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Distribution of artificial radionuclides (^{110m}Ag, ^{129m}Te, ^{134}Cs, ^{137}Cs) in surface soils from Miyagi Prefecture, northeast Japan, following the 2011 Fukushima Dai-ichi nuclear power plant accident

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The Tohoku earthquake and tsunami occurred off the Pacific coast of Japan in March 2011 and caused the Fukushima Dai-ichi nuclear power plant accident. To assess the distribution of artificial radionuclides in Miyagi Prefecture (northeastern Japan), we measured the activity concentrations of ^{110m}Ag, ^{129m}Te, ^{134}Cs, and ^{137}Cs in surface soil samples that were collected from 60–190 km north of the power plant approximately 1 month (April 16–29, 2011) after the earthquake. Since the studied samples were obtained from depths of 0–1 cm, the activity concentrations we determined were higher than those reported by the Japanese Government, whose measurements were based on soil samples obtained from depths of 0–5 cm. We found relatively high concentrations of radionuclides in surface soils in the southern and northern parts of Miyagi Prefecture. In the south, close to the border with Fukushima Prefecture, the total activity concentrations in the period April 16–29, 2011, reached approximately 27,600 Bq/kg dry soil. Relatively high total concentrations (2600–6600 Bq/kg dry soil) were also found in the northern part of Miyagi Prefecture (north Osaki, Kurihara, and Kesen-numa), but relatively low concentrations (400–1900 Bq/kg dry soil) were found in Sendai city and other areas in central Miyagi Prefecture (Higashimatsushima, south Osaki, Ishinomaki, and Onagawa). The concentrations found are consistent with the spatial variation in radiation dose rates in the air in Miyagi Prefecture.

Keywords: Fukushima Dai-ichi nuclear power plant accident, Miyagi Prefecture, radioactivity, caesium, surface soils

INTRODUCTION

The Fukushima Dai-ichi nuclear power plant (37°25′17″ N, 141°01′57″ E; Fig. 1a) incurred seriously damage because of the Tohoku Earthquake and tsunami that occurred off the Pacific coast on March 11, 2011 (magnitude 9.0; JMA, 2011). Consequently, large quantities of artificial radionuclides were released from the plant (NERH, 2011; Hirano *et al*., 2012; Katata *et al*., 2012). On April 12, 2011, the amount of radioactive caesium (^{137}Cs) released was estimated to be 15,000 TBq (1.5×10^{16} Bq; Chino *et al*., 2011; NERH, 2011). Using an atmospheric transport model and measurement data from 43 stations worldwide, Stohl *et al*. (2011) calculated that the power plant released approximately 3.5×10^{16} Bq of ^{137}Cs following the accident. Radionuclides from the power plant were dispersed in the air and ocean and deposited on land (Yasunari *et al*., 2011; Gudelis *et al*., 2012; Honda *et al*., 2012; Xiong *et al*., 2012). ^{134}Cs and ^{137}Cs were transported from the power plant to Taiwan (up to ~1.5×10^{-13} Bq/m^3 in aerosol samples; Huh *et al*., 2011), and airborne radionuclides were detected in Europe from March 22, 2011 (Masson *et al*., 2011; Beresford *et al*., 2012). Morino *et al*. (2011) reported that approximately 22% of the ^{137}Cs emitted from the power plant was deposited in Japan in March 2011. The Japanese Government reported the radionuclide activity concentrations in soils (0–5 cm depth) collected from Fukushima and southern Miyagi Prefecture (within a 100-km radius of the power plant) following the accident (MEXT, 2011a). On April 28, 2011, relatively high ^{134}Cs and ^{137}Cs activity concentrations (up to 8800 and 9400 Bq/kg, respectively; Kato *et al*., 2012) were found in the upper-

