Journal of Environmental Radioactivity 164 (2016) 291–299

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Radiocesium contamination in living and dead foliar parts of Japanese cedar during 2011–2015

Toshihiro Yoshihara*, Hideyuki Matsumura, Shin-nosuke Hashida, Ko Nakaya

Laboratory of Environmental Science, Central Research Institute of Electric Power Industry (CRIEPI), 1646 Abiko, Chiba 270-1194, Japan

A R T I C L E I N F O

Article history:
Received 17 February 2016
Received in revised form
27 July 2016
Accepted 7 August 2016

Keywords:
Radiocesium
Potassium
Fukushima
Japanese cedar
Translocation

A B S T R A C T

Radiocesium (^{137}Cs) activity concentrations, mainly derived from the Fukushima accident of March 2011, were measured in green foliar parts without separation by age (bulk green foliar parts; GL) and litterfall (LF) of Japanese cedar (*Cryptomeria japonica*) from 2011 to 2015. In all samples, ^{137}Cs concentrations decreased exponentially over time, but were always higher in LF (7.36–0.58 Bq g-DW^{-1}) than in GL (2.10–0.06 Bq g-DW^{-1}). The difference in the decreasing rate between GL and LF would reflect a difference in the dominant factor of the decrease between living and dead tissues (i.e., internal translocation and weathering, respectively). Over this same timeframe, potassium (K) concentrations in both GL and LF experienced repetitive periodical changes within a certain range (0.38–3.0 mg g-DW^{-1} for LF and 2.08–4.77 mg g-DW^{-1} for GL, respectively). Thus, there was no specific correlation between ^{137}Cs and K concentrations in LF and GL. However, analyses of the age classified green foliar parts (GL-S) and dead foliar parts still retained on trees (DL) could indicate another view. The annual changes in residual rates of both ^{137}Cs and K concentrations in GL-S demonstrated very similar two-phase reductions (i.e., a faster reduction in each expansion year than in the following years) and an obvious linear correlation between each other. Radiocesium concentration in DL were always higher than in any part of GL-S sampled at the same timing, but K concentrations showed the reverse relation. It is probable that ^{137}Cs is basically translocated from older parts to the developing parts (as long as the former are alive) via a seasonal nutritional flow of K; however, a part of ^{137}Cs translocation would cease considerably earlier than the cessation of K translocation.

1. Introduction

Forests effectively intercept aerosols (Bunzl and Kracke, 1988; Ould-Dada, 2002; Freer-Smith et al., 2005; Petroff et al., 2008); radionuclides in aerosols are also more effectively trapped by forests than by other land-use areas, particularly in cases where fallout occurs mainly in the form of dry deposition (Bunzl et al., 1989; Pröhl, 2009). Although fallout occurred mainly in the form of wet deposition in the case of the Fukushima Dai-ichi Nuclear Power Plant (FDNPP; located at 37.41868° N, 141.02215° E) accident, accumulation of radionuclides in forests was nevertheless observed (Amano et al., 2012; Katata et al., 2012; Kato et al., 2012a; Gonze et al., 2014). Forests extend over more than 70% of the Fukushima Prefecture (covering 975,000 ha of 1,378,000 ha); however, forest areas except vicinities of residential areas have been excluded from the governmental decontamination plan (MOE, 2012). Although this is understandable given the prioritization of economic and public health issue, it is critically important for people making living by forestry; concerns about the potential contamination of forest products, such as mushrooms, timber, and compost, are still being. Thus, at least, monitoring of the radionuclide deposition should be continued to expect the fate of radionuclide (Forestry Agency, 2014).

The widespread and serious contamination caused by radionuclides in the environment derived from the FDNPP accident has been documented in hundreds of papers (e.g., Yamada, 2012; Steinhauser et al., 2014; Evrard et al., 2015). Among those, a Chernobyl experience-based prediction suggested that a proportion of the radionuclides within tree parts of forests would disappear at most within 5 y (Hashimoto et al., 2013). In fact, many reports confirmed drastic reductions (e.g., of one-third of initial fallout per year) in the radiocesium inventory of the canopy within a few years following the FDNPP accident (Forestry Agency, 2014; Kato et al.,

* Corresponding author.
E-mail address: yoshiha@criepi.denken.or.jp (T. Yoshihara).

http://dx.doi.org/10.1016/j.jenvrad.2016.08.010
0265-931X/

2012a,b; Akama et al., 2013; Yoshihara et al., 2014b; Kajimoto et al., 2015). However, the reductions occur depending species and plant parts, and canopies should still retain a non-negligible level of contamination even 5 y after the fallout. Although statuses of leaf expansion at the fallout and the resulted receiving amount of radiocesium deposition mainly affect the specific rate of the reductions, another attribute in tree physiology (e.g., a radiocesium translocation activity within a tree body) would also contribute to induce at most three times variance in the reduction rates even if limited to only evergreen species (Yoshihara et al., 2014b; Kajimoto et al., 2015). On the other hand, the reduction in canopy contamination basically induces increase in forest floor contamination (Kato et al., 2012a; Kajimoto et al., 2015). It is noteworthy that litterfall derived organic matter inhibits the adsorption of radiocesium to clay minerals, and as a result affects the vertical distribution and thus the mobility of radiocesium, particularly at the surface soil layer of forest floor (Dumat et al., 2000; Koarashi et al., 2012). In addition, recent reports suggest that biotic lignin decomposition of litterfall and/or organic layers probably affects and complicates the fate of radiocesium on forest floors with a topographic heterogeneity (Hashida and Yoshihara, 2014, 2016; Koarashi et al., 2014). In other word, significant portion of radiocesium possibly continues to be mobile particularly in forest soils that are rich in organic matter relative to the amount of clay-sized particles (Koarashi et al., 2012). These may mean that biological recycling of ^{137}Cs will start with decomposition of organic materials in litter layers. Otherwise, these may relate to a possibility that deposited radiocesium discharges into watersheds through river systems and/or directly to areas adjacent to forests. In fact, high correlation can be seen between ^{137}Cs deposition and dissolved and particle ^{137}Cs concentration per unit weight of suspended substances in river water regardless of the river characteristics (Tsuji et al., 2014). Discharges of radiocesium from forests to river systems increase greatly following heavy rains, such as those generated by typhoons (Likuku, 2003; Nagao et al., 2013; Ueda et al., 2013; Evrard et al., 2015). Consequently, entire understanding of the fate of radionuclides in forests is important not only for the forest ecosystem, but also for economic and human health issues (Goor and Thiry, 2004; Kajimoto et al., 2015).

