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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

Terumi Dohi [a, *], Yoshihito Ohmura [b], Hiroyuki Kashiwadani [a, b], Kenso Fujiwara [a], Yoshiaki Sakamoto [a], Kazuki Iijima [a]

[a] Sector of Fukushima Research and Development, Japan Atomic Energy Agency, 6-6 Sakae-machi, Fukushima, Fukushima 960-8031, Japan
[b] Department of Botany, National Museum of Nature and Science, 4-1-1 Amakubo, Tsukuba, Ibaraki 305-0005, Japan

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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

* Corresponding author.
E-mail address: dohi.terumi@jaea.go.jp (T. Dohi).

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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