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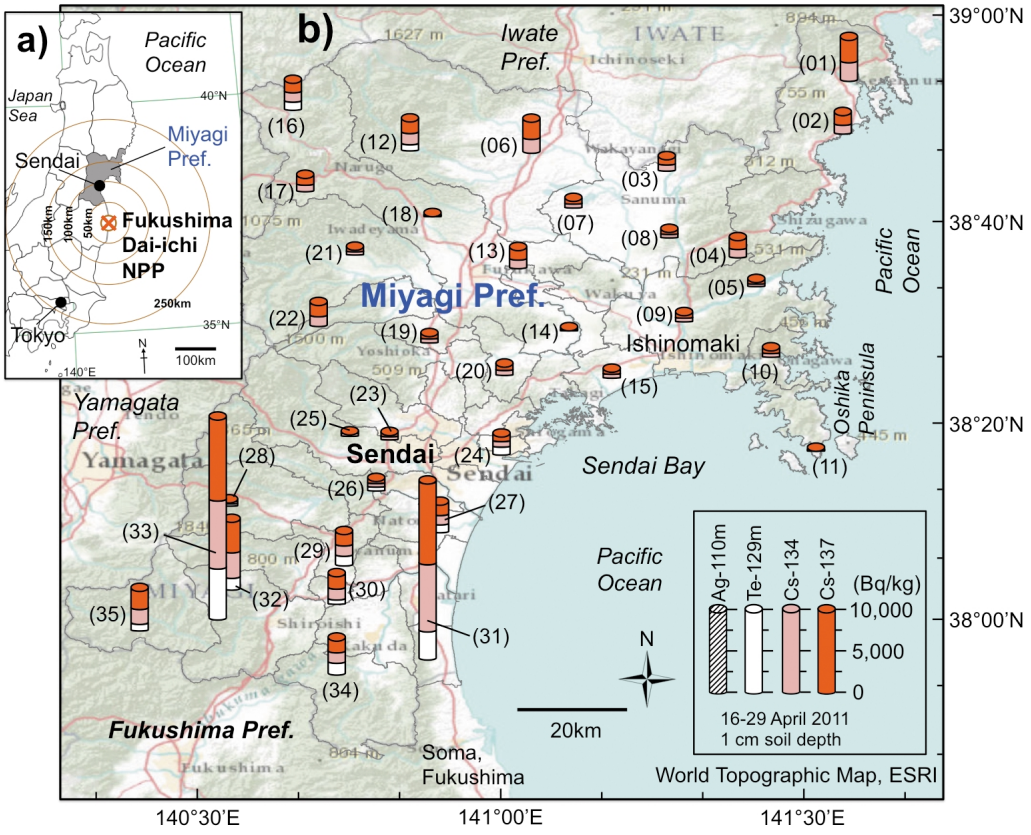

Fig. 1. a) Location of the study area (Miyagi Prefecture) and the Fukushima Dai-ichi Nuclear Power Plant (NPP). b) Distribution of artificial radionuclides (^{110m}Ag, ^{129m}Te, ^{134}Cs, ^{137}Cs) in surface soils (0–1 cm depth) in Miyagi Prefecture during the period April 16–29, 2011, following the March 2011 Fukushima Dai-ichi NPP accident (World Topographic Map; Esri, DeLorme, NAVTEQ, TomTom, Intermap, iPC, USGS, FAO, NPS, NRCAN, GeoBase, Kadaster NL, Ordnance Survey, Esri Japan, METI, Esri China (Hong Kong), and the GIS User Community). The numbers in parentheses indicate the soil sample number.

most layers (0.0–0.5 cm depth) of soil from the town of Kawamata, Fukushima Prefecture, approximately 50 km northwest of the power plant. In June 2011, the ^{137}Cs activity concentrations in agricultural soils of Miyagi Prefecture ranged from 24 to 1152 Bq/kg (MAFF, 2011).

