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Assessment of localized and resuspended ^{137}Cs due to decontamination and demolition in the difficult-to-return zone of Tomioka town, Fukushima Prefecture

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EDITOR'S NOTE:

This article is part of the special series "Lessons Learned & Consequences of the Fukushima-Daiichi Nuclear Power Plant Accident, 10 Years Later." Articles in this series report on the progress of the environmental monitoring, ecological recovery and resilience, and regulatory oversight associated with the 2011 Fukushima Daiichi nuclear power plant accident in Japan.

Abstract

As the next step that occurred more than one decade after the accident at the Fukushima Dai-ichi Nuclear Power Station (FDNPS), decontamination and demolition have been carried out in the Specified Reconstruction and Revitalization Base (SRRB) of the difficult-to-return zone around the FDNPS. However, the risk of internal exposure among workers due to airborne dust inhalation after building demolition operations has not been sufficiently evaluated. To evaluate the working environment and internal exposure risk due to inhalation in the SRRB of Tomioka town, Fukushima Prefecture, the cesium-137 (^{137}Cs) radioactivity levels in the airborne dust at building demolition sites were analyzed using gamma spectrometry. The ^{137}Cs radioactivity levels and resuspension factors of the airborne dust at the subject building sites in the difficult-to-return zone remained at high levels compared with those of the control, which was located in the evacuation order-lifted area in Tomioka town. However, the ^{137}Cs radioactivity levels did not increase significantly, despite demolition operations that used heavy machinery. In this case, no substantial increases in accident-derived ^{137}Cs levels due to decontamination and demolition in the SRRB of Tomioka town, Fukushima Prefecture, were observed in the airborne dust samples, which suggests that the ^{137}Cs radioactivity in the airborne dust is primarily associated with particles that are resuspended by localized winds accompanied by the transfer of construction vehicles as opposed to the decontamination and demolition operations. However, the internal exposure doses due to aspirating airborne dust containing ^{137}Cs were extremely low compared with the estimated annual effective doses of decontamination workers or the limits recommended by the Japanese government. Additionally, countermeasures such as wearing protective masks could help reduce the on-site inhalation of soil-derived radionuclides. *Integr Environ Assess Manag* 2022;18:1555–1563. © 2022 The Authors. *Integrated Environmental Assessment and Management* published by Wiley Periodicals LLC on behalf of Society of Environmental Toxicology & Chemistry (SETAC).

KEYWORDS: Demolition, Difficult-to-return zone, Internal exposure, Radiocesium, Resuspension

[Correction added on October 28, 2022, after first online publication: The data availability statement was changed to modify "manuscript" to "article" and "supplementary materials" to "Supporting Information Files".]

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INTRODUCTION

A decade has passed since a major nuclear accident occurred at the Fukushima Dai-ichi Nuclear Power Station (FDNPS), which resulted from severe damage to the infrastructure and included the loss of electrical power and reactor cooling function that were caused by the magnitude 9.0 Great East Japan Earthquake and the resulting tsunami (International Atomic Energy Agency [IAEA], 2015). The nuclear accident triggered the release of radionuclides such as iodine-131 (half-life [$t_{1/2}$] = 8 days), cesium-134 (^{134}Cs; $t_{1/2}$ = 2.1 years), and cesium-137 (^{137}Cs; $t_{1/2}$ = 30.1 years) into the atmosphere, which were eventually deposited in the surrounding terrestrial and marine environments (IAEA, 2015). After the FDNPS accident, decontamination and building demolition operations were conducted in residential areas and their adjacent forests, farmlands, and roads in Fukushima Prefecture, but excluded the difficult-to-return zones (estimated at >50 mSv/y by the national government as of March 2012); these operations were completed on March 19, 2018 (Ministry of the Environment [MOE], 2019). On the other hand, according to the Act on Special Measures for the Reconstruction and Revitalization of Fukushima that was outlined in 2017, six municipalities (e.g., Futaba town, Okuma town, Namie town, Tomioka town, Iitate village, and Katsurao village), located around the FDNPS, were making plans to construct a Specified Reconstruction and Revitalization Base (SRRB) in the difficult-to-return zones by aiming to lift evacuation orders and allow residents of the difficult-to-return zones to return home (MOE, 2019). At present, the Ministry of the Environment (MOE) is decontaminating the surface soil and conducting building demolitions in the difficult-to-return zones around these areas. In particular, the MOE is conducting decontamination and demolition efforts in many types of buildings, such as ferroconcrete buildings and wooden houses, with the aim of lifting the evacuation orders at the SRRB in the spring of 2022 or by the spring of 2023 (MOE, 2019).

Tomioka town is a town in Fukushima Prefecture that is located within a 20-km radius of the FDNPS. On April 1, 2017, the national government declared that the residents who had previously resided in approximately 88% of the town could return to their homes, as the radiation dose rates had reached acceptable levels (e.g., <20 mSv/y) (Fukushima Prefectural Government, 2014). Our previous studies have found that the external and internal exposure doses in residential living spaces (e.g., the evacuation order-lifted area) of Tomioka town have decreased to levels close to the public dose limit (e.g., 1 mSv/y) in the recovery and reconstruction period after the FDNPS accident (Matsuo et al., 2019; Yamaguchi et al., 2021).

