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Activity concentrations of radionuclides in lichens following the Fukushima nuclear accident

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Abstract: The activity concentration of ^{131}I, ^{134}Cs and ^{137}Cs radionuclides in lichens was traced one and a half months after the Fukushima nuclear accident. The samples were collected in Tsukuba City, which is located *c.* 170 km south of the Fukushima Daiichi nuclear power plant (NPP). The activity concentrations differed depending on species and habitat. For example, the maximum activity concentration of ^{137}Cs was 22596 Bq kg^{-1} dry weight in *Physcia orientalis* (collected from the trunk of *Zelkova serrata* on 30 June 2011), and 1928 Bq kg^{-1} in *Hyperphyscia crocata* (from the trunk of *Quercus myrsinaefolia* collected on 8 March 2012). The activity concentration of ^{137}Cs in *Dirinaria applanata* and *Phaeophyscia spinellosa* growing on vertical habitats decreased by *c.* 50% within a year, indicating radionuclides might have been washed off by rain. The radionuclides were apparently derived from the Fukushima NPP accident because: 1) one specimen collected at the same place one year before the accident did not contain radionuclides, 2) high activity concentrations of radionuclides were detected after the accident, 3) ^{131}I, which has a short half-life of 8 days, was detected one and a half months after the accident, and 4) the ratio of ^{134}Cs/^{137}Cs in lichens was 0·90–0·98 on 26 April 2011, which is consistent with the values reported for radiocesium from the Fukushima NPP accident.

Key words: Cesium 134, Cesium 137, fallout, foliose lichens, Iodine 131, monitoring

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Introduction

After the Tōhoku earthquake and tsunami, and the following nuclear disaster at the Fukushima Daiichi Nuclear Power Plant (NPP) on 11 March 2011, high activity concentrations of radiocesium (^{134}Cs, ^{137}Cs), radioiodine (^{131}I) and other radionuclides were reported from eastern Japan. Measurements came mainly from the atmosphere, surface soil, water and vegetables (MEXT 2013). However, lichens have not yet been investigated after the Fukushima NPP accident. Lichens are known to accumulate radionuclides and to reflect the total amount of fallout, since 1) they have no root and accumulate radionuclides in a passive way through the entire thallus, 2) they spread on a wide variety of substrata such as tree bark, rock, soil and concrete, and 3) they are long-lived organisms (Nimis 1996; Thomas & Gates 1999; Seaward 2002).

This paper reports on the activity concentrations of radionuclides (^{131}I, ^{134}Cs and ^{137}Cs) in lichens collected in Tsukuba City (*c.* 170 km south of the Fukushima NPP) using a low background gamma-ray detector. For comparison, we also measured these radionuclides in a specimen of *Phaeophyscia spinellosa* Kashiw. collected at the same locality one year before the Fukushima NPP accident.

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Materials and Methods

Collection of lichen samples

Collection information is summarized in Table 1. Lichen samples were collected after the Fukushima NPP accident in Tsukuba City, Ibaraki, Japan (36°06′N,

TABLE 1. *Collection information for lichen samples*

Collection date	Species*	Habitat	Specimen voucher**	Dry weight (g)
19/02/2010	PS	On concrete balustrade walls (mixed sample of both horizontal and vertical surfaces)	YO6878	13·84
26/04/2011	DA	On trunk of *Zelkova serrata*	YO8192	16·92
	PS	On concrete balustrade walls (mixed sample of both horizontal and vertical surfaces)	YO8191	11·38
30/06/2011	PO	On trunk of *Zelkova serrata*	YO8804	16·80
	PS	On horizontal surface of concrete balustrade walls	YO8802	11·86
		On vertical surface of concrete balustrade walls	YO8803	14·76
26/10/2011	DA	On trunk of *Zelkova serrata*	YO8806	23·64
	HC	On trunk of *Quercus myrsinifolia*	YO8807	27·42
	PO	On trunk of *Zelkova serrata*	YO8808	20·27
	PS	On horizontal surface of concrete balustrade walls	YO8809	19·83
		On vertical surface of concrete balustrade walls	YO8810	19·83
08/03/2012	DA	On trunk of *Zelkova serrata*	YO8811	12·92
	HC	On trunk of *Quercus myrsinifolia*	YO8812	18·50
	PS	On horizontal surface of concrete balustrade walls	YO8813	20·21
		On vertical surface of concrete balustrade walls	YO8814	13·04

* DA = *Dirinaria applanata*, HC = *Hyperphyscia crocata*, PO = *Physcia orientalis*, PS = *Phaeophyscia spinellosa*.
** Number after YO = specimen number of Yoshihito Ohmura. All specimens are housed in TNS.