The Japanese Government reported relatively high radiation dose rates in the air (0.2–0.5 μSv/h; MEXT, 2011b) on June 30, 2011 in the vicinity of Kurihara city, northern Miyagi Prefecture (Fig. 1b). These values were an order of magnitude higher than the average background values in Miyagi Prefecture prior to the accident (0.02–0.05 μSv/h; MEXT, 2011c). However, the distribution of radionuclides in soils throughout Miyagi Prefecture, including the central and northern parts beyond a 100-km radius from the power plant, was unknown. The main objective of our study was to assess the spatial distribution of radionuclides in surface soils from all areas of Miyagi Prefecture. To estimate the direct transport of radionuclides from the Fukushima Dai-ichi nuclear power plant to Miyagi Prefecture following the accident, we measured the activity concentrations of artificial radionuclides (^{110m}Ag, ^{129m}Te, ^{134}Cs, and ^{137}Cs) in surface soils (0–1 cm soil depth) collected over the period April 16–29, 2011 (approximately one month after the accident), from 35 sites in an area of Miyagi Prefecture 60–190 km north of the power plant. In 2011, the Japanese Government reported radionuclide activity concentrations in surface soils obtained from soil depths of 0–5 cm (MEXT, 2011a, d).

Depth profiles of radionuclides in soils from Fukushima have been reported (Kato *et al.*, 2012; Ohno *et al.*, 2012; Tanaka *et al.*, 2012). Kato *et al.* (2012) reported that following the accident, approximately 60% of the total radioactive caesium was in the top 0–1 cm of the soil in Fukushima Prefecture and 96% in the top 0–5 cm, and radionuclides were detected to a depth of 20 cm. Because of the reported high concentrations of radionuclides in surface soils (0–1 cm depth; Kato *et al.*, 2012) and from the perspective of risk and land-use assessment, we measured the activity concentrations of the radionuclides in the 0–1 cm surface-soil layer. Therefore, our results are not directly comparable to the Environ-

Table 1. ^{110m}Ag, ^{129m}Te, ^{134}Cs, and ^{137}Cs activity concentrations in surface soils (0–1 cm depth) from Miyagi Prefecture, Japan