In the difficult-to-return zone in Tomioka town, decontamination and demolition have been carried out to achieve the goal of lifting the evacuation order for the entire SRRB. For cases where decontamination activities and the related workers are engaged in workplaces such as those in the SRRB, the guidelines on the prevention of radiation hazards for workers, which were issued by the Japanese government (Ministry of Health, Labour and Welfare [MHLW], 2018), should be followed. Because the employers have decontamination workers who are engaged in highly contaminated environments due to soils and houses, including radiocesium, workers should measure their internal exposure doses in addition to receiving external exposure dose measurements by using personal dosimeters for those cases in which the average ambient dose rate exceeds 2.5 μSv/h (equivalent to 5 mSv/y based on 40 h/week and 52 weeks/year) (MHLW, 2018). Measurements of the internal exposure doses should be carried out using the methods described in the guidelines to control the concentrations in dust (the lower limit is 10 mg/m^3) and in soils contaminated with radioactivity (500 000 Bq/kg) during the decontamination work, according to the concentrations of radioactive materials that were released by the FDNPS accident (MHLW, 2018). However, the risk of internal exposure among workers due to airborne dust inhalation during building demolition has not been sufficiently evaluated. In particular, the national government and Tomioka town office pay attention to their internal radiation exposures due to the ^{137}Cs that is present around residents' houses, although they may not be directly exposed to airborne dust. Radiocesium has a relatively long half-life, and still exists in soil and plant samples that were collected around the FDNPS, including Tomioka town (Cui et al., 2020; MHLW, 2018; Tomioka Town Office, 2021). Additionally, ^{134}Cs and ^{137}Cs were the dominant radionuclides comprising the indoor surface contamination of wooden houses located within the evacuation areas after the FDNPS accident, and there was a distance dependence from the FDNPS (Yoshida et al., 2016). Therefore, in the present study, the ^{137}Cs radioactivity levels in the airborne dust at building demolition sites were analyzed using gamma spectrometry to evaluate the working environments and internal exposure doses due to inhalation in the SRRB of the difficult-to-return zone of Tomioka town, Fukushima Prefecture.

MATERIALS AND METHODS

Study site

The FDNPS (37°25′N, 141°02′E) is located on the east coast of Honshu Island, Japan, approximately 200 km northeast of Tokyo. "Building G" (37°22′00″N, 141°00′09″E) and "Building Y" (37°22′02″N, 140°59′48″E), which are situated in the difficult-to-return zone of Tomioka town, are located 6.65 and 6.79 km southwest of the FDNPS, respectively (Figure 1). The difficult-to-return zone, which covers approximately 12% of the total area of Tomioka town, is located within a 10-km radius of the FDNPS and is divided by the main road (R6) between the Yonomori and Oragahama districts (Figure 1) (MOE, 2019; Reconstruction Agency, 2018). Primary decontamination efforts in the Yonomori district, which was designated as a reconstruction and revitalization area by the Japanese government, started in July 2018 (Cui et al., 2020; MOE, 2019). Decontamination work in the Yonomori district has involved cleaning paved surfaces, roadsides, and street drains; removing topsoil; weeding and pruning trees; washing building surfaces; and demolishing buildings (Cui et al., 2020; MOE, 2019). On the other hand, a portion of the Oragahama district was designated as a radioactive waste storage area and forested area (e.g., nondecontaminated) in 2014. In the present study, the Yonomori district, including "Building G" and "Building Y" in the difficult-to-return zone of Tomioka town, is referred to as the decontamination and demolition area.

