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Short communication

Radiocaesium activity concentrations in parmelioid lichens within a 60 km radius of the Fukushima Dai-ichi Nuclear Power Plant

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ABSTRACT

Radiocaesium activity concentrations (^{134}Cs and ^{137}Cs) were measured in parmelioid lichens collected within the Fukushima Prefecture approximately 2 y after the Fukushima Dai-ichi Nuclear Power Plant accident. A total of 44 samples consisting of nine species were collected at 16 points within a 60 km radius of the Fukushima Dai-ichi Nuclear Power Plant. The activity concentration of ^{134}Cs ranged from 4.6 to 1000 kBq kg^{-1} and for ^{137}Cs ranged from 7.6 to 1740 kBq kg^{-1}. A significant positive correlation was found between the ^{137}Cs activity concentration in lichens and the ^{137}Cs deposition density on soil ($n = 44$), based on the calculated Spearman's rank correlation coefficients as $r = 0.90$ ($P < 0.01$). The two dominant species, *Flavoparmelia caperata* ($n = 12$) and *Parmotrema clavuliferum* ($n = 11$), showed strong positive correlations, for which the r values were calculated as 0.92 ($P < 0.01$) and 0.90 ($P < 0.01$) respectively. Therefore, *Flavoparmelia caperata* and *Parmotrema clavuliferum* are suggested as biomonitoring species for levels of radiocaesium fallout within the Fukushima Prefecture.

1. Introduction

Lichens are symbiotic organisms consisting of fungi and algae (and/or cyanobacteria), and are found in almost all terrestrial habitats e.g. on rock, tree bark and soil (Nash, 2008). Lichens have frequently been used as a means for obtaining information on radioactive fallout contamination for more than 50 y because (1) they grow on a wide variety of substrata, (2) they have no root system but through their entire thallus, accumulate greater activity concentrations of radionuclides in a passive way, and (3) they retain radionuclides in the thalli for long periods of time due to their longevity (Svensson and Lidén, 1965; Nimis, 1996; Thomas and Gates, 1999; Seaward, 2002). Many studies of radionuclide monitoring have been carried out in high-latitude regions using lichens, mainly for the following reasons; the fallout from atmospheric nuclear weapons testing effects on the food chains lichen-caribou-man (Nimis, 1996), and after the Chernobyl accident, much attentions were paid in not only high-latitude regions but also mid-latitude regions which were affected by the fission nuclides transferred from Chernobyl (Sloof and Wolterbeek, 1992; Nimis, 1996; Seaward, 2002; Sawidis et al., 2010; Iurian et al., 2011). Species commonly used for these studies were e.g. *Cladonia* spp., *Hypogymnia physodes* (L.) Nyl., *Pseudevernia furfuracea* (L.) Zopf, and *Umbilicaria* spp. These species have been studied since the first concerns of fallout from atmospheric nuclear weapons testing and, even more so, since the Chernobyl accident of 1986 (e.g. Tuominen and Jaakkola, 1973; Seaward, 2002; Puhakainen et al., 2007; Sawidis et al., 2010).

Large quantities of radionuclides, including ^{134}Cs and ^{137}Cs, were released into the atmosphere from the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident caused by an earthquake and tsunami on 11 March 2011 (Nuclear Emergency Response Headquarters Government of Japan, 2011; Fukushima Prefecture, 2012). In addition, ^{134}Cs and ^{137}Cs were also observed to have been deposited as fallout over a wide area of eastern Japan, especially within the Fukushima Prefecture (MEXT, 2012).

As lichens are known to be very effective tools to monitor environmental radionuclides in both time and space (Seaward, 2002), lichen samples can be used to examine and monitor radioactive fallout in the vicinity of the FDNPP. However, Japan belongs to a different phytogeographic area from Europe (Kurokawa, 1975) and therefore it is difficult to use the European boreal species of

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lichens for radionuclide monitoring and/or as an indicator of radionuclide fallout in Japan. After the FDNPP accident, Ohmura et al. (2013, 2015) investigated radioactive contamination in several lichen species occurring in an urban area, e.g. *Dirinaria applanata* (Fée) D.D. Awasthi, *Hyperphyscia crocata* Kashiw., *Phaeophyscia spinellosa* Kashiw., and *Physcia orientalis* Kashiw. These samples were collected in Tsukuba City located ca. 170 km south from the FDNPP. However, the Japanese Archipelago stretches from subarctic to subtropical zones, and the bioclimatic vegetation of Fukushima (hemitemperate climate) is somewhat different to that of Tsukuba (southern, middle and northern meridional climate) (Hämet-Ahti et al., 1974). In order to investigate the use of lichens as radionuclide monitors and/or indicators of fallout in and around the Fukushima Prefecture, dominant species of macrolichens should be studied. We focused on parmelioid lichens which commonly grow on the trunks of *Prunus* spp. (Japanese Cherry trees) within the Fukushima area. Parmelioid lichens (Parmeliaceae, Ascomycota) have mostly foliose, dorsiventral thalli, and usually rhizines (root-like filaments) are present on the lower surface (Crespo et al., 2010).

The purpose of this study is (1) to investigate the activity concentration of radiocaesium in parmelioid lichens around the area of the FDNPP and (2) to examine the relationship between the radiocaesium activity concentration in lichen and radiocaesium fallout.

2. Materials and methods

2.1. Study area and sampling

Samples were collected at 16 different points within a radius of 60 km from the FDNPP between 17 December 2012 and 5 February 2013. Sampling point's elevation varied between 10 and 510 m above sea level and also had varying air dose rates (Fig. 1 and Table 1). The distance and direction of the sampling points from the FDNPP were calculated by a Japanese government organization website tool (Geospatial Information Authority of Japan, 2013).