Sample name	Sampling date	Latitude	Longitude	City	Water content (%)	Dry sample wt. (g)	^{110m}Ag* (Bq/kg) 249.8 d**	^{129m}Te* (Bq/kg) 33.6 d**	^{134}Cs* (Bq/kg) 2.06 yr**	^{137}Cs* (Bq/kg) 30.07 yr**	$^{134}Cs/^{137}Cs$ (ratio)	$^{129m}Te/^{137}Cs$ (ratio)
Soil 01	2011.4.26	38°55′	141°34′	Kesen-numa	25.0	20.2	<5	<1100	3202 ± 147	3403 ± 154	0.94	
Soil 02	2011.4.26	38°49′	141°34′	Kesen-numa	35.6	26.7	<4	<700	1670 ± 76	1765 ± 80	0.95	
Soil 03	2011.4.20	38°45′	141°16′	Tome	22.4	78.9	3.0 ± 1.4	<930	1169 ± 54	1188 ± 55	0.98	
Soil 04	2011.4.20	38°37′	141°24′	Tome	25.8	95.2	9.2 ± 2.2	<1020	1584 ± 73	1638 ± 76	0.97	
Soil 07	2011.4.20	38°42′	141°07′	Tome	10.3	83.2	3.7 ± 0.8	<490	778 ± 36	824 ± 37	0.94	
Soil 08	2011.4.20	38°39′	141°16′	Tome	22.6	57.4	<4	<860	667 ± 31	718 ± 33	0.93	
Soil 06	2011.4.20	38°47′	141°03′	Kurihara	18.1	25.7	<8	<1900	2584 ± 119	2739 ± 125	0.94	
Soil 12	2011.4.21	38°47′	140°51′	Kurihara	14.2	84.8	<4	1000 ± 400	2081 ± 95	2137 ± 98	0.97	0.47
Soil 16	2011.4.21	38°52′	140°39′	Osaki	30.0	29.5	<4	1400 ± 500	1754 ± 80	1837 ± 84	0.95	0.76
Soil 17	2011.4.21	38°44′	140°41′	Osaki	29.0	32.4	<5	<1300	1247 ± 58	1309 ± 98	0.95	
Soil 18	2011.4.21	38°40′	140°53′	Osaki	11.4	67.0	<2	<430	246 ± 11	250 ± 11	0.98	
Soil 13	2011.4.21	38°36′	141°02′	Osaki	11.8	49.8	10.0 ± 1.0	<940	1651 ± 77	1748 ± 80	0.94	
Soil 14	2011.4.21	38°29′	141°07′	Osaki	9.2	101.3	<3	<530	207 ± 10	207 ± 10	1.00	
Soil 05	2011.4.16	38°34′	141°25′	Ishinomaki	2.2	49.5	<4	<1000	540 ± 25	604 ± 28	0.89	
Soil 09	2011.4.16	38°30′	141°18′	Ishinomaki	0.9	50.1	4.1 ± 1.6	<1100	712 ± 33	736 ± 34	0.97	
Soil 10	2011.4.16	38°27′	141°27′	Onagawa	20.0	43.1	8.2 ± 1.3	<820	727 ± 34	779 ± 36	0.93	
Soil 11	2011.4.29	38°17′	141°31′	Ishinomaki	18.1	98.1	<3	<560	291 ± 14	320 ± 15	0.91	
Soil 15	2011.4.26	38°25′	141°11′	Higashimatsushima	20.8	39.2	3.3 ± 1.6	<1000	752 ± 35	806 ± 38	0.93	
Soil 19	2011.4.21	38°28′	140°52′	Ohira	21.8	39.5	2.6 ± 0.7	<590	748 ± 34	811 ± 37	0.92	
Soil 20	2011.4.21	38°25′	141°01′	Osato	22.3	42.0	<5	<1200	940 ± 43	968 ± 44	0.97	
Soil 21	2011.4.21	38°37′	140°45′	Kami	12.6	61.7	<4	<960	634 ± 29	674 ± 31	0.94	
Soil 22	2011.4.21	38°30′	140°42′	Shikima	5.3	29.3	4.0 ± 1.7	<1100	1851 ± 85	1964 ± 89	0.94	
Soil 24	2011.4.21	38°18′	141°00′	Tagajo	9.8	71.3	4.6 ± 1.1	1300 ± 500	1022 ± 47	1095 ± 50	0.93	1.19
Soil 23	2011.4.21	38°19′	141°49′	Sendai	5.9	89.7	<3	<840	596 ± 28	654 ± 30	0.91	
Soil 25	2011.4.21	38°19′	141°45′	Sendai	9.4	42.5	<3	<650	322 ± 15	350 ± 16	0.92	
Soil 26	2011.4.25	38°14′	141°48′	Sendai	12.8	132.1	<3	620 ± 320	628 ± 29	652 ± 31	0.96	0.95
Soil 27	2011.4.25	38°10′	140°53′	Natori	6.2	141.9	4.4 ± 1.4	1300 ± 500	1785 ± 82	1853 ± 84	0.96	0.70
Soil 28	2011.4.29	38°12′	140°32′	Kawasaki	9.9	112.2	<2	430 ± 230	404 ± 19	414 ± 19	0.98	1.04
Soil 29	2011.4.25	38°07′	140°44′	Murata	23.8	103.0	<5	1500 ± 600	1959 ± 90	1971 ± 90	0.99	0.76
Soil 30	2011.4.25	38°03′	140°44′	Ogawara	4.9	149.2	6.4 ± 1.4	770 ± 330	2058 ± 94	2119 ± 96	0.97	0.36
Soil 31	2011.4.25	37°57′	140°53′	Yamamoto	34.7	55.9	36.4 ± 2.6	3970 ± 660	10,120 ± 460	10,490 ± 480	0.96	0.38
Soil 32	2011.4.29	38°04′	140°34′	Shiroishi	0.6	152.6	17.1 ± 2.8	1900 ± 600	4217 ± 193	4359 ± 198	0.97	0.44
Soil 33	2011.4.29	38°01′	140°32′	Shiroishi	0.3	141.7	48.4 ± 4.2	6800 ± 1300	10,210 ± 470	10,580 ± 480	0.97	0.64
Soil 34	2011.4.25	37°56′	140°44′	Marumori	17.5	100.7	<5	2000 ± 700	2016 ± 92	2066 ± 94	0.98	0.97
Soil 35	2011.4.29	38°00′	140°24′	Shichikasyuku	3.5	78.6	11.9 ± 1.6	1100 ± 500	2698 ± 123	2827 ± 128	0.95	0.39

