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Radiocesium contamination of the moss *Hypnum plumaeforme* caused by the Fukushima Dai-ichi Nuclear Power Plant accident

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ABSTRACT

We investigated ^{134}Cs and ^{137}Cs activity concentrations in the common Japanese moss species *Hypnum plumaeforme* collected from 32 sites within ca. 100 km radius of the Fukushima Dai-ichi Nuclear Power Plant. A total of 32 samples of *H. plumaeforme* were collected during the field surveys from November 2013 to September 2014. The maximum radiocesium activity concentrations in *H. plumaeforme* were 60.9 ± 1.8 kBq kg^{-1} for ^{134}Cs and 123 ± 2.3 kBq kg^{-1} for ^{137}Cs. The mean value for the ^{134}Cs/^{137}Cs was 1.17 ± 0.05, and the mean T_{ag} value was 0.09 ± 0.13. Positive correlations were obtained between total ^{134}Cs + ^{137}Cs activity concentrations in *H. plumaeforme* and the air dose rate with a correlation coefficient (r) of 0.55 ($P = 0.001$), and between ^{137}Cs activity concentration in *H. plumaeforme* and ^{137}Cs deposition density on soil with r of 0.55 ($P = 0.001$). These results suggest that the perennial moss species *H. plumaeforme* could be more suitable and useful as a qualitative indicator for the radiocesium pollution compared to vascular plants spreading over the lowlands including human habitation in Fukushima Prefecture.

1. Introduction

The Great East Japan Earthquake occurred on March 11, 2011, at a magnitude of 9.0. After the earthquake, the Fukushima Dai¨ichi Nuclear Power Plant (FDNPP) was struck by a huge tsunami that reached a height of > 13 m, which destroyed the FDNPP diesel generators and cooling systems. Following three successive hydrogen explosions at the buildings of Units 1, 3, and 4, approximately 520 (340–800) PBq of radioactive material was released into the atmosphere (Steinhauser et al., 2014). The releases of atmospheric radionuclides ^{134}Cs and ^{137}Cs was estimated to be 8–53.1 PBq (Chino et al., 2011; Masson et al., 2011; Stohl et al., 2012; Sugimoto, 2014), and ^{137}Cs was found to be widely dispersed throughout Fukushima and its adjacent prefectures in northeastern Japan. Most of the land within this region is covered with forest vegetation, including 71% of all land in the Fukushima Prefecture (Forestry Agency: http://www.rinya.maff.go.jp/j/keikaku/genkyou/h24/1.html), and because ^{137}Cs has a half-life of 30 years, decontamination of radiocesium in the environment is a crucial issue in Japan.

Bryophytes are non-vascular plants that form an important component of forest ecosystems, growing on various substrata (e.g., humus, soil, boulders, fallen logs, and tree trunks) and in various habitats (e.g., road banks, open pine forest floors, and open grasslands). Bryophytes have been used as bioindicators of air and water pollution (Gilbert, 1968; Kelly and Whitton, 1989; McLean and Jones, 1975; Vanderpoorten, 1999), heavy metal pollution (Berg et al., 1995; Harmens et al., 2010; Herpin et al., 1996; Lee et al., 2005; Markert et al., 1996; Onianwa, 2001), and radiocesium pollution (Dragović et al., 2004; Giovani et al., 1994; Livens et al., 1991; Papastefanou et al., 1989) within various environments. Unlike vascular plants, bryophytes absorb water and nutrients directly through their body surfaces, as they lack a water-repellent cuticle on their body surface and the water- and nutrient-conducting vascular systems in their body. Therefore, as bioindicators, bryophytes are generally more sensitive to pollutants than vascular plants.