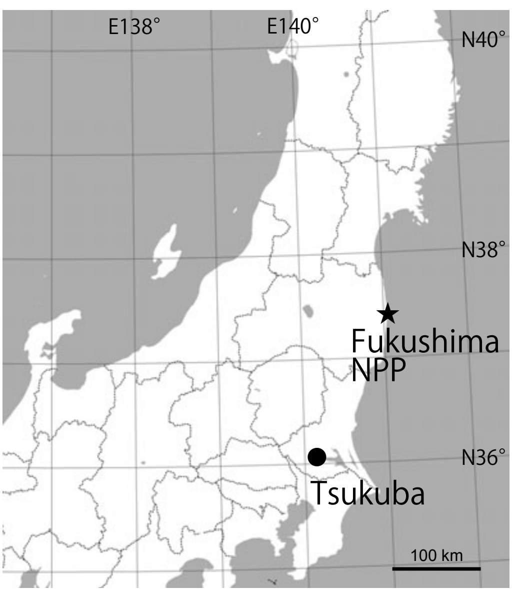

FIG. 1. Map of eastern Japan showing the locations of Tsukuba and the Fukushima NPP.

140°06′E; *c.* 25 m alt.) (Fig. 1) on 26 April, 30 June and 26 October 2011, and on 8 March 2012. A total of four lichen species from various habitats and substrata were investigated. The investigated species included *Dirinaria applanata* (Fée) D. D. Awasthi from trunks of *Zelkova serrata* (Thunb.) Makino, *Hyperphyscia crocata* Kashiw. from trunks of *Quercus myrsinifolia* Blume, *Physcia orientalis* Kashiw. from trunks of *Z. serrata*, and *Phaeophyscia spinellosa* from concrete balustrade walls. The samples of *P. spinellosa* from 26 April 2011 were mixed collections both from the horizontal and vertical surfaces of the balustrade walls, while on and after 30 June 2011, the samples were collected separately from the horizontal and vertical surfaces of the balustrade walls, respectively. A single specimen of *P. spinellosa* collected at the same locality on 19 February 2010, one year before the Fukushima NPP accident, was also examined. All voucher specimens are housed in the National Museum of Nature and Science, Tsukuba, Japan (TNS).

Radionuclide measurement

The lichen samples were dried at 40°C for 48 h and crushed by hand into small pieces *c.* 5 mm square. Cylindrical 100 ml plastic bottles (6·5 cm high, 5 cm diameter; U-8, AS ONE) were then filled with each crushed sample for radionuclide measurement. The dry weight of the samples is shown in Table 1. Radionuclide activity concentration (Bq kg^{-1}, dry weight) of ^{131}I, ^{134}Cs and ^{137} Cs was measured at the Center for Research in Isotopes and Environmental Dynamics, University of Tsukuba, using a low background gamma-ray detector for 2000 or 5000 s (IGC25190, Princeton Gamma Tech; MSA7800, SEIKO EG&G). The samples collected on 19 February 2010 and 26 April 2011 were measured after 2000 s, while the other samples were measured after 5000 s. The unique energies of the γ-rays (365 KeV for ^{131}I ; 604 KeV for ^{134}Cs; 661 KeV for ^{137}Cs) allowed identification and quantification of the radionuclides.