**Activity at sampling date.*
***Half-life.*

mental Quality Standards or the values reported from soil depths of 0–5 cm by MEXT (2011a); despite this, our data on the activity concentrations of radionuclides in surface soils and their spatial distribution in Miyagi Prefecture are critical for risk assessment and planning.

Materials and Methods

During the period April 16–29, 2011 (109–130 days following the Fukushima accident), 35 soil samples were collected from Miyagi Prefecture, Japan (37°46′ N–39°00′ N, 140°24′ E–141°34′ E; Table 1 and Fig. 1). At each site, the sample was collected from soil depths of 0–1 cm over an area of approximately 400–900 cm^2 using a plastic shovel. Each sample was air dried at room temperature for 1–2 weeks, coarse particles (the gravel fraction) were removed by dry sieving (2-mm opening sieve), and the fraction <2 mm was transferred to a plastic container (U-8 polystyrene vessel, 100 cm^3). The air-dried soil weight (<2 mm fraction) used for radionuclide activity measurements ranged from 20 to 153 g in weight (Table 1). The activity concentrations of the radionuclides ^{110m}Ag, ^{129m}Te, ^{134}Cs, and ^{137}Cs in the samples were measured by gamma-ray spectroscopy using a Ge semiconductor detector (GEM-25185, ORTEC). The measurements were performed at the Tokyo Metropolitan University during the period August 13–September 2, 2011 (approximately 5–6 months after the accident), and the activity concentrations of the radionuclides for each sampling time were calculated. The half-lives ($T_{1/2}$) of ^{110m}Ag, ^{129m}Te, ^{134}Cs, and ^{137}Cs are 249.6 days, 33.6 days, 2.06 years, and 30.07 years, respectively (Table 1). The detection limits were calculated from the three sigma values of the background counts. The counting efficiency for the soil sample with height of 5 cm was measured using calibrated gamma ray sources (MX033U8PP [^{109}Cd, ^{57}Co, ^{139}Ce, ^{51}Cr, ^{85}Sr, ^{137}Cs, ^{54}Mn, ^{88}Y, ^{60}Co] and MX035U8PP [^{134}Cs, ^{137}Cs]; Japan Radioisotope Association), and correction factor to sample height was obtained using the soils sampled in Fukushima prefecture with different heights (1, 2, 3, 4, 5, and 6 cm). The radionuclide activity concentrations for each sampling time were expressed as Bq per kg dry soil (<2 mm fraction).

Results and Discussion

Distribution of ^{110m}Ag (half-life: 249.8 days)

During the study period, ^{110m}Ag was detected in 16 of the soil samples from Miyagi Prefecture at activity concentrations that ranged from 0 (less than the detection limit) to 48 Bq/kg dry soil (Table 1). Relatively high values were found at Shiroishi city (Soils 32 and 33 with 17 and 48 Bq/kg dry soil, respectively) and Yamamoto town (Soil 31, 36 Bq/kg dry soil) in the south of Miyagi Prefecture. These areas are close to the border with Fukushima Prefecture (about 60 km north of the Fukushima Dai-ichi nuclear power plant). In other areas, the ^{110m}Ag activity concentrations were <12 Bq/kg dry soil. ^{110m}Ag was also detected in southern Miyagi Prefecture (Shiroishi city, Marumori city, and Yamamoto and Watari towns; Fig. 1b; MEXT, 2011d).