After the FDNPP accident, Nabihah et al. (2016) reported radiocesium contamination in the moss *Hyophila propagulifera* Broth., which frequently grows on calcareous substrata, such as mortar walls at roadsides or in ditches. It forms tufts that are few centimeters tall, with upright and less frequently branched stems. Concurrently with Nabihah et al. (2016), we initiated investigations on the moss *Hypnum plumaeforme* Wilson. This species belongs to a different phylogenetic group from *H. propagulifera* and has a different growth form, namely, wefts, which are woven carpets with many branched stems that are prostrate on the substratum, often interwoven within herbs. These two moss species are very common and easily recognizable in lowland as well as montane areas, not only in Fukushima and its adjacent prefectures but

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also throughout southwest Japan. *H. plumaeforme* is widely distributed throughout the temperate zones of Japan, Taiwan, Korea, Russia Far East, China, Nepal, and the Philippines (Ando, 1958) and is one of the most common bryophytes in lowland Japan. It grows in various environments inhabited by humans, such as urban areas, and forms large mats on various substrata, such as soil, humus, boulders, and tree trunks in sunny and dry to semi-shaded, moderately moistened places, such as road banks, open meadows, or the edges or floors of pine forests.

In the present study, we investigated the spatial distribution of radiocesium using *H. plumaeforme* collected within ca. 100 km radius of the FDNPP approximately 2.8–3.6 years after the FDNPP accident. We then studied the relationships between radiocesium (^{134}Cs and ^{137}Cs) activity concentration and the air dose rate at each sampling site and between ^{137}Cs activity concentration and ^{137}Cs deposition density on soil. The air dose rate and ^{137}Cs deposition density on soil were obtained from open source data from the Ministry of Education, Culture, Sports, Science and Technology (MEXT; MEXT, 2011) using Global Positioning System (GPS) data for all sampling site locations.

2. Materials and methods

2.1. Sampling of plant materials

The sampling locations were selected within ca. 100 km radius of the FDNPP. A total of 32 samples of *H. plumaeforme* were collected during the field surveys in November 2013 and in May, July, August, and September 2014 (Table 1). The samples used in the present study were collected from plants growing on a road bank. Plant size was approximately 10 cm and stem leaves ranged mostly from 1.5 to 3 mm long and from 0.7 to 1 mm wide. Sampling site locations are depicted in Fig. 1 using GPS data obtained at sampling and ArcGIS software. The distance and direction of the sampling sites from the FDNPP were also calculated using ArcGIS software. Before radiocesium measurements, the plant samples collected in the field were dried at room temperature for several days to prevent spoiling. Voucher specimens were deposited at the herbarium of Hiroshima University, Hiroshima, Japan (HIRO).

2.2. Corresponding data sources of air dose rate and ^{137}Cs deposition on soil

The air dose rate (μSv h^{-1}) and ^{137}Cs deposition density on soil (kBq m^{-2}) at the sampling points were obtained from the website "Extension Site of Distribution Map of Radiation Dose, etc.", of the Ministry of Education, Culture, Sports, Science and Technology (MEXT, 2011) and are shown in Table 2. Data used in the present study were measured on July 2, 2011 in the third airborne monitoring survey by MEXT, except for sample codes 21, 22, and 23 of which data were measured on November 5, 2011 because of no survey results on July 2, 2011.

2.3. Sample treatment

Each moss sample was identified by checking key morphological characteristics according to Ando (1958) using light microscopy. To measure the absorbed radiocesium in the samples, first the soil was separated from the air-dried samples using a sieve, and other bryophytes and/or vascular plants were removed under a stereoscopic microscope using forceps. The samples were then washed with water to scour the remaining soil attached to the moss surface and dried at room temperature for 5 day. The dried samples were crushed into a fine

Table 1
List of samples investigated in the present study.

