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Activity concentrations of environmental samples collected in Fukushima Prefecture immediately after the Fukushima nuclear accident

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Radionuclide concentrations in environmental samples such as surface soils, plants and water were evaluated by high purity germanium detector measurements. The contribution rate of short half-life radionuclides such as ^{132}I to the exposure dose to residents was discussed from the measured values. The highest values of the ^{131}I/^{137}Cs activity ratio ranged from 49 to 70 in the environmental samples collected at Iwaki City which is located to the south of the F1-NPS. On the other hand, the ^{132}I/^{131}I activity ratio in the same environmental samples had the lowest values, ranging from 0.01 to 0.02. By assuming that the ^{132}I/^{131}I activity ratio in the atmosphere was equal to the ratio in the environmental samples, the percent contribution to the thyroid equivalent dose by ^{132}I was estimated to be less than 2%. Moreover, the contribution to the thyroid exposure by ^{132}I might be negligible if ^{132}I contamination was restricted to Iwaki City.

On March 11, 2011, the power supplies for the cooling systems in the Fukushima Dai-ichi Nuclear Power Station (F1-NPS) were lost due to tsunami damage following the magnitude 9.0 Great East Japan earthquake[1,2]. Loss of cooling functions led to hydrogen explosions in three reactor units in the F1-NPS and artificial radionuclides such as radioiodine and radiocesium were released from the reactor buildings. These radionuclides have been detected around the world[3–7]. The most contaminated area in Fukushima Prefecture has been observed to the northwest from the F1-NPS[8].

On March 12, the Japanese Government had ordered the evacuation of residents within a 20-km radius area from the F1-NPS. A screening survey of radionuclide contamination of evacuees was carried out based on the evacuation decision. A first team of radiological professionals from Hirosaki University carried out the screening survey from March 15 to 19, 2011 at evacuation shelters and public facilities in Fukushima Prefecture[9]. The team had two purposes: to carry out the screening survey for evacuees and to evaluate ambient dose rate and activity concentrations. In such a nuclear power station accident with large releases of radioactive contaminates from the reactor containment, special attention must be paid to internal exposure to the thyroid by inhalation of released ^{131}I and ^{132}I. ^{131}I and ^{132}I have half-lives of 8 days and 2.3 hours, respectively, and therefore it is necessary to make air-borne activity measurements of these radionuclides quickly. Local health authorities measured the dose rate in the thyroid of 1,149 children under the age of 15 by 1-inch × 1-inch NaI(Tl) scintillation survey meter from March 24 to 30, 2011[10]. Tokonami *et al.*[11] measured the thyroid doses for 62 evacuees (including infants) using a 3-inch × 3-inch NaI(Tl) scintillation spectrometer and estimated the thyroid equivalent doses of all of them were below 50 mSv. However, contributions to the thyroid equivalent dose of ^{132}I and ^{132}Te as short half-life