Distribution of ^{129m}Te (half-life: 33.6 days)

Relatively high ^{129m}Te activity concentrations were found at Shiroishi city and Yamamoto town (up to 6800 Bq/kg dry soil; Table 1 and Fig. 1). This indicates that the radionuclides were dispersed rapidly in Miyagi Prefecture, because ^{129m}Te has a short half-life ($T_{1/2}$ = 33.6 days). The Japan Atomic Energy Agency simulated the radionuclide fallout in northeast Japan from the afternoon of March 12, 2011, following the Fukushima accident (JAEA, 2011), and Hosoda *et al*. (2011) reported increased dose rates in the air of Miyagi Prefecture during the period March–April 2011. Relatively high concentrations of ^{129m}Te were reported in Soma city, Fukushima (approximately 40 km north of the power plant), with concentrations of approximately 6000 Bq/kg dry soil in April 2011 and up to 1600 Bq/kg dry soil in June 2011 (0–5 cm depth; MEXT, 2012). ^{129m}Te was also detected in the south of Miyagi Prefecture (Shiroishi city, Marumori city, Kakuda city, and Yamamoto town; MEXT, 2011d). In this study ^{129m}Te was found south of Tagajo (Soils 24, 26–35), and in the northwest of Miyagi Prefecture (north Osaki and Kurihara; Soils 12 and 16; Fig. 1b).

Distributions of ^{134}Cs (half-life: 2.06 years) and ^{137}Cs (half-life: 30.07 years)

^{134}Cs and ^{137}Cs were detected in all 35 samples from Miyagi Prefecture, with activity concentrations ranging from 207 to 10,210 Bq/kg dry soil for ^{134}Cs, and from 207 to 10,580 Bq/kg dry soil for ^{137}Cs (Table 1). Relatively high ^{134}Cs and ^{137}Cs activity concentrations were detected in the south of Miyagi Prefecture (Soils 29–35; Fig. 1b). The concentrations were especially high in Shiroishi city and Yamamoto town (Soils 31 and 33, respectively; up to 10,580 Bq/kg dry soil) and were similar to the concentration ranges of ^{110m}Ag and ^{129m}Te (Table 1). The ^{137}Cs activity concentrations in the south of Miyagi Prefecture at Shichikasyuku (Soil 35) and Marumori (Soil 34) were 2830 and 2070 Bq/kg dry soil, respectively. In southern parts of Shiroishi and Marumori cities, relatively high atmospheric radiation dose rates of total radionuclides were reported (0.5–1.0 μSv/h on June 30, 2011; MEXT, 2011b).

In central Miyagi Prefecture (Sendai, Higashimatsushima, south Osaki, and Ishinomaki city; Soils 5, 9–11, 14–15, 23, 25, and 26), the activity concentrations were <1000 Bq/kg dry soil for each

radionuclide. However, the activity concentrations reached 1100 and 1850 Bq/kg dry soil in Tagajo (Soil 24) and Natori (Soil 27), respectively. Relatively high activity concentrations were also found in the north of Miyagi Prefecture (north Osaki, Kurihara city, and Kesen-numa city; up to ^{134}Cs = 3200 Bq/kg dry soil and ^{137}Cs = 3400 Bq/kg dry soil; Table 1). Although Kesen-numa city (Soil 1; Fig. 1b) is approximately 190 km north of the power plant, the activity concentrations there were higher than in soils from parts of southern Miyagi Prefecture (Soils 29 and 30). In western and central Tome city (Soils 3, 7, and 8), relatively low activity concentrations were found (670–1170 and 720–1190 Bq/kg dry soil for ^{134}Cs and ^{137}Cs, respectively; Fig. 1b). The air radiation dose rates in north Osaki, Kurihara city, Kesen-numa city, and in the eastern part of Tome city were relatively high (0.1–0.5 μSv/h on June 30, 2011; MEXT, 2011b). Therefore, the variations in the radionuclide activity concentrations found in the surface soils in this study are consistent with the air radiation dose rates reported for Miyagi Prefecture (MEXT, 2011b). In addition, the distribution of ^{137}Cs found in this study corresponds with the ^{137}Cs deposition rates in Miyagi Prefecture in the period March 11–29, 2011, as simulated by Morino *et al*. (2011).