Here, we provide data showing specific changes in radiocesium concentrations between 2011 and 2015 in the green foliar parts and litterfall of Japanese cedar (*Cryptomeria japonica*) from two ways of sampling: the continuous bulk sampling and age classified sampling. The ever green Japanese cedar is the most popular commercial woody species in Japan, planted across 54% of plantation forests in the Fukushima Prefecture (Forestry Agency, 2014). Japanese cedar is one of the species with long foliar part longevity (4–6 y), comparable to Norway spruce (*Picea abies*) and Momi fir (*Abies firma*), although its foliar part life is dependent on growth conditions and site locations (Sakaguchi, 1983; Miyaura and Hozumi, 1993; Fujimoto and Kimura, 2011). In this study, we also measured concentrations of potassium, the metabolism of which is most closely related to cesium metabolism in trees as a biological analogue (Ronneau et al., 1991; Myttenaere et al., 1993; Goor and Thiry, 2004), for a comparison to the radiocesium distribution in trees.

2. Materials and methods

2.1. Study site and target trees

The study site is located in Abiko (35.87815° N, 140.02487° E, Laboratory of Environmental Science, CRIEPI, the total area of 17.3 ha), approximately 200 km SSW of the FDNPP. Initial radionuclide fallout in Abiko was observed on March 16, 2011, in the form of dry-deposition; however, the majority of the fallout was observed on March 21, 2011, occurring with rainfall. In total, 60–100 kBq m^{-2} of radiocesium (^{134}Cs, and ^{137}Cs) were recorded from Abiko following the latter deposition event (Morino et al., 2011; Terada et al., 2012; Doi et al., 2013). Further details of the location can be obtained in other literature (Yoshihara et al., 2013, 2014a,b). Sampled individuals of Japanese cedar (*Cryptomeria japonica*) were located at the margin of three small forest stands and grew on bare ground. The basal soil type was comprised of pale ando soils overlain with a thin organic layer (National Land Agency, 1983). Trees were assumed to be approximately 30 y of age, based on the timing of planting. At the start of the observation period, the heights of stands were 13.2–18.0 m, and diameters at breast height were 24–36 cm.

2.2. Sampling procedure and radionuclide analysis

Litterfall (LF) and living foliar parts (GL), both of which include needles and small twigs (mostly under 3-y-old) were sampled between September 5, 2011 and February 20, 2015. The LF sampling was generally carried out monthly, but the sampling program was adjusted over the period, based on the volume of LF. In total, 30 LF samples were obtained from each of three stands growing at least 20 m apart in a small forest comprised only of Japanese cedar. Radiocesium (^{137}Cs) and potassium concentrations were measured independently for each stand, as explained below. However, the possibility of some mixing of LF from targeted stand with LF from non-targeted neighboring stands can not be excluded. Two box-type traps, each measuring 1 × 1 × 1 m^3, were randomly placed under each target stand to collect LF before reaching the ground. The LF collected during the same sampling period by the two traps was mixed and treated as one sample. Sampling of GL was carried out at the same time as LF sampling. A total volume of approximately 0.5 kg-FW was harvested from at least two independent parts of the bottom of the tree crown; these were then mixed and treated as one GL sample, without any classification by year of expansion.

Additional GL sampling was performed in one day during each growing season between 2011 and 2015 (i.e, August 7, 2011, May 24, 2012, June 1, 2013, June 14, 2014, June 13, 2015). A total volume of approximately 1 kg-FW was collected from each of two (top and bottom, 2012–2015) or three relative foliar positions (top, middle, and bottom, 2011) in the canopy and precisely classified by year of expansion (age classes of foliar parts, Fig. 1 and Table 1; Yoshihara et al., 2013, 2014b). Yearly classification was performed for parts expanded in 2011 (i.e., during the year of deposition) and after,

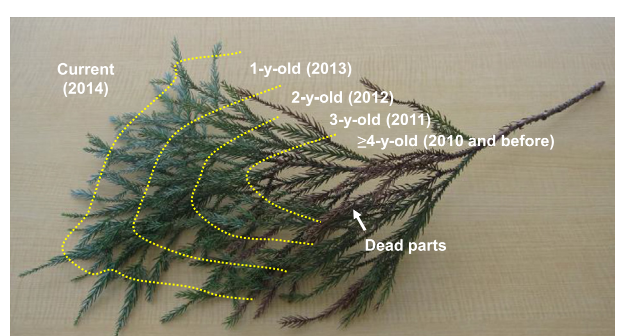

Fig. 1. A typical status of age classification in a cluster of foliar parts. The cluster was collected in June 14, 2014. Dotted lines are showing approximate positions for classification (GL-S) at the sampling time. The possible expansion year is also indicated in parentheses. Arrows indicate dead foliar parts still retained on trees (DL).