Table 1 | Summary of the sampling site locations, date and type of samples

Sampling site	Site ID	Distance from NPS (Direction)	Longitude (degree)	Latitude (degree)	Sampling date	Ambient dose rate (μGy h^{-1})	Sample			
							Soil	Water	Plant	Filter
Koriyama Fukushima Pref.	KO-1	58.5 km (W)	37.371	140.375	2011/3/17	5.13	0–5 cm	Tap water	*Rhododendron* × *pulchrum*	1168 L (9.7 hours)
Koriyama Fukushima Pref.	KO-2	58.3 km (W)	37.394	140.375	2011/3/17	9.70	0–5 cm	Ground water	*Chamaecyparis obtusa*	NC***
Fukushima Fukushima Pref.	FU	62.9 km (NW)	37.763	140.468	2011/3/17	7.39 (0.252*)	0–5 cm	Rain water (RA) River water (RI)	*Chamaecyparis pisifera*	1320 l* (11 hours)
Iwaki Fukushima Pref.	IW-1	44.4 km (SSW)	37.045	140.865	2011/3/18	1.09	0–5 cm	Tap water	*Camellia sasanqua*	NC***
Iwaki Fukushima Pref.	IW-2	63.7 km (SSW)	36.889	140.768	2011/3/18	0.414	0–5 cm	Tap water	*Chamaecyparis pisifera*	404 L (3.4 hours)
Aizuwakamatsu Fukushima Pref.	AI	96.4 km (W)	37.485	139.945	2011/3/18	0.494	0–5 cm	Sewer (SE) Snow water (SN)	*Sasa kurilensis* (SK) *Aucuba japonica* (AJ)	NC***
Kawamata Fukushima Pref.	KA	46.0 km (NW)	37.669	140.615	2011/3/19	4.50	0–5 cm	Tap water	*Camellia japonica*	266 L (2.2 hours)
Shirakawa Fukushima Pref.	SH	78.6 km (SW)	37.122	140.229	2011/3/19	2.12	0–5 cm	Tap water	*Cedrus deodara*	NC***
Marumori Miyagi Pref.	MA	52.1 km (NNW)	37.817	140.717	2011/3/19	NM**	0–5 cm	Tap water	*Chamaecyparis pisifera*	NC***
Kunimi Fukushima Pref.	KU	67.2 km (NNW)	37.907	140.577	2011/3/19	2.93	0–5 cm	Tap water	NC***	NC***
Zao Miyagi Pref.	ZA	79.5 km (NNW)	38.070	140.660	2011/3/19	0.450	0–5 cm	NC***	NC***	NC***

*Air-borne radionuclides were collected on a fourth floor building balcony. Ambient dose rate was measured at the filter sampling point.
**NM: Not measured
***NC: Not collected

radionuclides were not considered. Balanov *et al.*[12] determined the average percent contribution of short half-life radionuclides to thyroid dose of residents in Chernobyl was about 30%.

The authors collected environmental samples such as surface soils, leaves and water immediately after the accident. In this study, radionuclide concentrations in these environmental samples were evaluated, and the percent contribution of short half-life radionuclides to the exposure dose to residents was discussed. On the other hand, airborne radionuclide concentrations provide important information on the estimation of internal dose due to inhalation in nuclear disasters. In such an emergency, a simple technique to measure airborne radionuclide concentrations without an AC power supply is needed. Immediately after the accident, radioactive aerosol sampling was also carried out using a glass fiber filter and a battery-powered pump at several locations in Fukushima Prefecture. Furthermore, the inhalation exposure from radioactive aerosols for residents in a measurement period was also discussed.

Results

Ambient dose rate at each measurement site. Ambient dose rate at each measurement site is shown in **Table 1**. Ambient dose rates ranged from 0.414 to 9.70 μGy h^{-1}. The highest ambient dose rate of 9.70 μGy h^{-1} was observed at Koriyama City (KO-2), located to the west of F1-NPS, on March 17, 2011. Ambient dose rate at Iwaki City (IW-2), located to the south of F1-NPS, had the lowest value of 0.414 μGy h^{-1} on March 18, 2011. As previously reported[8], ambient dose rates in the northwest and west directions such as Kawamata Town (KA), Fukushima City (FU) and Koriyama City (KO) were observed to have higher values ranging from 4.50 to 9.70 μGy h^{-1}. However, the ambient dose rate measured for a fourth floor balcony in Fukushima City had the lowest value of 0.252 μGy h^{-1}.

Air-borne radionuclide concentrations at four sampling sites. ^{131}I was detected at three sites (KO-1, FU and IW-2) as shown in **Table 2**. Air-borne ^{131}I aerosol concentration at Iwaki City was the highest, 10 ± 3 mBq m^{-3}. On the other hand, air-borne concentrations of ^{134}Cs and ^{137}Cs at the fourth site, Kawamata Town, were the highest with values of 89 ± 23 and 66 ± 18 mBq m^{-3}, respectively. The highest ambient dose rates of ^{131}I, ^{134}Cs and ^{137}Cs at Koriyama City were 2 ± 1, 6 ± 4 mBq m^{-3} and under the detection limit (ND), respectively.