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TABLE 2. *Activity concentrations of radionuclides in lichens collected from Tsukuba City. Values in each cell show from the top activity concentrations (Bq kg^{-1}, dry weight) of ^{131}I, ^{134}Cs, ^{137}Cs, and the ratio of ^{134}Cs/^{137}Cs.*

	Collection date				
Species*	19/02/2010	26/04/2011	30/06/2011	26/10/2011	08/03/2012
DA	–	3706±111 3470±113 3836±139 0·90	–	ND 13578±107 17949±134 0·76	ND 6557±93 9394±125 0·70
HC	–	–	–	ND 948±27 1096±35 0·86	ND 1348±34 1928±45 0·70
PO	–	–	ND 19843±273 22596±373 0·88	ND 16961±129 19491±169 0·87	–
PS_h	–	–	ND 13787±267 15541±328 0·89	ND 10742±103 12243±135 0·88	ND 8017±85 11184±112 0·72
PS_v	–	–	ND 10209±236 11342±260 0·90	ND 6950±83 7989±108 0·87	ND 4014±76 5925±102 0·68
PS (mixture of horizontal and vertical samples. Showing mean value after 2011/6/30)	ND ND ND –	8873±217 13056±300 13296±293 0·98	ND 11998±252 13442±294 0·89	ND 8846±93 10116±122 0·87	ND 6016±81 8555±107 0·70

ND = not detected (below detection limit). *See Table 1 and Fig. 2 for the abbreviations.

Results

The radionuclide activity concentrations of ^{131}I, ^{134}Cs and ^{137}Cs in the lichens examined differed depending on species and habitat (Table 2). The maximum activity concentrations of ^{137}Cs measured were 22596±373 Bq kg^{-1} in *Physcia orientalis* (collected from the trunk of *Zelkova serrata* on 30 June 2011), 17949±134 Bq kg^{-1} in *Dirinaria applanata* (collected from the trunk of *Z. serrata* on 26 October 2011), 15541±328 Bq kg^{-1} in *Phaeophyscia spinellosa* (collected from the horizontal surface of concrete balustrade walls on 30 June 2011), 11342±260 Bq kg^{-1} in *P. spinellosa* (collected from the vertical surface of concrete balustrade walls on 30 June 2011), and 1928±45 Bq kg^{-1} in *Hyperphyscia crocata* (collected from the trunk of *Quercus myrsinaefolia* on 8 March 2012).

Phaeophyscia spinellosa collected at the monitoring site on 19 February 2010 (*c.* one year before the Fukushima NPP accident) showed no measurable radionuclide activities, while after the NPP accident, high activity concentrations of ^{131}I, ^{134}Cs and ^{137}Cs were detected for this species. ^{131}I was detected in *D. applanata* and *P. spinellosa* collected on 26 April 2011, but not in samples collected on and after 30 June 2011. The activity concentrations of ^{134}Cs and ^{137}Cs were almost equal in all samples on 26 April 2011: 3470±113 and 3836±139 Bq kg^{-1} in *D. applanata* (^{134}Cs/^{137}Cs = 0·90), and 13056±300 and 13296±293 Bq kg^{-1} in *P. spinellosa* (^{134}Cs/^{137}Cs = 0·98). The activity concentrations of ^{134}Cs decreased with time and became much lower than those of ^{137}Cs for all samples by 8 March 2012, on which date the ratio of ^{134}Cs/^{137}Cs reached 0·68–

0·72 (i.e., *D. applanata*: 0·70; *H. crocata*: 0·70; *P. spinellosa* on horizontal surface of balustrade walls: 0·72; *P. spinellosa* on vertical surface of balustrade walls: 0·68).

The activity concentration of radiocesium in *D. applanata* from the trunk of *Z. serrata* increased from 26 April to 26 October 2011, and then decreased to 8 March 2012, while that of *P. spinellosa* from balustrade walls stayed constant from 26 April to 30 June 2011 and then decreased. The activity concentrations of ^{137}Cs in samples of *P. spinellosa* from vertical surfaces of balustrade walls were 73% lower than in those from horizontal surfaces on 30 June 2011, 65% lower on 26 October 2011, and 53% lower on 8 March 2012.

Discussion

Radionuclides in lichens collected at Tsukuba City after the Fukushima NPP accident at the investigated sites were apparently derived from the NPP because 1) one specimen collected at Tsukuba one year before the accident did not contain detectable radionuclides, 2) high activity concentrations of radionuclides were detected after the accident, 3) ^{131}I, which has a short half-life of 8 days, was detected one and a half months after the accident, and 4) the ratio of ^{134}Cs/^{137}Cs in lichens was 0·90–0·98 on 26 April 2011, which is consistent with values reported for radiocesium derived from the Fukushima NPP accident [e.g. 0·8–0·9 in Kinoshita *et al.* (2011); 0·9 in Masson *et al.* (2011); 1·0 in Yamamoto *et al.* (2012)]. In comparison, the ratio of ^{134}Cs/^{137}Cs in the Chernobyl fallout in 1986 was reported at 0·5–0·6 (Arvela *et al.* 1990; De Cort *et al.* 1998).