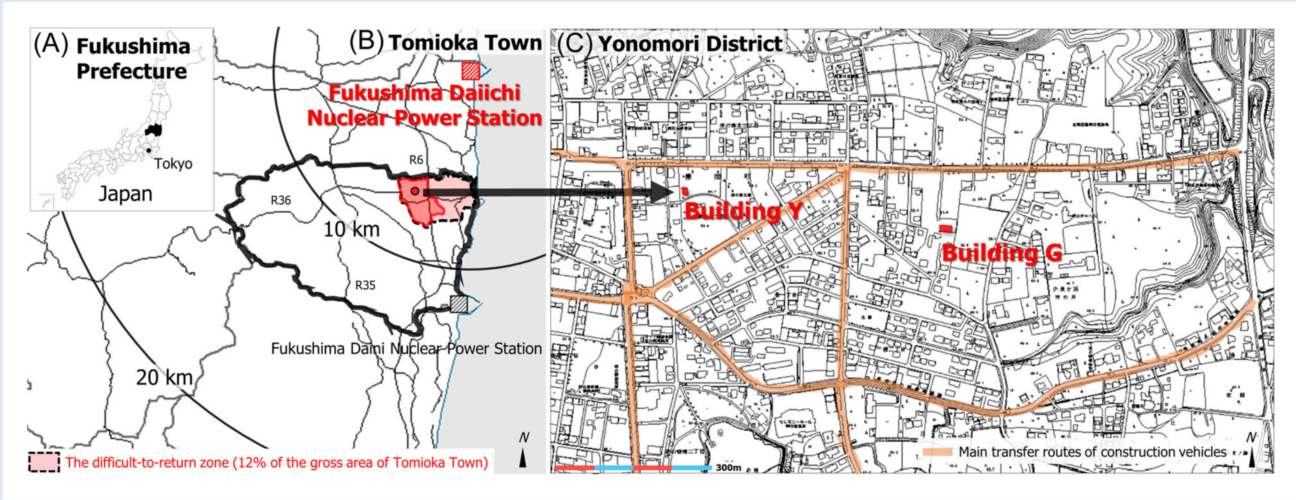

FIGURE 1 Location of airborne dust sampling sites "Building G" and "Building Y," Yonomori district, Tomioka town, Fukushima Prefecture. (A) Outline map of Japan. A black-painted area shows the location of Fukushima Prefecture and (B) location of Tomioka town, Fukushima Prefecture. The bold line shows a boundary of Tomioka town; 10 and 20 km show the radius from Fukushima Dai-ichi Nuclear Power Station. (C) Enlarged map in Yonomori district of the difficult-to-return zone, Tomioka town (scale 1:5000). Map (B) reprinted from Green Map III under a CC BY license, with permission from Tokyo Shoseki Co., Ltd.; original copyright 2003. Map (C) reprinted from Administration-support GIS (ALANDIS) under Tomioka town office license, with permission from Asia Air Survey Co., Ltd. (https://www.ajiko.co.jp/en/service/mapping.html)

Airborne dust sampling

Airborne dust samples (e.g., particles captured on filters: filter dust samples) were collected at the site of "Building G" between September 5, 2019 and February 27, 2020 (before the decontamination and demolition work) and between May 27 and August 18, 2020 (during the decontamination and demolition work). Filter dust samples were also collected at the site of "Building Y" between February 2 and 3, 2021 (before the decontamination and demolition work) and between March 3 and 24, 2021 (during the decontamination and demolition work). For comparison, filter dust samples were also collected at the Tomioka Town Office (37°20′N, 141°0′E) in the evacuation order-lifted area between August 13, 2019 and May 22, 2020, and between April 4 and 12, 2021. All samples were collected on 203 × 254-mm quartz-fiber filters (GB100R-810A; SIBATA Scientific Technology Ltd.) at flow rates of 1000 L/min using a high-volume air sampler with a single-stage impactor (HV-1000R; SIBATA Scientific Technology Ltd.). The cumulative flows were in the range of 349.8–403.9 m^3 (5 h 50 min to 6 h 45 min) for "Building G" and 250.5–429.9 m^3 (4 h 10 min to 7 h 10 min) for "Building Y." The collection method was certified based on the standard method series of radioactivity measurements certified by the Ministry of Education, Culture, Sports, Science and Technology and the Nuclear Regulation Authority in Japan (Environmental Radioactivity and Radiation in Japan, 2021).

Measurement of radionuclides using gamma spectrometry

After collection, all samples (single tared quartz-fiber filters) were dried at room temperature (15–25 °C) for one month. After preparation, the filter dust samples, including the circular quartz-fiber filters, were placed into polypropylene containers (U8), and their masses (i.e., the airborne dust, including the quartz-fiber filter) were then calculated and measured based on the height of the samples (the airborne dust, including the quartz-fiber filter) before measuring the radioactivity. The radioactivity levels in the filter dust samples collected at the site of "Building G" were analyzed from five samples collected in 2019 (before decontamination and demolition) and in 19 samples collected in 2020 (during decontamination and demolition). In addition, the radioactivity levels in the filter dust samples collected at the site of "Building Y" were analyzed in two samples (before decontamination and demolition) and in 13 samples (during decontamination and demolition) that were collected in 2021. As a control, the radioactivity levels were analyzed for five filter dust samples that were collected at the Tomioka town office during the present study period. Finally, the samples were analyzed using a high-purity germanium detector (ORTEC GMX series; Ortec International Inc., Ltd.) coupled to a multichannel analyzer (MCA7600; Seiko, EG&G Co., Ltd.) for 80 000 s. The measurement time was set to the level at which the target radionuclide (^{137}Cs) was detectable at 0.1 mBq/m^3. The gamma-ray peaks adopted for the measurements were 604.66 keV for ^{134}Cs (2.1 years) and 661.64 keV for ^{137}Cs (30 years). The decay corrections were set to the sampling dates. Detector efficiency calibrations for different measurement geometries, including the density and thickness of the samples, were performed using mixed activity standard volume sources (Japan Radioisotope Association). Detector efficiency calibrations for different measurement geometries were performed using mixed activity standard volume sources, including radiocesium (Japan Radioisotope Association).

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All soil samples were processed and analyzed at Nagasaki University, Nagasaki, Japan. Sampling, processing, and analysis were carried out using the standard method series of radioactivity measurements certified by the Ministry of Education, Culture, Sports, Science and Technology and the Nuclear Regulation Authority in Japan (Environmental Radioactivity and Radiation in Japan, 2021).