Sample code	Location	Elevation (m)	Collecting date (day/month/year)	Distance from the FDNPP (km)	Specimen voucher
1	Ohkura, Iitate-mura, Soma-gun, Fukushima	188.0	25/3/2014	39.7	HIRO1030303
2	Mano Dam, Ohkura, Iitate-mura, Soma-gun, Fukushima	196.1	25/3/2014	38.0	HIRO1030304
3	Mano Dam, Ohkura, Iitate-mura, Soma-gun, Fukushima	209.1	25/3/2014	38.7	HIRO1030305
4	Yamakami, Soma-shi, Fukushima	127.6	17/7/2014	42.2	HIRO1030306
5	Matsugafusa Dam, Yamakami, Soma-shi, Fukushima	412.9	26/3/2014	47.8	HIRO1030307
6	Koyasu Shrine, Furumichi, Miyakoji-machi, Tamura-shi, Fukushima	423.3	30/9/2014	18.4	HIRO1030308
7	Yamakiya, Kawamata-machi, Date-gun, Fukushima	625.1	6/9/2014	35.0	HIRO1030309
8	Shirane, Yanagawa-machi, Date-shi, Fukushima	154.9	18/7/2014	57.8	HIRO1030310
9	Ishida, Ryozen-machi, Date-shi, Fukushima	436.0	17/7/2014	48.2	HIRO1030311
10	Ohta, Nihonmatsu-shi, Fukushima	254.6	16/7/2014	47.6	HIRO1030312
11	Kaminagaori, Nihonmatsu-shi, Fukushima	228.9	7/9/2014	47.7	HIRO1030313
12	Watanoshiba, Iino-machi, Fukushima-shi, Fukushima	192.7	7/9/2014	51.1	HIRO1030314
13	Shinobuyama Park, Fukushima-shi, Fukushima	271.9	29/9/2014	63.5	HIRO1030315
14	Shinobuyama Park, Fukushima-shi, Fukushima	185.7	29/9/2014	63.3	HIRO1030316
15	Asakawa, Matsukawa-machi, Fukushima-shi, Fukushima	218.2	7/9/2014	58.6	HIRO1030317
16	Ochiai, Katsurao-mura, Futaba-gun, Fukushima	449.2	30/9/2014	24.3	HIRO1030318
17	Katsurao, Katsurao-mura, Futaba-gun, Fukushima	645.5	30/9/2014	26.7	HIRO1030319
18	Kamikawauchi, Kawauchi-mura, Futaba-gun, Fukushima	450.8	30/9/2014	21.6	HIRO1030320
19	Ohgaki Gam, Murohara, Namie-machi, Futaba-gun, Fukushima	154.7	17/7/2014	16.3	HIRO1030321
20	Hirusone, Namie-machi, Futaba-gun, Fukushima	217.9	30/9/2014	19.7	HIRO1030322
21	Takase, Namie-machi, Futaba-gun, Fukushima	14.3	12/11/2013	7.0	HIRO1030323
22	Ohsuge, Tomioka-machi, Futaba-gun, Fukushima	80.0	30/9/2014	6.6	HIRO1030324
23	Yamada, Futaba-machi, Futaba-gun, Fukushima	32.1	17/7/2014	3.3	HIRO1030325
24	Mimigai, Odaka-ku, Minamisoma-shi, Fukushima	2.5	17/7/2014	12.0	HIRO1030326
25	Takanokura Dam, Takanokura, Haramachi-ku, Minamisoma-shi, Fukushima	181.7	26/3/2014	26.6	HIRO1030327
26	Yokokawa Dam, Baba, Haramachi-ku, Minamisoma-shi, Fukushima	163.2	11/11/2013	23.0	HIRO1030328
27	Fukouno, Haramachi-ku, Minamisoma-shi, Fukushima	64.2	11/11/2013	28.6	HIRO1030329
28	Nukazawa, Motomiya-shi, Fukushima	244.3	7/9/2014	54.1	HIRO1030330
29	Nakakomatsu, Inawashiro-machi, Yama-gun, Fukushima	518.1	16/7/2014	81.7	HIRO1030331
30	Ohuchi, Marumori-machi, Igu-gun, Miyagi	67.4	18/7/2014	51.6	HIRO1030332
31	Kamitakihigashi, Marumori-machi, Igu-gun, Miyagi	71.9	18/7/2014	54.4	HIRO1030333
32	Sakunami, Aoba-ku, Sendai, Miyagi	228.3	30/8/2014	103.6	HIRO1030334