Radionuclide concentrations of environmental samples at each sampling site. ^{132}Te, ^{131}I, ^{134}Cs and ^{137}Cs were detected in soil samples which were collected at all sampling sites as shown in **Table 3**. The maximum values of these radionuclides in the soil samples were observed at Fukushima City (FU), and their respective values were 2.2 × 10^5, 1.5 × 10^5, 2.8 × 10^4 and 2.9 × 10^4 Bq kg^{-1} wet. ^{129m}Te, ^{136}Cs and ^{132}I were detected in soil samples which were collected at most of the sampling sites in Fukushima Prefecture. Maximum values of these radionuclides (and site ID) were 4.1 × 10^4 (FU), 9.2 × 10^3 (KO-2) and 3.3 × 10^4 Bq kg^{-1} wet (FU). Furthermore, ^{140}La was also detected at several sampling sites and the maximum value (and site ID) was 1.8 × 10^4 Bq kg^{-1} wet

Table 2 | Radionuclide concentrations for samples collected on a glass fiber filter at four sampling sites

Site ID	Radionuclide concentrations (mBq m^{-3})*		
	^{131}I (364 keV)	^{134}Cs (795 keV)	^{137}Cs (662 keV)
KO-1	2.3 ± 0.8	5.6 ± 4.3	ND**
FU	2.9 ± 0.9	8.7 ± 3.8	7.7 ± 3.0
IW-2	10.0 ± 2.6	20.1 ± 15.2	15.2 ± 11.4
KA	ND**	88.8 ± 22.8	66.3 ± 18.0

*Radionuclide concentrations were corrected to the sampling date.
**ND: Under the detection limit.

Table 3 | Radionuclide concentrations for soil samples

Site ID	Radionuclide concentrations (kBq kg^{-1} wet)							
	^{132}Te	^{131}I	^{134}Cs	^{137}Cs	^{129m}Te	^{136}Cs	^{132}I	^{140}La
KO-1	31.59 ± 0.23	28.32 ± 0.16	6.02 ± 0.05	6.09 ± 0.07	6.37 ± 0.80	1.08 ± 0.03	5.35 ± 0.05	5.01 ± 0.46
KO-2	46.14 ± 0.27	42.81 ± 0.20	5.32 ± 0.05	5.54 ± 0.07	7.11 ± 0.79	9.20 ± 0.03	6.84 ± 0.05	4.67 ± 0.53
FU	223.90 ± 0.62	150.10 ± 0.38	28.35 ± 0.11	29.43 ± 0.17	40.52 ± 2.12	4.85 ± 0.07	33.25 ± 0.15	18.54 ± 1.10
KA	75.32 ± 0.34	93.90 ± 0.28	9.05 ± 0.06	9.64 ± 0.09	14.79 ± 1.04	1.55 ± 0.03	10.78 ± 0.07	6.42 ± 0.55
IW-1	2.06 ± 0.06	16.01 ± 0.11	0.26 ± 0.01	0.30 ± 0.02	ND*	0.05 ± 0.01	0.29 ± 0.01	ND*
IW-2	2.52 ± 0.07	15.72 ± 0.12	0.35 ± 0.01	0.32 ± 0.02	0.99 ± 0.25	ND*	0.33 ± 0.01	ND*
AI	14.97 ± 0.15	6.91 ± 0.08	2.30 ± 0.03	2.40 ± 0.04	2.00 ± 0.39	0.41 ± 0.02	2.18 ± 0.03	ND*
SH	15.11 ± 0.17	11.42 ± 0.11	2.07 ± 0.03	2.22 ± 0.05	2.66 ± 0.52	0.38 ± 0.02	2.10 ± 0.03	1.89 ± 0.35
MA	4.64 ± 0.09	10.59 ± 0.10	0.50 ± 0.02	0.49 ± 0.02	ND*	0.07 ± 0.01	ND*	ND*
KU	59.73 ± 0.31	51.70 ± 0.21	6.33 ± 0.05	6.55 ± 0.07	7.80 ± 0.80	1.06 ± 0.03	8.59 ± 0.06	ND*
ZA	11.75 ± 0.14	19.31 ± 0.13	1.37 ± 0.03	1.42 ± 0.04	ND*	0.26 ± 0.02	1.65 ± 0.02	ND*

*ND: Under the detection limit.