A total of 44 lichen samples were collected from trunks of *Prunus* spp., Japanese cherry, at a height of between 1 and 2 m above the ground. All samples were deposited as voucher specimens in the National Museum of Nature and Science, Tsukuba, Japan (herbarium code: TNS). Collection data are summarized in Table 1.

2.2. Calculation of ^{137}Cs deposition density on soil and air dose rate measurement

The ^{137}Cs inventory in soil at each sampling point was estimated by means of the GIS software (ArcGIS, ESRI Japan) with the Inverse Distance Weighted (IDW) (Watson and Philip, 1985) before being decay corrected to the date of either at the final sampling date (5 February 2013) or at each sampling date of lichens. The data for the interpolation was obtained by MEXT, who collected soil samples within a 100 km radius of the FDNPP between June and July of 2011 (MEXT, 2011a, 2011b; Saito et al., 2015).

Air dose rates, which were defined as the ambient equivalent dose rate of gamma radiation including the contribution from natural radiation, H*(10), at each sampling point was measured at a height of 1 m above the ground by means of a NaI scintillation survey meter (ALOKA, TCS-172B). These results are summarized in Table 2. The reading uncertainty in case of digital indication was within 15% (ALOKA, TCS-172B instruction manual).

2.3. Identification of lichens

Lichen identification was carried out based on both their morphological characteristics using a dissecting stereomicroscope and additionally on analysis of their secondary metabolites (lichen substances). The metabolites were determined by thin layer chromatography (TLC) using the "solvent B system" as detailed in Culberson and Kristinsson (1970) and Culberson and Johnson (1982).

2.4. Measurement for radiocaesium in lichens

Lichen samples were cleaned with tweezers to remove bark and any debris and then air-dried at room temperature for a month. Samples were placed into 50 ml cylindrical plastic containers (38.0 mm $\phi \times$ 59.5 mm high) for radioactivity measurement. Dry weights of samples ranged from 1.74 to 4.84 g (Table 1).

The activity concentration of ^{134}Cs and ^{137}Cs in lichen samples were measured by γ-ray spectrometry using a CsI scintillation detector (shielded by lead with 50 mm thickness), coupled to a multichannel analyzer (FD-08Cs40, Techno-X, Osaka, Japan). Areas under the energy peaks 795 keV and 801 keV assigned to ^{134}Cs, and 661 keV for ^{137}Cs were calculated by spectrum analysis software (CsAnalyzer, Techno-X). Peak positions were calibrated using ^{137}Cs/^{40}K mixed source so that each channel width was adjusted to be 1 keV. The energy resolution (full width at half-maximum, FWHM) for the detector was between 9 and 11% at 662 keV for ^{137}Cs. One sample of *Parmotrema tinctorum*, the radiocaesium (^{134}Cs + ^{137}Cs) activities of which was defined by means of germanium (Ge) semiconductor detector (GC2518-7500SL-2002CSL, Canberra, Tokyo, Japan), was used as a calibration source for measurement parmelioid lichen samples. The Ge detector was calibrated with the 50 ml mixed source solution which was prepared by spiking 170 μl and 50 μl of standard solution of ^{137}Cs (CS010) and ^{152}Eu (EU010), respectively, and filled in the plastic container used for the CsI measurement. Both standard solutions were certified by Japan Radioisotope Association (Tokyo, Japan). The activities for ^{134}Cs and ^{137}Cs in all lichen samples were decay corrected to the date of either at the final sampling date or at each sampling date in order to analyze the relationship between radiocaesium activity concentration in lichens and the ^{137}Cs deposition density on soil or the air dose rate at each of the sampling points. The ratio of ^{134}Cs activity to ^{137}Cs activity in lichens was corrected to that of the date on 11 March 2011.

2.5. Calculation for the aggregated transfer factor (T_{ag}) of ^{137}Cs in soil-to-lichen

The aggregated transfer factor (T_{ag}) is defined as the ratio of the radionuclide activity concentration in plants or any other natural or semi-natural product (Bq kg^{-1} fresh or dry weight, depending on the product) divided by the total deposition density on the soil (Bq m^{-2}) (IAEA, 2009). The T_{ag}'s for ^{137}Cs in soil-to-lichen were calculated for each specimen as the ratio of ^{137}Cs activity concentration in lichen divided by the ^{137}Cs inventory in soil (after decay correction to the date of the final sampling of lichens).

3. Results

3.1. ^{137}Cs deposition density on soil and air dose rate

The ^{137}Cs deposition density on soil at the investigated sampling points was estimated to be between 45.8 kBq m^{-2} (Point 12) and 2920 kBq m^{-2} (Point 10), the values of which were decay-corrected to that of the final sampling date of lichens (5 February 2013). The measured air dose rate at each of the sampling locations ranged from 0.2 μSv h^{-1} (Point 12 and 14) to 20.8 μSv h^{-1} (Point 11). The values of air dose rate at Points 11, 12, 13 and 14 were not used as there was snow cover when the measurements were made