Internal radiation exposure by inhalation

After measuring the filter dust samples, the internal effective doses due to inhalation at the "Building G" and "Building Y" sites were estimated based on the ^{137}Cs radioactivity levels using the following equation:

$$H_{\text{int}} = C \cdot D_{\text{int}} \cdot e, \quad (1)$$

where C is the average detected ^{137}Cs radioactivity (Bq/m^3) (e.g., ^{137}Cs concentrations in filter dust samples), D_{int} is the effective dose coefficient for inhaled particulates (e.g., activity median aerodynamic diameters of 1 and 5 μm) for workers based on the International Commission on Radiological Protection (ICRP) Publications 68 and 119 (e.g., 1 μm: 4.8×10^{-6} mSv/Bq and 5 μm: 6.7×10^{-6} mSv/Bq for ^{137}Cs) (ICRP, 1994, 2012), and e is quoted from the reference values for the daily ventilation rates used for dosimetric modeling for adult workers based on ICRP Publication 89 (e.g., 13.5 m^3/8 h/d for decontamination and demolition workers and 9.6 m^3/8 h/d for office workers) (ICRP, 2002). In this methodology, the average daily inhalation was estimated based on internal exposure doses due to inhalation during occupational activities among heavy manual workers.

Limited resuspension factor (L-RF)

To evaluate ^{137}Cs radioactivity levels in airborne dust, including resuspended soil particles that are due to ^{137}Cs contamination of the surface soil, the L-RF values used in the present study were calculated using the following equation:

$$K = {^{137}Cs_{\text{invent}}}/{^{137}Cs_{\text{cont}}}, \quad (2)$$

where $^{137}Cs_{\text{invent}}$ is the ^{137}Cs radioactivity level of airborne dust (Bq/m^3) and $^{137}Cs_{\text{cont}}$ is the ^{137}Cs contamination in the surface soil layer (0–0.5 cm) (Bq/m^2) collected in the present study (Abe et al., 2021; Ochiai et al., 2016). Because the decontamination in the Yonomori district of the SRRB used the same schedule and technical methods, the surface soil contamination levels that were closest to the airborne dust sampling points (in time) were adopted as the reference values (Cui et al., 2020). Actually, there are some parks, agricultural lands, forests (the boundary with the SRRB), and deposited grounds around the "Building G" and "Building Y" sites at the SRRB in the Yonomori district (Supporting Information: Appendix S2) (MOE, 2019).

RESULTS

In the present study, the only prevalent dose-forming artificial radionuclide that was recovered from the filter dust samples in the difficult-to-return zone of Tomioka town was ^{137}Cs. No other artificial radionuclides were detected, except for the natural radionuclides derived from soils and rocks (e.g., uranium-238 series: thorium-234, radium-226, lead-214, bismuth-214, and lead-210; thorium-232 series: actinium-228, thorium-228, lead-212, and tallium-208; and potassium-40) and the natural radionuclide derived from the atmosphere (beryllium-7 [^{7}Be]) (data not shown). First, the ^{137}Cs radioactivity levels in the airborne dust at the "Building G" site in the difficult-to-return zone of Tomioka town, Fukushima Prefecture, between 2019 and 2020 are shown in Table 1 and Supporting Information Appendix S1. The range of the ^{137}Cs radioactivity levels was <0.11–0.73 mBq/m^3 before demolition from September 2019 to February 2020 and <0.10–0.62 mBq/m^3 during demolition from May to August 2020. On the other hand, the range of the ^{137}Cs radioactivity levels at the Tomioka town office (the control building) was <0.034–0.15 mBq/m^3 from August 2019 to May 2020. To evaluate the inhalation risk due to airborne dust, including ^{137}Cs, during decontamination and demolition, the internal exposure dose rates were also estimated to be $<6.8 \times 10^{-6}$–4.7×10^{-5} μSv/d (d refers to daily working hours [8 hours/day]) due to the inhalation of particulates with activity median aerodynamic diameters of 1 μm and/or $<9.5 \times 10^{-6}$–6.6×10^{-5} μSv/d due to the inhalation of particulates with activity median aerodynamic diameters of 5 μm before demolition from September 2019 to February 2020. Moreover, the doses were estimated to be $<6.7 \times 10^{-6}$–4.0×10^{-5} μSv/d due to the inhalation of particulates with median aerodynamic diameters of 1 μm and/or $<9.3 \times 10^{-6}$–5.6×10^{-5} μSv/d due to the inhalation of particulates with median aerodynamic diameters of 5 μm during demolition from May to August 2020. On the other hand, the doses were estimated to be $<1.6 \times 10^{-6}$–7.1×10^{-6} μSv/d due to the inhalation of particulates with median aerodynamic diameters of 1 μm and/or $<2.2 \times 10^{-6}$–1.0×10^{-5} μSv/d due to the inhalation of particulates with median aerodynamic diameters of 5 μm at the Tomioka town office (the control building) from August 2019 to May 2020. According to MOE survey data, the ambient dose rates measured at 1 m above the ground using a survey meter at 23 points around "Building G" were 0.70–2.0 μSv/h before demolition (February 29, 2020) and 0.27–0.56 μSv/h after demolition (from August 28 to September 1, 2020). Therefore, the rates of decrease due to decontamination and demolition ranged from 44% to 83% for each point. Moreover, the L-RFs ranged from 1.1×10^{-8} to 7.5×10^{-8} m^{-1} before demolition from September 2019 to February 2020 and from 0.47×10^{-8} to 2.8×10^{-8} m^{-1} during demolition from May to August 2020. On the other hand, the L-RFs at the Tomioka town office (the control building) ranged from 0.056×10^{-8} to 0.25×10^{-8} m^{-1} from August 2019 to May 2020.