(FU). ^{132}Te,^{131}I,^{134}Cs, ^{136}Cs and ^{137}Cs were detected in the plant samples which were collected at all sampling sites as shown in **Table 4**. Maximum values of these radionuclides (and site ID) were 4.1×10^5 (KO-1), 3.7×10^5 (IW-1), 1.5×10^5 (KO-1), 2.7×10^4 (KO-1) and 1.6×10^5 Bq kg^{-1} wet (KO-1). ^{131}I activity concentration in Iwaki City (IW) had the highest value. ^{129m}Te and ^{132}I were detected in the plant samples which were collected at most sampling sites in Fukushima Prefecture. Maximum values of these radionuclides (and site ID) were 6.6×10^4 (KO-1) and 5.8×10^4 Bq kg^{-1} wet (KO-2). Furthermore, ^{140}La was also detected at several sampling sites and the maximum value (and site ID) was 4.8×10^4 Bq kg^{-1} wet (KO-2). ^{132}Te, ^{131}I, ^{134}Cs, ^{137}Cs, ^{136}Cs and ^{132}I were detected from some water samples as summarized in **Table 5**. Maximum values of these radionuclides (and site ID) were 1.8×10^3 (AI-SN), 1.3×10^5 (FU-RI), 6.8×10^2 (AI-SN), 8.5×10^2 (AI-SN), 1.4×10^2 (AI-SN) and 2.4×10^2 Bq L^{-1} (AI-SN). A maximum value was observed in a snow sample collected at Aizuwakamatsu City, which is located approximately 96 km from the F1-NPS. On the other hand, the activity concentrations of ^{129m}Te and ^{140}La were below detection limits.

Discussion

Radon decay products were collected more than 99% on the 1st stage as the result of performance test of the filter sampling system in the radon chamber of the National Institute of Radiological Sciences, Japan (NIRS). Moreover, radionuclide distributions on the glass fiber filter obtained by the imaging plate measurements seemed to be homogeneous. Therefore, the simple filter sampling system used for this study was an effective technique for the collection of airborne radionuclide in an emergency situation. The airborne ^{131}I activity concentration at Iwaki City was observed as the highest value of 10 mBq m^{-3} on March 18, 2011. According to the estimation of thyroid equivalent dose for an infant by SPEEDI (System for Prediction of Environmental Emergency Dose Information), high equivalent doses were shown not only in the northwest direction from F1-NPS but also in the south direction such as along the coast in Iwaki City[13]. According to the simulation results by Katata *et al.*[14], a radioactive plume including ^{131}I was released in the south direction from F1-NPS in the morning on March 15, and it reached Iwaki City. No rainfall was observed around Iwaki City (Yamada monitoring station) on March 15 according to meteorological observation data of the Japan Meteorological Agency[15]. Rainfall of 0.5–2.0 mm was observed at Iwaki City from 2 PM to 4 PM on March 16, and no rainfall was observed until 7 AM on March 21[15]. This fact suggested that the contamination in Iwaki City was dry deposition. The maximum values of thyroid equivalent dose for residents in Namie Town were estimated to be 33 mSv according to Tokonami *et al.*[11]. Moreover, they estimated the atmospheric ^{131}I activity concentration on March 15 was 23 kBq m^{-3}. This estimated value was the ^{131}I activity concentration of particulate and gaseous forms. According to Momoshima *et al.*[16], the ^{131}I collected on activated charcoal accounted for 30 to 67% of the total ^{131}I. ^{131}I activity concentration was corrected to the value of March 15, 2011 for the physical half-life, and it was evaluated as 13 mBq m^{-3}. ^{131}I activity concentration as gaseous forms was estimated to be 30 mBq m^{-3}, assuming that ^{131}I gaseous forms were 70% of the total amount. Moreover, according to the simulation results by Morino *et al.*[17], all the species in the radioactive plume from F1-NPS were released toward the Pacific Ocean during the period from March 17 to 19. This fact might indicate the internal exposure by inhalation of ^{131}I at Iwaki City during the period from March 17 to March 19, 2011 was negligible.