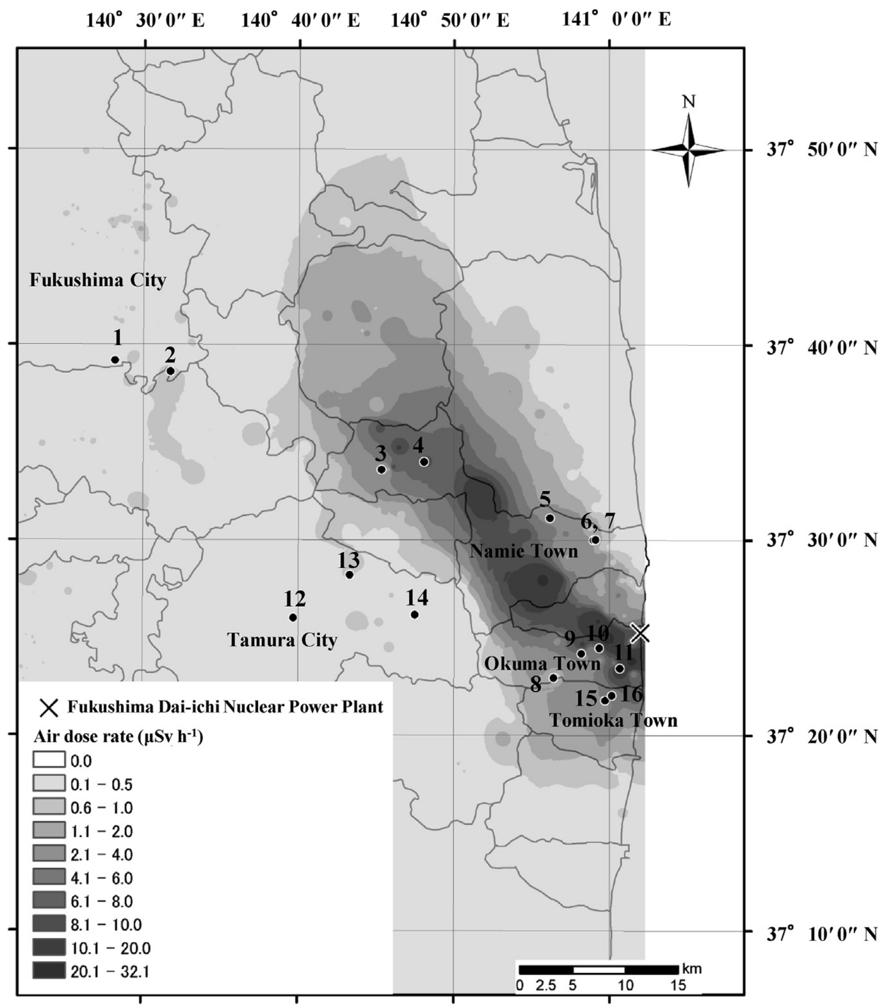

Fig. 1. Locations of lichen sampling points superimposed an air dose rate map measured by MEXT in June and July 2011 (MEXT, 2011a and 2011b).

(Table 2). Spearman's rank correlation coefficient (r) was calculated for the relationship between air dose rate and the ^{137}Cs deposition density on soil at each of the lichen sampling points (Fig. 2). These data sets ($n = 13$) show a strong positive correlation ($r = 0.92$, $P < 0.01$).

3.2. Lichen species

Collected samples of parmelioid lichens were examined and identified as the following nine species: *Canoparmelia aptata* (Kremp.) Elix & Hale, *Flavoparmelia caperata* (L.) Hale, *Myelochroa leucotyliza* (Nyl.) Elix & Hale, *Parmotrema austrosinense* (Zahlbr.) Hale, *P. clavuliferum* (Räsänen) Streimann, *P. reticulatum* (Taylor) M. Choisy, *P. tinctorum* (Nyl.) Hale, *Punctelia borreri* (Sm.) Krog, and *P. rudecta* (Ach.) Krog.

Among them, the common species were *Flavoparmelia caperata* (collected from 12 of 16 sampling points), followed by *Parmotrema clavuliferum* (11 points), and *P. tinctorum* (7 points). Other species were collected at only a few points (4–1 points) (Table 1).

3.3. Radiocaesium (^{134}Cs and ^{137}Cs) activity concentration in lichens

The activity concentration of ^{134}Cs and ^{137}Cs in the collected parmelioid lichens (total 44 samples) are shown in Table 2. Activity concentrations ranged from 4.6 ± 1.5 to 1000 ± 20 kBq kg^{-1} for ^{134}Cs and from 7.6 ± 1.6 to 1740 ± 20 kBq kg^{-1} for ^{137}Cs. The highest radiocaesium activity concentration was detected in *Parmotrema clavuliferum* collected at Point 11 which was located ca. 4 km southwest from the FDNPP. The lowest activity concentration was detected in *P. reticulatum* at Point 12 which was located approximately 33 km west from the FDNPP (Table 1 and Fig. 1). The radiocaesium activity concentration in the lichen samples generally increased with increase in the ^{137}Cs inventory in soil estimated by IDW.

The ratio of maximum and minimum ^{137}Cs activity concentrations in lichens collected at the same sampling location ($n \geq 3$) varied from 1.2 to 3.3. For example, the minimum ^{137}Cs activity concentration, 475 ± 11 kBq kg^{-1}, was observed in *Flavoparmelia caperata* and the maximum, 562 ± 12 kBq kg^{-1}, in *Parmotrema clavuliferum* at Point 4, while the minimum, 202 ± 7 kBq kg^{-1} in *P. tinctorum* and the maximum, 668 ± 14 kBq kg^{-1} in *P. clavuliferum* at Point 16.

The ^{134}Cs/^{137}Cs activity ratio of lichens corrected to the date, 11 March 2011 were in the range from 0.969 to 1.10 (Table 2). The mean ± standard deviation for the ratio obtained was 1.03 ± 0.03.

3.4. The aggregated transfer factor (T_{ag}) of ^{137}Cs in soil-to-lichen

The aggregated transfer factors (T_{ag}) for soil to lichen ($n = 44$) ranged from 0.12 to 0.99. The fundamental statistic of T_{ag}, i.e., median, mean, standard deviation (SD), and coefficient of variation

Table 1
Information of lichen samples.