Second, the ^{137}Cs radioactivity levels in the airborne dust at the "Building Y" site in the difficult-to-return zone of Tomioka town, Fukushima Prefecture, in 2021 are shown in Table 2 and Supporting Information Appendix S2. The ranges of the ^{137}Cs radioactivity levels were 0.35–0.36 mBq/m^3 before demolition from February 2 to 3, 2021

TABLE 1 ^{137}Cs radioactivity in the airborne dust at building sites in Tomioka town, Fukushima Prefecture, between 2019 and 2020

Measurement point	Period	*n*	^{137}Cs (mBq/m^3)	Internal exposure dose rate (μSv/d)[a]		L-RFs × 10^{-8} (m^{-1})[b]	Remark
				Inhaled particulates (activity median aerodynamic diameter of 1 μm)	Inhaled particulates (activity median aerodynamic diameter of 5 μm)		
Building G (difficult-to-return zone)	September 2019 to February 2020[c]	4 (3)[d]	<0.11–0.73 (0.085–0.098)[e]	$<6.8 \times 10^{-6}$ to 4.7×10^{-5}[f]	$<9.5 \times 10^{-6}$ to 6.6×10^{-5}	1.1–7.5	Before demolition
	May to August 2020[g]	20 (16)	<0.10–0.62 (0.091–0.11)	$<6.7 \times 10^{-6}$ to 4.0×10^{-5}	$<9.3 \times 10^{-6}$ to 5.6×10^{-5}	0.47–2.8	During demolition
Tomioka town office (evacuation order-lifted area)	August 2019 to May 2020[h]	3 (2)	<0.034–0.15 (0.023–0.097)	$<1.6 \times 10^{-6}$ to 7.1×10^{-6}	$<2.2 \times 10^{-6}$ to 1.0×10^{-5}	0.056–0.25	Control building[i]

Abbreviation: L-RF, limited resuspension factor.
[a]Calculated using Equation (1). Daily ventilation rates were quoted from 13.5 m^3/8 h/d for the decontamination and demolition workers and 9.6 m^3/8 h/d for office workers.
[b]Calculated using Equation (2), $^{137}Cs_{cont}$ (surface soil samples) were 9.7×10^3 Bq/m^2 before demolition, 21.9×10^3 Bq/m^2 during demolition, and 60.6×10^3 Bq/m^2 for the control building.
[c]Airborne dust was collected four times (September 5, 2019, November 21, 2019, December 5, 2019, and February 27, 2020).
[d]Sample number of ^{137}Cs detection.
[e]Detection limit at the time of ^{137}Cs detection.
[f]Minimum–maximum (values less than those indicated were not detected).
[g]Airborne dust was collected 20 times (from May 27 to August 6, 2020).
[h]Airborne dust was collected three times (August 13–14, 2019, February 26, 2020, and May 22, 2020).
[i]Tomioka Town Food Inspection Department within the Tomioka town office.

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TABLE 2 ^{137}Cs radioactivity in the airborne dust at building sites in Tomioka town, Fukushima Prefecture, from January to April, 2021

Measurement point	Period	*n*	^{137}Cs (mBq/m^3)	Internal exposure dose rate (µSv/d)[a]: Inhaled particulates (activity median aerodynamic diameter of 1 µm)	Internal exposure dose rate (µSv/d)[a]: Inhaled particulates (activity median aerodynamic diameter of 5 µm)	L-RFs $\times 10^{-8}$ (m^{-1})[b]	Remark
Building Y (difficult-to-return zone)	February 2 to 3, 2021[c]	2 (2)[d]	0.35–0.36 (0.091–0.098)[e]	2.2×10^{-5} to 2.3×10^{-5}[f]	3.1×10^{-5} to 3.2×10^{-5}	1.6	Before demolition
	March 3 to 24, 2021[g]	13 (9)	<0.13–2.3 (0.089–0.15)	$<8.2\times10^{-6}$ to 1.5×10^{-4}	$<1.1\times10^{-5}$ to 2.0×10^{-4}	0.58–10	During demolition
Tomioka town office (evacuation order-lifted area)	April 9 to 12, 2021[h]	2 (0)	N.D. (0.081–0.082)	-	-	-	Control building[i]