Radionuclide concentrations in the environmental samples collected at Fukushima City and Koriyama City were higher than those

Table 4 | Radionuclide concentrations for plant samples

Site ID	Radionuclide concentrations (kBq kg^{-1} wet)							
	^{132}Te	^{131}I	^{134}Cs	^{137}Cs	^{129m}Te	^{136}Cs	^{132}I	^{140}La
KO-1	414.50 ± 1.49	167.60 ± 0.77	149.70 ± 0.47	155.20 ± 0.68	66.04 ± 5.27	27.48 ± 0.23	ND*	ND*
KO-2	372.80 ± 1.21	304.00 ± 0.83	62.86 ± 0.27	63.17 ± 0.38	64.33 ± 3.97	11.71 ± 0.14	58.20 ± 0.26	47.77 ± 2.39
FU	53.46 ± 0.46	125.30 ± 0.51	5.08 ± 0.08	4.87 ± 0.11	8.54 ± 1.18	0.95 ± 0.04	8.26 ± 0.09	2.96 ± 0.90
KA	120.30 ± 0.73	126.80 ± 0.57	13.10 ± 0.13	13.72 ± 0.19	21.38 ± 2.28	2.33 ± 0.07	17.92 ± 0.14	ND*
IW-1	28.69 ± 0.43	365.40 ± 0.97	5.09 ± 0.08	5.19 ± 0.12	4.60 ± 1.45	1.03 ± 0.05	4.39 ± 0.08	ND*
IW-2	22.69 ± 0.35	247.10 ± 0.75	5.62 ± 0.08	5.71 ± 0.12	ND*	1.14 ± 0.05	ND*	ND*
AI-SK	49.16 ± 0.70	15.31 ± 0.32	15.26 ± 0.21	16.36 ± 0.31	9.07 ± 2.86	2.76 ± 0.10	ND*	ND*
AI-AJ	17.91 ± 0.29	4.37 ± 0.13	4.79 ± 0.08	5.15 ± 0.12	ND*	0.84 ± 0.04	2.59 ± 0.06	ND*
SH	241.60 ± 1.03	142.10 ± 0.61	49.26 ± 0.25	49.88 ± 0.31	40.28 ± 3.12	8.76 ± 0.13	36.16 ± 0.22	46.04 ± 5.17
MA	105.50 ± 0.77	97.28 ± 0.57	17.35 ± 0.17	17.90 ± 0.25	24.21 ± 3.18	3.07 ± 0.09	15.81 ± 0.15	ND*

*ND: Under the detection limit.

Table 5 | Radionuclide concentrations for water samples

Site ID	Radionuclide concentrations (kBq L^{-1})							
	^{132}Te	^{131}I	^{134}Cs	^{137}Cs	^{129m}Te	^{136}Cs	^{132}I	^{140}La
KO-1	1.31 ± 0.05	2.91 ± 0.05	0.09 ± 0.01	0.11 ± 0.01	ND*	ND*	ND*	ND*
KO-2	ND*	ND*	ND*	ND*	ND*	ND*	ND*	ND*
FU-RA	0.27 ± 0.02	1.22 ± 0.03	0.04 ± 0.01	0.04 ± 0.01	ND*	ND*	ND*	ND*
FU-RI	1.45 ± 0.09	128.00 ± 0.33	0.06 ± 0.01	0.07 ± 0.01	ND*	ND*	0.23 ± 0.01	ND*
KA	ND*	0.14 ± 0.01	ND*	ND*	ND*	ND*	ND*	ND*
IW-1	ND*	0.03 ± 0.01	ND*	ND*	ND*	ND*	ND*	ND*
IW-2	ND*	ND*	ND*	ND*	ND*	ND*	ND*	ND*
AI-SE	ND*	0.01 ± 0.00	ND*	ND*	ND*	ND*	ND*	ND*
AI-SN	1.82 ± 0.05	0.27 ± 0.02	0.68 ± 0.02	0.85 ± 0.03	ND*	0.14 ± 0.01	0.24 ± 0.01	ND*
SH	ND*	ND*	ND*	ND*	ND*	ND*	ND*	ND*
MA	0.04 ± 0.01	0.12 ± 0.01	ND*	ND*	ND*	ND*	ND*	ND*
KU	ND*	0.08 ± 0.01	ND*	ND*	ND*	ND*	ND*	ND*

*ND: Under the detection limit.