Sampling point	Latitude	Longitude	Distance and direction from the FDNPP	Elevation (m)	Habitat	Sampling date	Species[a]	Specimen voucher	Dry weight[b] (g)
1. Matsukawamachi, Fukushima City	37° 39′ 11.27″ N	140° 28′ 10.19″ E	56 km NW	200	Open place with a clump of *Prunus* sp. in the park	17 Dec 2012	CA	TD 86	4.06
							FC	TD 90	2.45
2. Iinomachi, Fukushima City	37° 38′ 37.06″ N	140° 31′ 44.32″ E	51 km NW	180	Open place with scattered *Prunus* sp. along the lakeside	17 Dec 2012	CA	TD 84	3.76
							FC	TD 82	2.65
							ML	TD 85	4.59
							PC	TD 81	2.26
							PT	TD 78	1.97
							PuR	TD 83	4.84
3. Shimotsushima, Namie Town	37° 33′ 37.70″ N	140° 45′ 22.40″ E	29 km NW	430	Open place with scattered *Prunus* on the school grounds	20 Dec 2012	FC	TD101	2.41
							PC	TD102	2.02
4. Akougi, Namie Town	37° 34′ 00.90″ N	140° 48′ 05.20″ E	26 km NW	360	Open place with scattered *Prunus* sp. and a deciduous tree on a public square	20 Dec 2012	FC	TD106	2.32
							PC	TD107	2.14
							PR	TD108	2.59
5. Tatsuno, Namie Town	37° 31′ 07.72″ N	140° 56′ 13.24″ E	14 km NW	60	Open place with scattered *Prunus* sp. along a road	23 Jan 2013	PA	TD127	1.74
							PC	TD128	2.00
							PT	TD126	2.69
6. Sakata, Namie Town	37° 30′ 01.74″ N	140° 59′ 00.67″ E	10 km NW	10	Open place with scattered *Prunus* sp. along the river bank	23 Jan 2013	CA	TD122	4.79
							PT	TD118	2.19
7. Sakata, Namie Town	37° 30′ 02.14″ N	140° 59′ 08.86″ E	10 km NW	10	Open place with scattered *Prunus* sp. along the river bank	23 Jan 2013	FC	TD125	2.66
							PT	TD124	2.23
8. Oogawara, Okuma Town	37° 22′ 58.76″ N	140° 56′ 26.76″ E	9 km SW	140	Open place with scattered *Prunus* sp. along the lakeside	24 Jan 2013	FC	TD131	3.14
							PC	TD130	2.32
9. Shimonogami, Okuma Town	37° 24′ 11.03″ N	140° 58′ 13.10″ E	6 km W	70	Open place with scattered *Prunus* sp. on the school ground	5 Feb 2013	PA	TD144	1.74
10. Ottozawa, Okuma Town	37° 24′ 28.76″ N	140° 59′ 22.54″ E	4 km SW	70	Open place with scattered *Prunus* sp. on the school ground	5 Feb 2013	FC	TD146	3.57
							PA	TD145	1.86
11. Kumagawa, Okuma Town	37° 23′ 26.70″ N	141° 00′ 40.76″ E	4 km SW	40	Open place with scattered *Prunus* sp. on the school grounds	24 Jan 2013	PC	TD133	1.99
							PT	TD132	2.49
12. Tokiwa, Tamura City	37° 26′ 01.89″ N	140° 39′ 41.84″ E	33 km W	450	Open place with scattered *Prunus* sp. along the river bank	28 Jan 2013	FC	TD136	2.86
							PC	TD137	2.46
							PR	TD138	2.47
							PuB	TD139	2.89
13. Iwaisawa, Tamura City	37° 28′ 14.71″ N	140° 43′ 20.30″ E	28 km W	510	Open place with scattered *Prunus* sp. in the park	29 Jan 2013	FC	TD141	2.95
							PC	TD142	2.60
							PuR	TD143	4.41
14. Furumichi, Tamura City	37° 26′ 12.50″ N	140° 47′ 31.61″ E	21 km W	400	Open place with scattered *Prunus* sp. on the school grounds	28 Jan 2013	FC	TD134	3.26
							PC	TD135	2.40
15. Yonomori, Tomioka Town	37° 21′ 48.50″ N	140° 59′ 44.70″ E	7 km SW	50	Open place with scattered *Prunus* sp. along a road	18 Dec 2012	FC	TD 95	3.29
							PA	TD 99	1.98
							PC	TD 97	2.38
							PT	TD 98	2.07
							PuB	TD 96	3.51
16. Yonomori, Tomioka Town	37° 22′ 03.85″ N	141° 00′ 10.88″ E	6 km S	50	Open place with scattered *Prunus* sp. along a road	5 Feb 2013	FC	TD147	2.61
							PC	TD149	1.94
							PT	TD148	2.21

[a] CA = *Canoparmelia aptata*, FC = *Flavoparmelia caperata*, ML = *Myelochroa leucotyliza*, PA = *Parmotrema austrosinense*, PC = *Parmotrema clavuliferum*, PR = *Parmotrema reticulatum*, PT = *Parmotrema tinctorum*, PuB = *Puncteria borreri*, PuR = *Punctelia rudecta*.
[b] The weight precision is ±0.01 g.

(CV), are shown in Table 3. The median and mean values of T_{ag} in whole lichen samples were 0.37 and 0.40, respectively. The widest range between minimum and maximum T_{ag} values (from 0.12 to 0.99) was found in *Flavoparmelia caperata*, and the CV value was 85%. In contrast, the narrowest range (from 0.23 to 0.88) of T_{ag} was shown in *Parmotrema clavuliferum*, and the CV value was 38%.

Statistical analysis using the Steel–Dwass test showed that the difference of T_{ag} among the nine parmelioid lichen species was not significant. However, among the three common species, *Flavoparmelia caperata*, *Parmotrema clavuliferum* and *P. tinctorum*, the T_{ag} in *P. clavuliferum* was significantly higher than that in *Flavoparmelia caperata* ($P < 0.01$), and a similar result was also obtained by the Mann–Whitney's *U* test ($P < 0.01$).

3.5. Lichens and fallout

The radiocaesium activity concentration in lichens were compared to the estimated deposition in soil and measured air dose rates (which could be considered as a proxy for the radioactive fallout). The Shapiro–Wilk test ($P < 0.05$) revealed that ^{137}Cs activity concentration in lichen samples followed neither a normal nor a log-normal distribution. To examine the relationships

Table 2
Radiocaesium concentrations (kBq kg^{-1} dry weight, ± counting error) of lichen samples.

