	0.022	0.009	0.013	0.010	0.009	0.087
7	0.040	0.023	0.029	0.022	0.012	0.019
8	0.039	0.021	0.016	0.026	0.009	0.021

4. Conclusion

Indoor and outdoor dose rates were measured in eight houses where radionuclides were deposited on indoor surfaces to examine the distribution and main infiltration pathways of radionuclides in a house. These measurements indicated that vents, windows, and doors are the main infiltration pathways, whereas radionuclides hardly infiltrate through sockets and air conditioning outlets. A significant difference between the measured dose rates on a bookshelf surface (where the surface area is locally large) and on the wall near the bookshelf was not found. This measurement result implies that radionuclides discharged from the Fukushima-Daiichi nuclear power plant did not deposit locally on rough surfaces (such as bookshelves).

The contribution of various contaminated surfaces to indoor dose rate was examined by a dose rate calculation using EGS5. The calculations suggested that radionuclides likely deposited near openings. However, we cannot reveal exactly the distribution of radionuclides on the ceiling and floor near the openings because we cannot distinguish the complicated contribution of radiations from the many locations surrounding the detector using only the dose rate measurements. This measurement method should be combined with other measurement methods that can measure radioactivity on a surface; for example, a smear method could be used to examine, in more detail, the distribution and main infiltration pathways of radionuclides in a house.

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