for samples collected in other sites (**Table 2**). Radionuclide concentrations of soil samples collected at Fukushima City on March 22 were reported by Taira *et al.*[18]. Although ^{132}Te, ^{132}I and ^{140}La were not detected, activity concentrations of other radionuclides in that report were similar values to the present results. According to the simulation results by Katata *et al.*[14], the radioactive plume including ^{131}I was released to the northwest direction from F1-NPS in the evening on March 15, and it reached Fukushima City and Koriyama City. Katata *et al.* also reported that the radioactive contamination by wet deposition (rainfall) was observed around these areas in the evening on March 15. Activity ratio of each radionuclide based on ^{137}Cs activity concentration is shown in **Table 6** and **Table 7**. The obtained radionuclide concentrations were corrected to the value of March 15, 2011 for each physical half-life. Since the number of detected radionuclides in water samples was small, only the activity ratio results for soil and plant samples are shown in this table. Tagami *et al.*[19] reported the average value of the ^{134}Cs/^{137}Cs activity ratio of soil samples which were collected 20 km south of F1-NPS was 0.9. Moreover, ^{134}Cs/^{137}Cs activity ratios of tea leaves (collected 300 km southwest from F1-NPS) and camellia leaves (collected 220 km south from F1-NPS) were also reported to be 0.98 ± 0.09 and 0.92 ± 0.05[20]. In this study, the average values (range) of the ^{134}Cs/^{137}Cs activity ratio of all soil and plant samples were 1.0 (0.89–1.1) and 1.0 (0.93–1.0), respectively. These values were similar to the previous study[19,20]. range) of ^{131}I/^{137}Cs activity ratio of all soil and plant samples were 16.4 (2.9–54) and 16.5 (0.85–70), respectively. Average values (range) of ^{131}I/^{137}Cs activity ratio of soil and plant samples at Iwaki City were 51 (49 and 54) and 57 (43 and 70), respectively. On the other hand, average values of ^{131}I/^{137}Cs activity ratio of soil and plant samples excluding Iwaki City were 8.7 and 6.4, respectively. These results suggested that the generation sources of radioactive plume which was released on March 15, 2011 to each area differed.

Average values (range) of ^{132}I/^{131}I activity ratio of all soil and plant samples were 0.09 (0.02–0.32) and 0.19 (0.01–0.59), respectively (**Table 6** and **Table 7**). Especially, ^{132}I/^{131}I activity ratio of soil and plant samples at Iwaki City had the lowest values, and they were 0.02 and 0.01, respectively. The percent contribution to the thyroid equivalent dose of ^{132}I was not considered in the report by Tokonami *et al*[11]. According to ICRP Publication 72, dose coefficients of ^{131}I and ^{132}I to an adult are 2.2×10^{-8} and 2.9×10^{-10} (Sv/Bq), respectively[21]. If it was assumed that the ^{132}I/^{131}I activity ratio in the atmosphere was equal to the ratio in the environmental samples, the percent contribution to the thyroid equivalent dose by ^{132}I was estimated to be less than 2%. Moreover, the contribution by ^{132}I to the thyroid exposure might be negligible (less than 0.03%) if ^{132}I was restricted to Iwaki City. However, if ^{132}Te is taken into the body, ^{132}I will be generated by radioactive decay of ^{132}Te, and the generated ^{132}I will accumulate in the thyroid[12]. Thus, it will be necessary to examine this process in the human body.

The authors have already reported on the thyroid equivalent dose for residents who lived in the northwest direction from F1-NPS[11]. Although the local health authorities were reported on the screening survey of the thyroid dose in Iwaki City which was contaminated by ^{131}I at the same level as the northwest region, no detailed examination in this area was carried out by the Japanese government. Furthermore, since the residents were not evacuated from Iwaki City, many children who lived in this city might have been exposed to radioiodine. Therefore, it is important to clarify the thyroid equivalent dose for children who lived in a south direction from F1-NPS (especially coastal areas) immediately after the accident, and it is also important to continue to make ultrasound examinations of the thyroid for residents.