Sampling point	Species[a]	Activity concentration (kBq kg^{-1})		$^{134}Cs/^{137}Cs$[b]	Measured air dose rate (μSv h^{-1})	^{137}Cs deposition density on soil (kBq m^{-2})
		^{134}Cs	^{137}Cs			
1	CA	32.0 ± 2.5	55.1 ± 2.8	1.05 ± 0.10	0.7	133
	FC	22 ± 3	38.3 ± 3.1	1.06 ± 0.16		
2	CA	27.5 ± 2.4	48.9 ± 2.8	1.02 ± 0.11	0.8	176
	FC	19 ± 2	33.8 ± 2.8	1.01 ± 0.15		
	ML	42.8 ± 2.7	74.1 ± 3.1	1.05 ± 0.08		
	PC	57.1 ± 4.4	102 ± 5	1.02 ± 0.09		
	PT	47.4 ± 4.3	84.4 ± 5.1	1.02 ± 0.09		
	PuR	23.9 ± 2.0	43.7 ± 2.3	0.995 ± 0.097		
3	FC	157 ± 7	285 ± 8	0.999 ± 0.053	4.3	1560
	PC	352 ± 11	634 ± 13	1.01 ± 0.04		
4	FC	274 ± 9	475 ± 11	1.05 ± 0.04	8.8[c]	2410
	PC	318 ± 11	562 ± 12	1.03 ± 0.04		
	PR	316 ± 10	550 ± 11	1.05 ± 0.04		
5	PA	219 ± 10	406 ± 12	0.981 ± 0.052	5.2	1040
	PC	206 ± 9	374 ± 10	0.998 ± 0.051		
	PT	256 ± 8	479 ± 10	0.971 ± 0.038		
6	CA	176 ± 5	328 ± 6	0.972 ± 0.035	2.4	419
	PT	209 ± 8	379 ± 10	1.00 ± 0.05		
7	FC	48.9 ± 3.8	91.7 ± 4.5	0.969 ± 0.089	1.5	462
	PT	87.8 ± 5.5	162 ± 7	0.985 ± 0.073		
8	FC	54.1 ± 3.7	92.4 ± 4.2	1.06 ± 0.09	2.1	780
	PC	273 ± 10	471 ± 11	1.06 ± 0.04	1.6	
9	PA	697 ± 18	1210 ± 20	1.04 ± 0.03	9.3	1690
10	FC	390 ± 9	680 ± 10	1.04 ± 0.03	11.0	2920
	PA	517 ± 15	901 ± 17	1.04 ± 0.04		
11	PC	1000 ± 20	1740 ± 20	1.05 ± 0.02	(20.5)[d]	1980
	PT	645 ± 14	1130 ± 20	1.04 ± 0.03	(20.8)[d]	
12	FC	5.8 ± 1.5	9.9 ± 1.6	1.06 ± 0.32	(0.2)[d]	45.8
	PC	8.1 ± 1.8	14 ± 2	1.05 ± 0.28		
	PR	4.6 ± 1.5	7.6 ± 1.6	1.10 ± 0.42		
	PuB	9.4 ± 1.8	16 ± 2	1.06 ± 0.23		
13	FC	13 ± 2	22.4 ± 2.2	1.03 ± 0.19	(0.5)[d]	128
	PC	36.1 ± 3.4	63.0 ± 3.8	1.04 ± 0.12		
	PuR	28.6 ± 2.3	49.4 ± 2.6	1.05 ± 0.10		
14	FC	17 ± 2	31.3 ± 2.4	1.00 ± 0.15	(0.2)[d]	81.8
	PC	17 ± 3	28.7 ± 2.8	1.08 ± 0.19		
15	FC	486 ± 10	873 ± 12	1.01 ± 0.03	7.5	881
	PA	200 ± 9	354 ± 10	1.03 ± 0.05		
	PC	220 ± 8	395 ± 10	1.01 ± 0.05		
	PT	244 ± 9	436 ± 11	1.02 ± 0.05		
	PuB	270 ± 8	486 ± 9	1.01 ± 0.03		
16	FC	120 ± 6	214 ± 7	1.02 ± 0.06	7.6	1450
	PC	374 ± 12	668 ± 14	1.02 ± 0.04		
	PT	113 ± 6	202 ± 7	1.02 ± 0.07		

The radiocaesium concentration in lichen samples and the ^{137}Cs deposition density on soils were decay corrected to the date of final sampling (5 Feb 2013).
The fundamental statistics (median, mean, standard deviation (SD) and coefficient of variation (CV)) of radiocaesium activity concentration in lichen samples were obtained and are as follows:
138 kBq kg^{-1} (median), 196 kBq kg^{-1} (mean), 216 kBq kg^{-1} (SD), 110% (CV) in ^{134}Cs activity concentration, and.
249 kBq kg^{-1} (median), 348 kBq kg^{-1} (mean), 378 kBq kg^{-1} (SD), 109% (CV) in ^{137}Cs activity concentration.
[a] See Table 1 for abbreviations.
[b] The ratios were obtained by decay-correcting the date to March 11, 2011.
[c] The data was used the indicated value of the monitoring post at the sampling point.
[d] The values showing in the parentheses were measured on snow-covered ground.