^{134}Cs/^{137}Cs and ^{129m}Te/^{137}Cs ratios in the surface soils

During the study period, the ^{134}Cs/^{137}Cs ratios in surface soils from throughout Miyagi Prefecture were approximately 1 (range 0.89–1.00; average, 0.95; Table 1). Prior to March 11, 2011, the amount of ^{134}Cs ($T_{1/2}$ = 2.06 years) in the soil was negligible, and most of the ^{137}Cs ($T_{1/2}$ = 30.07 years) present in the soil originated from nuclear weapons tests and the Chernobyl nuclear power plant accident (Hisamatsu and Takizawa, 1990; Andersson and Roed, 1994). Following the Fukushima Dai-ichi nuclear power plant accident, the ^{134}Cs/^{137}Cs ratio in drainage from discharge canals of the power plant was 1.00 (March 21, 2011; TEPCO, 2011). A constant ^{134}Cs/^{137}Cs ratio (average = 0.92; from April 14, 2011 to May 3, 2011) in the surface seawater of the western North Pacific, approximately 200 km east of the power plant, was also reported (Honda *et al*., 2012). In addition, the ^{134}Cs/^{137}Cs ratios in soils from depths of 0–5 cm from Fukushima Prefecture were approximately 1.0 in June 2011 (MEXT, 2011a). Considering the constant ^{134}Cs/^{137}Cs ratio in our study (average, 0.95; Table 1), we conclude that ^{137}Cs found in the surface soils of Miyagi Prefecture area derived from the Fukushima Dai-ichi nuclear power plant.

In contrast, the ^{129m}Te/^{137}Cs ratios in the soils from Miyagi Prefecture varied widely, ranging from 0.36 to 1.19 (Table 1). Relatively high ^{129m}Te/^{137}Cs ratios were observed in Marumori (Soil 34), Kawasaki (Soil 28), Sendai (Soil 26), and Tagajo (Soil 24). Varying ^{129m}Te/^{137}Cs ratios were also reported in Fukushima Prefecture (MEXT, 2011d). Relatively high ^{129m}Te/^{137}Cs ratios (1.4–1.8 in April 2011) were found in Naraha town, Fukushima Prefecture (approximately 20 km south of the power plant; Tagami *et al*., 2011). Caesium is readily adsorbed by surface soils and its migration rate in soil is relatively low (Andersson and Roed, 1994; Ohta *et al*., 2012). However, tellurium migrates in soil because in oxic aqueous environments, it exists mainly in the form of oxyanions or hydroxide ions (Harada and Takahashi, 2009). The variations in the ^{129m}Te/^{137}Cs ratio may have been a consequence of ^{129m}Te migration in the soil following radionuclide deposition and/or its physical properties, such as boiling point. The boiling points of Ag, Te, and Cs are 2160, 990 and 670°C, respectively (MEXT, 2011d).

Total radionuclides in the surface soils from Miyagi Prefecture following the Fukushima nuclear accident

The total artificial radionuclide activity concentrations (^{110m}Ag, ^{129m}Te, ^{134}Cs, and ^{137}Cs) in surface soils from throughout the Miyagi area during the study period ranged from 400 to 26,920 Bq/kg dry soil (Table 1 and Fig. 1b). In the south (close to the border of Fukushima Prefecture) and north (north Osaki, Kurihara city, and Kesen-numa city) of Miyagi Prefecture, the total radionuclide activity concentrations were relatively high (up to approximately 27,600 Bq/kg dry soil and 6600 Bq/kg dry soil, respectively; Table 1). In contrast, the total activity concentrations were relatively low (400–1900 Bq/kg dry soil) in Sendai (the capital city of Miyagi Prefecture) and other central areas of Miyagi Prefecture (Higashimatsushima, south Osaki, Ishinomaki, and Onagawa; Table 1). The artificial radionuclide activity concentrations in the south of Miyagi Prefecture, close to the border of Fukushima Prefecture, were up to 70 times those from central Miyagi Prefecture.