Abbreviations: L-RF, limited resuspension factor; N.D., not detected.
[a]Calculated using Equation (1). Daily ventilation rates were quoted from 13.5 m^3/8 h/d for the decontamination and demolition workers.
[b]Calculated using Equation (2); $^{137}Cs_{cont}$ (surface soil sample) was 21.9×10^3 Bq/m^2 before and during demolition.
[c]Airborne dust was collected two times (February 2–3, 2021).
[d]Sample number of ^{137}Cs detection.
[e]Detection limit at the time of ^{137}Cs detection.
[f]Minimum–maximum (values less than those indicated were not detected).
[g]Airborne dust was collected 13 times (March 3–24, 2021).
[h]Airborne dust was collected two times (April 9–12, 2021).
[i]Tomioka Town Food Inspection Department within the Tomioka town office.

and <0.13–2.3 mBq/m^3 during demolition from March 3 to May 24, 2021. On the other hand, ^{137}Cs was not detected at the Tomioka town office (the control building) from April 9 to 12, 2021. In addition, the internal exposure dose rates resulting from the inhalation of airborne dust, including ^{137}Cs, during decontamination and demolition, were estimated to range from 2.2×10^{-5} to 2.3×10^{-5} µSv/d due to inhalation of particulates with activity median aerodynamic diameters of 1 µm and/or 3.1×10^{-5} to 3.2×10^{-5} µSv/d due to inhalation of particulates with an activity median aerodynamic diameter of 5 µm before demolition from February 2 to 3, 2021. Moreover, the internal exposure dose rates were estimated at $<8.2\times10^{-6}$ -1.5×10^{-4} µSv/d due to inhalation of particulates with median aerodynamic diameters of 1 µm and/or $<1.1\times10^{-5}$–2.0×10^{-4} µSv/d due to inhalation of particulates with median aerodynamic diameters of 5 µm during demolition from March 3 to May 24, 2021. According to MOE survey data, the ambient dose rates that were measured at 1 m above ground level using a survey meter at 30 points around "Building Y" were 0.33–2.1 µSv/h before demolition (June 20, August 31, September 15, and October 16, 2017), 0.17–0.65 µSv/h after decontamination of the surface soil around "Building Y" (December 15, 2017), and 0.12–0.52 µSv/h after demolition (January 21, 2021). Therefore, the rates of decrease due to decontamination and demolition were 13%–83% at each point. Moreover, the L-RFs were 1.6×10^{-8} m^{-1} before demolition from February 2, 2021 and ranged from 0.58×10^{-8} to 10×10^{-8} m^{-1} during demolition from March 3 to May 24, 2021.

DISCUSSION

The difficult-to-return zone in Tomioka town, where approximately 4800 people had lived before the FDNPS accident, remains quite large at 8.5 km^2 (Tomioka Town Office, 2017). Although the ambient dose rates ranged from 1.9 to 3.8 µSv/h before the decontamination and demolition work in 2017, the ambient dose rates (median) in the SRRB decreased dramatically, from 1.0 to 0.32 µSv/h, as a result of the decontamination work conducted in 2018–2019 (Cui et al., 2020; MOE, 2019; Yamaguchi et al., 2021). Although in the Yonomori district (SRRB) in Tomioka town, demolition work was conducted using standard methods, such as cleaning rooms and removing interior materials; demolishing interior materials, roof and window frames, walls, and frameworks; removing walls and roof materials; and removing concrete foundations using heavy machinery, the annual external effective dose (median) for the decontamination workers in the SRRB was estimated to be 0.66 mSv/y (less than the public dose limit, 1 mSv/y) from July 2018 to July 2019 (Cui et al., 2020) (Supporting Information: Appendices S1 and S2).