between radiocaesium activity concentration in our lichen samples to the ^{137}Cs deposition density on soil and air dose rate, Spearman's rank correlation coefficient (r) was calculated. Strong positive correlations were observed between ^{137}Cs activity concentration in the lichens and ^{137}Cs deposition density on soil ($r = 0.90$, $P < 0.01$), and between radiocaesium ($^{134}Cs + ^{137}Cs$) activity concentration in lichens and air dose rate ($r = 0.84$, $P < 0.01$). For a better understanding these relationships, simple regression analyses were performed (Figs. 3 and 4). The coefficients of determination (R^2) were obtained for ^{137}Cs deposition density on soil as 0.53 and for air dose rate as 0.60, respectively. The statistical significance of both regression equations were confirmed ($P < 0.001$). The coefficients r of relationship among three lichen species ($n \geq 6$) and ^{137}Cs deposition density on soil were 0.92 ($n = 12$, $P < 0.01$) in *Flavoparmelia caperata*, 0.90 ($n = 11$, $P < 0.01$) in *Parmotrema clavuliferum*, and 0.71 ($n = 7$, $P > 0.05$) in *P. tinctorum*, indicating that statistically significant positive correlations were observed for *Flavoparmelia caperata* and *Parmotrema clavuliferum*. The values r for the relationships with air dose rate were 0.85 ($n = 9$, $P < 0.01$) in *Flavoparmelia caperata*, 0.54 ($n = 7$, $P > 0.05$) in *Parmotrema clavuliferum* and 0.60 ($n = 6$, $P > 0.05$) in *P. tinctorum*, showing that a statistically significant positive correlation was observed only for *Flavoparmelia caperata*.

4. Discussion

4.1. Radiocaesium activity concentration in lichens

The activity concentration of ^{137}Cs in parmelioid lichens collected within 4–56 km of the FDNPP ranged from 7.6 to

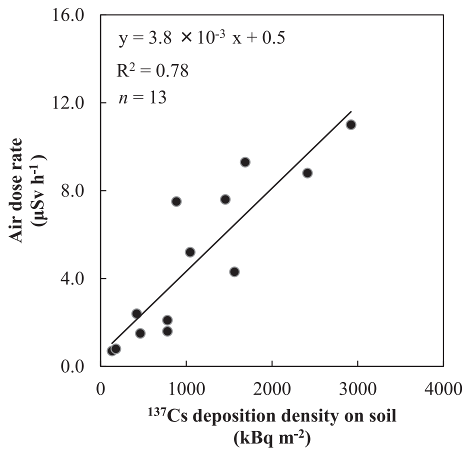

Fig. 2. The relationship between the air dose rate and the ^{137}Cs deposition density on soil at lichen sampling points (without snow cover). Air dose rate was measured at each sampling date. The ^{137}Cs deposition density was decay corrected to the date of each lichen sampling. The regression equation was obtained by simple linear regression analysis ($P < 0.001$). The slope and intercept values of the regression line were $3.8 \times 10^{-3} \pm 1.3 \times 10^{-3}$ and 0.5 ± 1.8, respectively (95% confidence interval). The Spearman's rank correlation coefficient was calculated as 0.92 ($P < 0.01$).

1740 kBq kg^{-1} (Tables 1 and 2). The maximum value of ^{137}Cs activity concentration measured in this study was 1740 ± 20 kBq kg^{-1} in *Parmotrema clavuliferum* that was collected ca. 4 km southwest of the FDNPP. This value is much higher than post-Fukushima reports: 35 kBq kg^{-1} (^{137}Cs) in *Physcia orientalis* collected in Tsukuba City (Ohmura et al., 2015), and 0.051 kBq kg^{-1} (^{137}Cs) in *Usnea diffracta* collected in Kunashiri Island (Ramzaev et al., 2014). In contrast, the maximum value detected in our study is an order of magnitude lower than those in foliose lichens collected from 1.5 km southwest of the Chernobyl NPP after the Chernobyl NPP accident which released approximately 85 PBq of ^{137}Cs (UNSCEAR, 2000), in which the activity concentrations were 14.6 MBq kg^{-1} in *Parmelia sulcata* Taylor and 5.84 MBq kg^{-1} in *Hypogymnia physodes* (Biazrov, 1994). But the value in this study is higher than those reported from other parts of Europe after the Chernobyl NPP accident: 61.2 kBq kg^{-1} in *H. physodes* collected in Austria and 26.7 kBq kg^{-1} in *Xanthoparmelia somloënsis* (Gyeln.) Hale collected in northern Greece (Sawidis et al., 1997; Heinrich et al., 1999). The ^{137}Cs activity concentration measured in lichens affected by atmospheric nuclear weapon tests were lower than that in our study (i.e., 2.3 kBq kg^{-1} in *Nephroma arcticum* (L.) Torss. from Finland, 0.173 kBq kg^{-1} in *Umbilicaria cylindrica* (L.) Delise from Poland, and 1.33 kBq kg^{-1} in *Hypogymnia physodes* from Austria) (Salo and Miettinen, 1964; Kwapuliński et al., 1985; Eckl et al., 1984).

4.2. Relationships between radiocaesium activity concentration in lichens and ground deposition/air dose rate

Significant positive correlations were obtained between ^{137}Cs activity concentration in the examined lichens and ^{137}Cs deposition density on soil (Fig. 3), and between total radiocaesium activity concentration (^{134}Cs + ^{137}Cs) in lichens and air dose rate (Fig. 4). The analyses of the two dominant species, *Flavoparmelia caperata* and *Parmotrema clavuliferum*, showed significant positive correlations regarding the ^{137}Cs deposition density on soil. A positive correlation between the concentrations of radionuclides in epiphytic parmelioid lichen and atmospheric deposition was also reported in *Parmelia sulcata* (Sloof and Wolterbeek, 1992), and our results are consistent with this study. The Spearman's rank correlation coefficient (r) for total radiocaesium activity concentration (^{134}Cs + ^{137}Cs) in lichens and air dose rate ($r = 0.84$), measured at the sampling 2 y after from the FDNPP accident was lower than the r value for the relationship between ^{137}Cs in lichens and ^{137}Cs inventory in soil ($r = 0.90$) collected in June and July 2011 (Figs. 3 and 4). Similarly, the r values for the relationship between total radiocaesium activity and air dose rate were lower than those for the ^{137}Cs inventory in soil obtained for the three species, *Flavoparmelia caperata*, *Parmotrema clavuliferum* and *P. tinctorum*. Ohmura et al. (2015) also found a temporal change in the coefficient of determination (R^2) value for the relationship between air dose rate and the radiocaesium activity concentration in *Physcia orientalis* sampled 1 y after the accident ($R^2 = 0.80$) and again 2 y after the accident ($R^2 = 0.65$). The reduction of coefficients, r and R^2, can be attributed to the changes of the radiocaesium contaminated situation surrounding the sampling points that might be caused by not only radioactive decay but also the weathering effect (Mikami et al., 2015a). Thus, even though the weathering was ongoing in the surrounding environment, *Flavoparmelia caperata* and *Parmotrema clavuliferum* could well keep radiocaesium in the thalli at least 2 y after from the accident.