In this study, we assessed the spatial distribution of ^{110m}Ag, ^{129m}Te, ^{134}Cs, and ^{137}Cs in surface soils (0–1 cm depth) from the Miyagi area during the period April 16–29, 2011, following the Fukushima Dai-ichi nuclear power plant accident in March 2011. MEXT (2011a) reported the concentrations of radionuclides in soil (0–5 cm depth) from the south of Miyagi Prefecture within a 100-km radius of the power plant. The trends in our results from southern Miyagi Prefecture (Fig. 1b) are consistent with the values reported by MEXT (2011a). In MEXT (2012), the soil activity concentrations showed large variations within a relatively small area. For example, the ^{137}Cs activity concentrations in soil from Kawamata town (area = 128 km^2) in Fukushima Prefecture varied from 770 to 34,000 Bq/kg dry soil in the period March–May 2011. Thus, it is difficult to estimate the inventory of the radionuclides based on our analysis and to compare the results quantitatively with MEXT (2011a), since (i) we did not collect a large number of samples at each site and

(ii) our data are confined to the soil from depths of 0–1 cm. However, the inventories of ^{134}Cs and ^{137}Cs were roughly estimated to be 2–123, 1–15, and 1–7 kBq/m^2 in the south, north and central areas of Miyagi Prefecture, respectively, on the basis of our survey and assuming that 60% of the radiocaesium was distributed within the 0–1 cm depth layer. The values of the total cumulative amounts of ^{134}Cs and ^{137}Cs are consistent with those of the airborne survey in Miyagi Prefecture reported by MEXT (2011b), demonstrating the validity of our results.

Because the half-life of ^{137}Cs is relatively long ($T_{1/2}$ = 30.07 years), continuous and long-term monitoring of the activity concentrations in soils is needed in Miyagi Prefecture and eastern Japan. Although surface soils and clay particles adsorb radioactive caesium (Squire and Middleton, 1966; Chikasawa *et al*., 2005; Ohta *et al*., 2012), migration of the radionuclides into river water, groundwater, and vegetation should be monitored.

Conclusions

The spatial distributions of ^{110m}Ag, ^{129m}Te, ^{134}Cs, and ^{137}Cs in surface soils (0–1 cm depth) from Miyagi Prefecture (northeastern Japan) during the period April 16–29, 2011, following the Fukushima Dai-ichi nuclear power plant accident in March 2011, can be summarized as follows:

1) ^{134}Cs and ^{137}Cs were found in all 35 samples at concentrations of 207–10,210 Bq/kg dry soil and 207–10,580 Bq/kg dry soil, respectively. The ^{134}Cs activity concentrations were similar to those of ^{137}Cs (^{134}Cs/^{137}Cs ratio = 0.87–0.98; average, 0.93). In addition, radionuclides with relatively short half-lives (^{110m}Ag, $T_{1/2}$ = 249.8 days and ^{129m}Te, $T_{1/2}$ = 33.6 days) were detected at 16 and 12 of the sampling locations, respectively. Therefore, we conclude that the artificial radionuclides were rapidly dispersed and deposited in all areas of Miyagi Prefecture following the Fukushima accident.

2) There were high total activity concentrations (up to approximately 27,600 Bq/kg dry soil) of the four radionuclides in surface soils from the south of Miyagi Prefecture, close to the border of Fukushima Prefecture. The total activity concentrations in southern Miyagi Prefecture were up to approximately 70 times higher than those from central Miyagi Prefecture (the Sendai city, Higashimatsushima, south Osaki, Ishinomaki, and Onagawa).

3) Relatively high total activity concentrations of the artificial radionuclides were found in northern Miyagi Prefecture (north Osaki, Kurihara city and Kesen-numa city; up to approximately 6600 Bq/kg dry soil). The variations in the radionuclide activity concentrations in the surface soils were consistent with the radiation dose rates in the air.

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