In the present study, ^{137}Cs, the only artificial radionuclide, was detected intermittently in filter dust samples collected at the SRRB in the Yonomori district. The ^{137}Cs radioactivity levels in the airborne dust at the "Building G" (<0.10–0.73 mBq/m^3) and "Building Y" (<0.13–2.3 mBq/m^3) sites in the difficult-to-return zone were still at high levels compared with those of the control samples (<0.034–0.15 mBq/m^3) obtained from the evacuation order-lifted area in Tomioka town (Tables 1 and 2). Moreover, the L-RFs around "Building G" (1.1×10^{-8} to 7.5×10^{-8} m^{-1} before demolition and 0.47×10^{-8} to 2.8×10^{-8} m^{-1} during demolition) and "Building Y" (1.6×10^{-8} m^{-1} before demolition and 0.58×10^{-8} to 10×10^{-8} m^{-1} during demolition) were still at high levels compared with those at the control location ($<0.25 \times 10^{-8}$ m^{-1}) (Tables 1 and 2). According to another report, the atmospheric ^{137}Cs radioactivity levels changed temporarily to ranges from 10^{-1}–10^{0} Bq/m^3 to 10^{-5}–10^{-1} Bq/m^3 inside and outside the difficult-to-return zone during the first seven years after the FDNPS accident (Abe et al., 2021). In particular, the atmospheric ^{137}Cs radioactivity levels at urban sites significantly decreased after the first five years following the FDNPS accident due to anthropogenic activities such as the decontamination processes conducted around the monitoring sites (Abe et al., 2021). Similar to the atmospheric ^{137}Cs concentrations, the L-RFs were also lower than those during the initial two years both inside and outside the difficult-to-return zone, which ranged from 6.7×10^{-11} to 2.8×10^{-8} m^{-1} and from 1.7×10^{-10} to 2.0×10^{-7} m^{-1}, respectively (Abe et al., 2021). Although the L-RFs varied by one order of magnitude depending on location and whether a sampling site was inside or outside the difficult-to-return zone, the large-scale remediation conducted may reflect the reduction rate in L-RFs as atmospheric aerosols that were derived from a wide area (Abe et al., 2021). Based on the land category classification, such as urban lands, agricultural, paddy fields, and catchments (with facilitated radiocesium wash-off or not) in addition to anthropogenic influences, the L-RFs did change. However, in the present study, the L-RFs during demolition around "Building G" and "Building Y" showed expected and similar tendencies. Furthermore, the L-RF values in the present study were similar to the annual mean values of the L-RFs (e.g., 0.32×10^{-8} to 3.3×10^{-8} m^{-1} observed at Chernobyl in 1986–1991 and 0.030×10^{-8} to 130×10^{-8} m^{-1} observed in 14 European cities during the 2–3-year period after the Chernobyl accident), although the environmental fate of radiocesium radioactivity varies between Fukushima and Chernobyl (Ochiai et al., 2016). In the present study, atmospheric circulation was confirmed to have occurred during the sampling period by examining the levels of ^{7}Be, which is a natural radionuclide and tracer present in the troposphere and stratosphere (data not shown) that was detected in filter dust samples. According to meteorological observations such as wind direction, wind speed, and precipitation obtained by the Japan Meteorological Agency, Japan (https://www.jma.go.jp/jma/en/Activities/observations.html), and backward trajectory analyses using the NOAA HYSPLIT Trajectory Model (https://www.ready.noaa.gov/HYSPLIT_traj.php), a positive relationship was not always confirmed between atmospheric advection and the ^{137}Cs radioactivity levels at the "Building G" and "Building Y" sites (Supporting Information Appendices S1 and S2). These findings suggested that the ^{137}Cs detected in the present case may be driven from the areas around "Building G" and "Building Y" in the difficult-to-return zone. In other words, these radioactivity levels may not be due to resuspended ^{137}Cs that was associated with the demolition work at "Building G" and "Building Y," but might be due to resuspended ^{137}Cs that was caused by decontamination work such as concrete removal at the "Building G" and "Building Y" sites. Despite the demolition work using heavy machinery, the ^{137}Cs radioactivity levels did not increase significantly (Tables 1 and 2; Supporting Information Appendices S1 and S2). Therefore, the L-RFs (m^{-1}) estimated in the present study may reflect the indirect effects of airborne dust, including ^{137}Cs that was resuspended by wind and/or heavy machinery, as opposed to the direct effect of the demolition work at the sampling sites. In particular, the frequent passage of many trucks through the area as part of the decontamination and demolition work in the SRRB might have caused ^{137}Cs to be resuspended (Cui et al., 2020). Although the present data over a small area are limited, these data are expected to be useful for accurately estimating additional radiation exposures due to radiocesium in the future. However, the dispersal distances of particles differ greatly depending on their particle sizes. Further investigations, such as compositional analysis of particles, including radiocesium, are needed to estimate their origins.

However, the internal exposure dose rates due to aspiration of airborne dust, including ^{137}Cs, were extremely low compared with the estimated annual effective dose of decontamination workers or the annual effective dose limits recommended by the Japanese government (Tables 1 and 2) (MHLW, 2018). Additionally, the external exposure doses due to ^{137}Cs in the surface soil were low because the decontamination process, which included removing surface soil and fallen leaves and stripping topsoil from gardens, was close to being fully complete in the SRRB during the recovery and reconstruction period after the FDNPS accident (Supporting Information Appendices S1 and S2) (Cui et al., 2020; Matsuo et al., 2019; Yamaguchi et al., 2021). In the difficult-to-return zone of Tomioka town, the mean ambient dose rates were 1.4 (12)–1.8 (16) µSv/h (mSv/y) outdoors in 2018 and 1.5 (13)–1.6 (14) µSv/h (mSv/y) outdoors in 2019 (Yamaguchi et al., 2021). Moreover, in the difficult-to-return zone, the mean radiocesium concentrations in surface soils (0–5 cm) within the assembly halls were 3012 (19–5720) Bq/kg-dry in 2018 and 1805 (20–5873) Bq/kg-dry in 2019 for ^{134}Cs and 31 603 (243–58 719) Bq/kg-dry in 2018 and 25 658 (223–83 001) Bq/kg-dry in 2019 for ^{137}Cs (Yamaguchi et al., 2021). The annual ambient dose equivalents from the surface soil were estimated to be 0.35 (3.1) µSv/h (mSv/y) in 2018 and 0.44 (3.9) µSv/h (mSv/y) in 2019 (Yamaguchi et al., 2021). Although the amounts of radiocesium in the decontamination and demolition workers at "Building G" and "Building Y," which were determined using a whole-body counter, were not assessed directly, their internal exposure doses were estimated to be extremely limited. The ambient dose rates did not exceed 2.5 µSv/h within "Building G" or "Building Y" before and after demolition

(Supporting Information Appendices S1 and S2). Nevertheless, careful attention should be paid to radiation safety for workers so that they can protect themselves appropriately to avoid unnecessary internal exposure through inhalation of particulates derived from the FDNPS accident, including ^{137}Cs. Therefore, wearing masks and controlling environmental contamination could further reduce the risk of internal exposure due to inhalation of airborne dust, including ^{137}Cs, and that of external exposure by managing the external doses.