4.3. The interspecific differences of T_{ag} among the parmelioid lichens

The T_{ag} in soil-to-lichen, i.e. the ratio of ^{137}Cs activity concentration in lichen against ^{137}Cs deposition density on soil, can

Table 3
The soil-to-lichen aggregated transfer factor (T_{ag}) for the Fukushima derived ^{137}Cs (m^2 kg^{-1}).

Species[a]	n	T_{ag}				
		Range	Median	Mean	Standard deviation	Coefficient of variation (%)
FC	12	0.12–0.99	0.20	0.28	0.24	85
PC	11	0.23–0.88	0.45	0.47	0.18	38
PT	7	0.14–0.90	0.48	0.49	0.23	48
PA	4	0.31–0.72	0.40	0.45	0.18	40
CA	3	0.28–0.78	0.41	0.49	0.26	53
PR	2	0.17–0.23	0.20	0.20	NC[c]	NC[c]
PuB	2	0.35–0.55	0.45	0.45	NC[c]	NC[c]
PuR	2	0.25–0.39	0.32	0.32	NC[c]	NC[c]
ML	1	NA[b]	0.42	0.42	NC[c]	NC[c]
ALL	44	0.12–0.99	0.37	0.40	0.21	53

The ^{137}Cs deposition density on soil and ^{137}Cs activity concentration in lichen were decay corrected to the final sampling date (5 Feb 2013).
[a] See Table 1 for abbreviations.
[b] NA = not applicable.
[c] NC = not calculated.

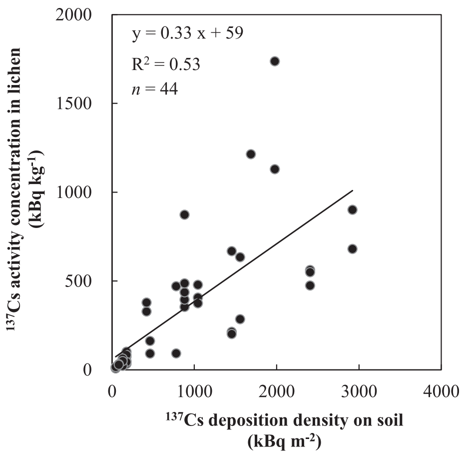

Fig. 3. The relationship between the ^{137}Cs activity concentration in all 44 lichen samples and the ^{137}Cs deposition density on soil at each of the lichen sampling points. The ^{137}Cs activity concentration in lichen samples and the ^{137}Cs deposition density were decay corrected to the date of the final sampling (5 Feb 2013). The regression equation was calculated by simple linear regression analysis ($P < 0.001$). The slope and intercept values of the regression line were 0.33 ± 0.09 and 59 ± 120, respectively (95% confidence interval). The Spearman's rank correlation coefficient was calculated as 0.90 ($P < 0.01$).

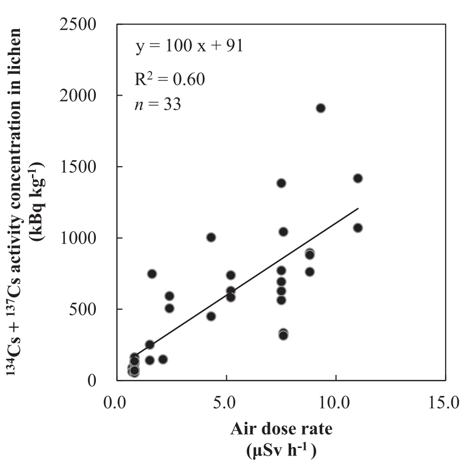

Fig. 4. The relationship between ^{134}Cs + ^{137}Cs activity concentration in lichen and the air dose rate at sampling points (without snow cover). The radiocaesium activity concentration in lichen was decay corrected to the date of each lichen sampling. Air dose rate was measured at each lichen sampling date. The regression equation was calculated by simple linear regression analysis ($P < 0.001$). The slope and intercept values of the regression line were 100 ± 30 and 91 ± 180, respectively (95% confidence interval). The Spearman's rank correlation coefficient was calculated as 0.84 ($P < 0.01$).

be adopted as a normalized radionuclide accumulation level at the sampling points (IAEA, 2009) (Table 3). Focusing on *Flavoparmelia caperata* and *Parmotrema clavuliferum*, the difference in T_{ag} values was significant based on the results of both the Steel–Dwass test and the Mann–Whitney's *U* test. The significant differences among *Cetraria* species and, among other lichen species grown at the same habitat were also reported in studies by Hanson (1971) and Sawidis et al. (1997). Such interspecific difference should be taken into account for the application as biomonitor in the future researches.