Table 6 | Activity ratio in soil samples of seven radionuclides to ^{137}Cs and the ^{132}I/^{131}I activity ratio

Site ID	^{132}Te/^{137}Cs	^{131}I/^{137}Cs	^{134}Cs/^{137}Cs	^{129m} Te/^{137}Cs	^{136}Cs/^{137}Cs	^{132}I/^{137}Cs	^{140}La/^{137}Cs	^{132}I/^{131}I
KO-1	5.2	4.7	1.0	1.0	0.18	0.88	0.82	0.19
KO-2	8.3	7.7	1.0	1.3	1.7	1.2	0.84	0.16
FU	7.6	5.1	1.0	1.4	0.16	1.1	0.63	0.22
KA	7.8	9.7	0.94	1.5	0.16	1.1	0.67	0.11
IW-1	7.0	54	0.89	–	0.16	0.97	–	0.018
IW-2	7.8	49	1.1	3.1	–	1.0	–	0.021
AI	6.2	2.9	1.0	0.83	0.17	0.91	–	0.32
SH	6.8	5.2	0.93	1.2	0.17	0.95	0.85	0.18
MA	9.4	21	1.0	–	0.14	–	–	–
KU	9.1	7.9	1.0	1.2	0.16	1.3	–	0.17
ZA	8.3	14	1.0	–	0.18	1.2	–	0.085

Table 7 | Activity ratio in plant samples of seven radionuclides to ^{137}Cs and the ^{132}I/^{131}I activity ratio

Site ID	^{132}Te/^{137}Cs	^{131}I/^{137}Cs	^{134}Cs/^{137}Cs	^{129m} Te/^{137}Cs	^{136}Cs/^{137}Cs	^{132}I/^{137}Cs	^{140}La/^{137}Cs	^{132}I/^{131}I
KO-1	2.7	1.1	1.0	0.43	0.18	–	–	–
KO-2	5.9	4.8	1.0	1.0	0.19	0.92	0.76	0.19
FU	11	26	1.0	1.8	0.19	1.7	0.61	0.066
KA	8.8	9.2	0.95	1.6	0.17	1.3	–	0.14
IW-1	5.5	70	0.98	0.89	0.20	0.85	–	0.012
IW-2	4.0	43	0.98	–	0.20	–	–	–
AI-SK	3.0	0.94	0.93	0.55	0.17	–	–	–
AI-AJ	3.5	0.85	0.93	–	0.16	0.50	–	0.59
SH	4.8	2.8	1.0	0.81	0.18	0.72	0.92	0.25
MA	5.9	5.4	1.0	1.4	0.17	0.88	–	0.16

Methods

Environmental sampling. Environmental sampling sites in Fukushima Prefecture are shown in **Fig. 1**. This figure was made using the Generic Mapping Tools (GMT) created in 1988 by Wessel and Smith[22]. The types of environmental samples were summarized in **Table 1**. The sampling sites were selected after considering direction and distance from the F1-NPS. Moreover, the environmental sampling sites were located at evacuation shelters and public facilities. The distance between F1-NPS and each sampling site was about 44–96 km. A 1 kg soil sample from 5 cm below the surface was collected at each sampling site. Moreover, a plant sample was also collected at each soil sampling site. Plant species are summarized in **Table 1**. Rain water, river water and snow were collected at some sampling sites. Ambient radioactive aerosols were collected by a two-stage sampling technique with glass fiber filters (Whatman GF/F, $\phi = 47$ mm) and a battery-powered pump (MP-Σ300, Sibata Scientific Technology Ltd.). The sampling flow rate was set to 2.0 L min^{-1} at each sampling site. The sampling time and total sampling volume are given in **Table 1**. The weather at each site was fair during the period from March 17 to 19.

Measurement of ambient dose rate. *In-situ* gamma-ray spectra at several sampling sites for the estimation of ambient dose rate were obtained using a 3-inch × 3-inch NaI(Tl) scintillation spectrometer (JSM-112, Hitachi-Aloka Co.). Measurements at every site were carried out 1 m above the uncovered soil surface. Measurements at Fukushima City (FU) were carried out not only outside but also on a balcony of a fourth floor building (the disaster countermeasures office). This balcony was made of concrete. Counting time was set to 300 s at every site. The obtained gamma-ray pulse height distributions were unfolded by a 60 × 60 response matrix for the evaluation of ambient dose rates[23]. This calculation software assumed that the fallout formed an infinite plane source on the ground.