There are several limitations to the present study. First, filter dust samples were not collected continuously at "Building G" and "Building Y" because the sample collections were carried out under restricted conditions (the SRRB in the difficult-to-return zone) that resulted from the FDNPS accident. In addition, environmental monitoring surveys were not performed directly by using dosimetry. Second, no airborne particulate matter (PM) samples were collected by a multistage impactor, which is used to separate particles with aerodynamic diameters less than 2.5 µm ($PM_{2.5}$) and greater than $PM_{2.5}$. Third, the L-RF values for ^{137}Cs assume homogeneous contamination in the atmosphere (i.e., a simple ratio of airborne dust radioactivity to surface contamination). Furthermore, local atmospheric wind-derived resuspension of contaminated soil particles depends on accident-derived radionuclide deposition in areas around the FDNPS (Igarashi et al., 2011). According to our follow-up investigation after demolition, ^{137}Cs was even intermittently detected from two types of filter dust samples ($PM_{2.5}$ [<2.5 µm] and PM_{10} [>2.5 µm]) that were collected at the main road near "Building G" (<0.12–0.15 mBq/m^3) and "Building Y" (<0.25 mBq/m^3). The L-RF values were 0.50–0.64 × 10^{-8} m^{-1} near "Building G" and 1.2 × 10^{-8} m^{-1} near "Building Y." These values were similar to the values presented in the present study. Although the current environmental radioactivity levels and/or the external effective doses due to ^{137}Cs may be gradually decreasing compared to the levels detected immediately after demolition in the SRRB, these results show that radiocesium nuclides continue to be a potential long-term source of contaminated particulate matter. In other words, the ^{137}Cs radioactivity in the SRRB airborne dust may be primarily associated with particles that are resuspended by compound factors such as weather conditions (localized winds) and decontamination and demolition work (transfer of construction vehicles). Further investigations with detailed conditions, including bioaerosols and/or dosimetric modeling for workers, are therefore needed to confirm the effects of decontamination and demolition on ^{137}Cs resuspension.

CONCLUSION

For the case of "Building G" and "Building Y," the radioactivity levels in airborne dust due to accident-derived ^{137}Cs did not increase dramatically as a result of the decontamination and demolition of buildings in the SRRB (the difficult-to-return zone) of Tomioka town, Fukushima Prefecture. These findings suggest that the ^{137}Cs radioactivity levels in airborne dust may be primarily associated with particles that are resuspended by localized winds accompanied by transfers from construction vehicles such as large trucks rather than being caused by the decontamination and demolition activities at the sampling sites. However, the internal exposure doses due to aspiration of airborne dust, including ^{137}Cs, were low compared with the estimated annual effective doses of the decontamination workers or the annual effective dose limits recommended by the Japanese government. Additionally, suitable countermeasures, such as wearing protective masks, could help decrease the on-site inhalation of soil-derived radionuclides. This case of Tomioka town within the SRRB may be the first working environment model for evaluating the environmental contamination and internal exposure dose rates due to artificial radionuclides derived from a nuclear disaster.

AUTHOR CONTRIBUTION

Yasuyuki Taira, Noboru Takamura, and Shigekazu Hirao contributed to the study design; Yasuyuki Taira, Makiko Orita, Hitomi Matsunaga, and Shigekazu Hirao collected the data; Yasuyuki Taira, Masahiko Matsuo, and Shigekazu Hirao analyzed the data; and Yasuyuki Taira and Shigekazu Hirao led the writing. All authors assisted in the study design, contributed to the data collection, assisted in the interpretation of data, and made critical revisions of the manuscript. All authors approved the final version of the manuscript.

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CONFLICT OF INTEREST

The authors declare no conflicts of interest.

DATA AVAILABILITY STATEMENT

All data are presented in the article and the Supporting Information files.

SUPPORTING INFORMATION

Raw data. Sample data such as airborne dusts and soils collected in the present study.

Appendix S1. Detailed information about the demolition progress and ^{137}Cs radioactivity at the site of "Building G," located within the Yonomori district in the difficult-to-return zone (SRRB) of Tomioka town, Fukushima Prefecture. Localized resuspended ^{137}Cs radioactivity and photos are fit into the calendar from May 27 to August 18, 2020.

Appendix S2. Detailed information about the demolition progress and ^{137}Cs radioactivity at the site of "Building Y," located within the Yonomori district in the difficult-to-return zone (SRRB) of Tomioka town, Fukushima Prefecture. Localized-resuspended ^{137}Cs radioactivity and photos are fit into the calendar from March 3 to 30, 2021.

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