4.4. The ^{134}Cs/^{137}Cs activity ratios of lichen

The ratios of ^{134}Cs and ^{137}Cs for the lichen samples (decay-corrected to the date, 11 March 2011) ranged from 0.969 to 1.10. These ratios are consistent with those for radiocaesium released during the FDNPP accident (0.89–1.2) that emitted approximately 12 PBq of ^{137}Cs (Nuclear Emergency Response Headquarters Government of Japan, 2011; Fukushima Prefecture, 2012; Masson et al., 2011; Chino et al., 2011). The mean value for the ^{134}Cs/^{137}Cs activities ratio in our lichen samples, 1.03 ± 0.03 ($n = 44$), was compatible with both the mean values of 0.98 ± 0.05 ($n = 15$) for lichen collected from Tsukuba and 0.99 ± 0.10 ($n = 56$) for lichen collected from the Sakhalin region (Ohmura et al., 2013; Ramzaev et al., 2014). These agreements indicate that the lichens can keep radionuclides composition even they are collected from around or far from the FDNPP. Our mean value also revealed to be in agreement with other environmental samples (plants and soil) collected in the Fukushima Prefecture and around the FDNPP by Komori et al. (2013), which the value was 1.00 ± 0.02 ($n = 24$) taken. Therefore, spatial variations of the ^{134}Cs/^{137}Cs values in the lichens can be also found as well as these environmental samples, even the small differences, as a possible cause of multiple radioactive plumes from the FDNPP (Komori et al., 2013; Mikami et al., 2015b).

4.5. The applicability of lichens as biomonitors and/or bioindicators of radiocaesium fallout

Our study suggests that the nine parmelioid lichens show potential for use as biomonitors of radiocaesium fallout because their radiocaesium activity concentrations even at 2 y after the accident had still good relationship with ^{137}Cs inventory in soil measured quite soon after the accident. Among these lichen species, for long-term ^{137}Cs-monitoring around the FDNPP, *Flavoparmelia caperata* and *Parmotrema clavuliferum* could be especially useful species because of the following features: (1) wide distribution in the study area, (2) the longevity of the species, (3) good accumulators of radiocaesium, (4) the significant positive correlation between radiocaesium activity concentration in these lichens and fallout (radiocaesium inventory in soil), and (5) ease of collection and relative inexpensive of sampling and analysis. These features fundamentally satisfy the criteria for the selection of biomonitoring lichen species (Shukla et al., 2014). Since the applicability and usefulness of lichens in ^{137}Cs monitoring have been proven for more than a decade after the Chernobyl NPP accident (e.g. Sawidis et al., 2010; Cevik and Celik, 2009; Kirchner and Daillant, 2002), the long-term monitoring of ^{137}Cs around the FDNPP using *Flavoparmelia caperata*, *Parmotrema clavuliferum* and/or other species should be a useful means by which to study the dynamics of ^{137}Cs in environmental ecosystems (e.g. weathering and transport behavior).

The total amount of radiocaesium fallout is usually estimated by the radiocaesium deposition density on soil. However, deposited radiocaesium was considered to be penetrated shortly into the topsoil (Schimmack et al., 1989; Koarashi et al., 2012). In addition, the depth profile of radiocaesium in soil varies with soil type and land-use conditions (e.g. croplands, grasslands, and forests) (Al-Masri, 2006; IAEA, 2006; Koarashi et al., 2012; Takahashi et al., 2015), meaning that depth profile is necessary for the accurate estimation of the total soil inventory. However, lichens (foliose and fruticose) can easily be stripped from substrata, and can be more quickly measured for radiocaesium activity concentration even if only a small quantity is available since lichens uptake radionuclides efficiently through their entire thallus. These characteristics may provide quick estimation of inventory. The coefficient of variation (CV) in T_{ag} of the parmelioid lichen samples was 53% (Table 3). One of the factors related to such variation might be differences in environmental conditions. The influence of growth position of lichens on a tree trunk was reported for *Pseudevernia furfuracea*, which showed an increase of ^{137}Cs activity concentration of lichen with increasing height on tree trunk (Heinrich et al., 1999). The variation of ^{137}Cs activity concentration of lichens growing on exposed rock surfaces with different facing directions were reported for *Umbilicaria hirsuta* (Sw. ex Westr.) Ach. and the ratios for ^{137}Cs were approximately 10:3:2 for the northeast, southeast and southwest, directions respectively (Seaward et al., 1988). The variation in radioactivities in lichens is expected to reduce to improve quantitatively of deposition density by choosing less disturbed samples. Therefore, the effect of environmental conditions needs further investigation.

The situation of radioactive contamination has been changed because of the different velocity of radionuclide reduction depending on the places (e.g. fast in urban area and slow in forests) (Saito, 2013, 2014). Behavior of radiocaesium in lichens growing above the ground, namely e.g. on tree trunk and rock, is expected to be unaffected by surface situation for long periods of time. Such lichens could be appropriate bioindicators of radioactive fallout amount.

Even though, the effective half-lives of radiocaesium in lichens is one of the factors which may influence the estimation of the amount of fallout as has been pointed out by Tuominen and Jaakkola (1973). The effective half-lives of radiocaesium in lichens are estimated to be between 2.7 and 10.1 y (Puhakainen et al., 2007; Iurian et al., 2011), although they may vary depending on species type or different environmental factors (Cevik and Celik, 2009). Furthermore, ^{137}Cs-leaching from the substrate (e.g. tree trunk) should be also taken into consideration since it may cause an increase of ^{137}Cs in lichens with time (Sloof and Wolterbeek, 1992). Further research of the half-lives of ^{137}Cs and effect of ^{137}Cs-leaching in *Flavoparmelia caperata* and *Parmotrema clavuliferum* would contribute for more accurate estimation of fallout in Fukushima.

5. Conclusion

The radiocaesium activity concentration in parmelioid lichen species collected around the FDNPP (within a 60 km radius) 2 y after the accident was investigated. The relationship between the activity concentration of ^{137}Cs in lichens and the ^{137}Cs inventory in soil shows there is a significant positive correlation. Focusing on the relationships for the two dominant species, *Flavoparmelia caperata* and *Parmotrema clavuliferum*, the ^{137}Cs accumulation level in *P. clavuliferum* is significantly higher than that in *Flavoparmelia caperata* shown by comparison of their aggregated transfer factor (T_{ag}).

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