Evaluation of radionuclide concentrations. Quick measurement is necessary for evaluation of the short half-life nuclide concentrations. Although gravel and roots were removed from the soil samples, a drying processing was not carried out for the environmental samples. The plant sample was cut about 1 square centimeter from a leaf. The water sample (100 mL) had several grams of NaCl added as a carrier for the evaluation of radionuclide concentration. Two filter samples were enclosed in a container. Every environmental sample was enclosed in a cylindrical polypropylene container of 48 mm × 55 mm. Radionuclide concentrations of each sample were measured with a high-purity germanium (HPGe) detector (GEM-100210, ORTEC). The measurement time was set at 600 s for the evaluations of short half-life radionuclides such as ^{131}I, ^{132}I and ^{132}Te. For evaluation of long half-life radionuclides such as ^{134}Cs and ^{137}Cs, measurement time was set at more than 16,000 s. The radionuclide concentrations in environmental samples excluding filter samples were corrected to the value on March 15, 2011 by each physical half-life. On the other hand, radionuclide concentrations of filter samples were corrected to the sampling date.

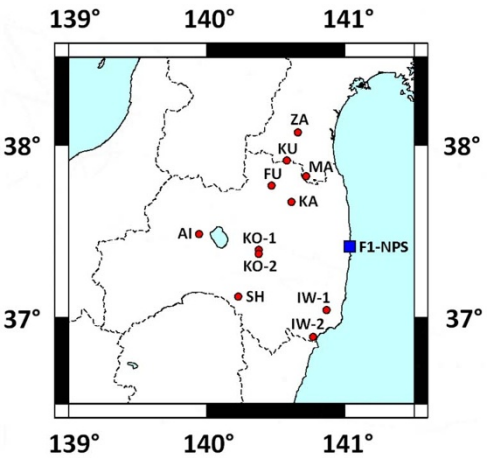

Figure 1 | The location of the environmental sampling points. Sampling sites in Fukushima Prefecture were selected after considering their direction and distance from F1-NPS.

Evaluation of surface distribution of radioactive aerosols on the filter. The surface distribution of radioactive aerosols on the filter is also important due to the counting efficiency in the HPGe detector measurement. Therefore the surface distribution with the same system as for *in-situ* sampling was evaluated using a radioactive aerosol chamber (internal volume: approximately 25-m^3) at NIRS. This radioactive aerosol chamber is environmentally controlled for temperature and relative humidity. Radon is used as the radioactive source. The temperature and relative humidity can be controlled in the range of 5 to 30°C with an error of 0.5°C, and 30 to 90% with an error of 3%, respectively[24]. In this study, radon concentration, temperature and relative humidity were set to 10,000 Bq m^{-3}, 20°C and 60%, respectively. Carnauba wax was used as the aerosol material and the particle size had the distribution which made approximately 100 nm maximum[25]. Two glass fiber filters with a battery-powered pump which were used for the in-situ sampling were used for the performance test. The sampling flow rate was set to 2 L min^{-1} and radon decay products were collected during 3.5 h. After aerosol samples were collected, the gross alpha measurements were recorded over consecutive 1 minute intervals during a total recording period of 60 minutes. Moreover, an imaging plate technique (BAS-MS 2025, Fuji Film Co.) was used in order to obtain the distribution images of the radon decay products on the glass fiber filters. All radionuclides other than ^{3}H, which has a low beta energy of 18.6 keV, can be detected by this technique[26]. Information in the imaging plate was read out after 3 days using a reading system (FLA-5100, Fuji Film Co.). Gradation and resolution for the reading system were set to 16 bits and 25 μm, respectively.

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Author contributions

M.H., S.T. and I.K. designed the study; M.H., S.T., S.M., M.O., Masatoshi Yamada, A.N., Mitsuaki Yoshida, H.Y. and I.K. carried out field measurements and sample preparations; H.T. and Masatoshi Yamada measured filter samples; M.H., A.S., S.S., N.A., H.K. and Y.M. analyzed gamma spectrum; M.H., Y.O. and T.I. carried out experiment at NIRS radon chamber; M.H., S.T. and T.K. wrote the manuscript; S.T. supervised the study. All authors contributed extensively to discussions about this work and in reviewing the manuscript.

Additional information

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