The radionuclide activity concentrations in the lichens examined were much higher (e.g., 22596 ± 373 Bq kg^{-1} for ^{137}Cs in *Physcia orientalis*) than the values reported in mushrooms, plants and soil collected in Tsukuba City, among which the highest value was 335 Bq kg^{-1} for ^{137}Cs in a soil sample (Tsukuba City 2013). The capacity of lichens for accumulating radionuclides from atmospheric deposition has been well documented, particularly for 'Reindeer Moss' (*Cladonia* spp.), since the period of maximum atmospheric nuclear weapons fallout in the early 1960s (Nimis 1996; Seaward 2002). The main reasons why lichens accumulate high activity concentrations of radionuclides are their lack of roots, large surface area, and longevity (Thomas & Gates 1999). In addition, sampling design may have caused the differences in activity concentration of radionuclides. While the lichens in this study were only 2–3 mm thick and grew exposed on the surface of the substrata, the soil samples, for example, were collected at a depth of 5 cm below the soil surface (Tsukuba City 2013).

The activity concentrations of radionuclides were different depending on species and habitat. The four species examined in this study are similar in gross morphology, all being foliose and adnate to the substratum. The radiocesium activity concentrations in *Hyperphyscia crocata* growing on the trunk of *Quercus myrsinaefolia*, an evergreen tree, were much lower than those in the lichen taxa growing on concrete balustrade walls or trunks of *Zelkova serrata*, a deciduous tree. However, it is uncertain whether the differences in radiocesium activity concentration were due to differences in lichen species or habitat. *Phaeophyscia spinellosa* had different activity concentrations of radiocesium when growing on horizontal or vertical substratum surfaces (Table 2). A similar difference in activity concentrations of radiocesium was also reported for *Hypogymnia physodes* (L.) Nyl. after the Chernobyl accident (Guillitte *et al.* 1994). The higher values in the samples from horizontal surfaces are consistent with the general notion that lichens reflect the total amount of fallout (Nimis 1996), as lichens growing on horizontal substrata present a larger surface area available for fallout accumulation, compared to lichens from vertical substrata. The trends of radiocesium activity concentrations in *Dirinaria applanata* were distinctive; that is, they first increased and then decreased. The samples were collected from the trunks of the deciduous tree *Zelkova serrata*, bearing no leaves at the time of the accident, and, because of this, may have been accumulated in the lichen by stem flow from the branches and twigs during the initial fallout. Later, after the radionuclides had been blown off the branches and twigs, the radiocesium activity concentrations in *D. applanata* might have decreased, probably by being washed out from the lichen thalli. The other species investigated in this study might be less affected by radionuclide accumulation by run-off water as they either grow on low balustrade walls, or on the trunk of an evergreen tree which might accumulate the radionuclides on their leaves (Hashimoto *et al.* 2012) and thus shelter the lichens.

Although lichens still hold high activity concentrations of radionuclides one year after the Fukushima NPP accident (maximum 11184 Bq kg^{-1} of ^{137}Cs on 8 March 2012), the activity concentration of ^{137}Cs drastically decreased (*c.* 50%) in *D. applanata* and *P. spinellosa*, especially in samples growing on vertical habitats. Since the half-life of ^{137}Cs is *c.* 30 years, the loss of radiocesium from lichens might be caused by washing off by rain during the first year after the NPP accident. An exponential loss of ^{137}Cs was reported for *Cladonia stellaris* (Opiz) Pouzar & Vĕzda (Puhakainen *et al.* 2007), but ^{137}Cs has still been detected in this species 18 years after the Chernobyl accident (e.g. 94400 Bq kg^{-1} in 1987 and 1560 Bq kg^{-1} in 2004). A similar decrease in ^{137}Cs concentration in lichens might be observed in the polluted areas around the Fukushima NPP in the future, and long-term monitoring of lichens in Japan should be conducted for further data collection on the Chernobyl and Fukushima NPP accidents.

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