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Changes in the distribution of radiocesium in the wood of Japanese cedar trees from 2011 to 2013

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ABSTRACT

The changes in the distribution of ^{137}Cs in the wood of Japanese cedar (*Cryptomeria japonica*) trunks within three years after the Fukushima Dai-ichi Nuclear Power Plant (FDNP) accident in 2011 were investigated. Thirteen trees were felled to collect samples at 6 forests in 2 regions of the Fukushima prefecture. The radial distribution of ^{137}Cs in the wood was measured at different heights. Profiles of ^{137}Cs distribution in the wood changed considerably from 2011 to 2013, and the process of ^{137}Cs distribution change in the wood was clarified. From 2011 to 2012, the active transportation from sapwood to heartwood and the radial diffusion in heartwood proceeded quickly, and the radial ^{137}Cs distribution differed according to the vertical positon of trees. From 2012 to 2013, the vertical diffusion of ^{137}Cs from the treetop to the ground, probably caused by the gradient of ^{137}Cs concentration in the trunk, was observed. Eventually, the radial ^{137}Cs distributions were nearly identical at any vertical positions in 2013. Our results suggested that the active transportation from sapwood to heartwood and the vertical and radial diffusion in heartwood proceeded according to the vertical position of the tree and ^{137}Cs distribution in the wood approached the equilibrium state within three years after the accident.

1. Introduction

Although four years have passed since the Fukushima Dai-ichi Nuclear Power Plant (FDNP) accident, radiocesium contamination is still persistent in East Japan. In Fukushima, high concentrations of radiocesium were deposited northwest of FDNP, in regions such as Namie Town and Iidate Village. High radiocesium concentrations were also deposited in the central region of Fukushima, including the cities of Fukushima, Nihonmatsu and Koriyama, and a particularly high level of radiocesium contaminated the forests in this area. Forestry, one of the major industries of Fukushima prefecture, are supported by the forests that constitute approximately 70% of the Fukushima area. In Japan, Japanese cedar (*Cryptomeria japonica*) is the most propagated tree in plantations and is one of the most valuable for the timber industry. Therefore, it is important to understand radiocesium contamination and its change in the wood of Japanese cedar trees.

The large forest area in Europe was also contaminated with radiocesium caused by the Chernobyl accident. International Atomic Energy Agency (IAEA) reported that the forest contamination process was divided into two phases, the early phase and the steady phase (IAEA, 2009). During the early phase, radioactive materials that were trapped on the forest, such as branches, leaves and bark were translated to the ground with rainfall and fallings leaves immediately. During the steady phase, radiocesium in the soil that was absorbed into the tree through the roots circulated in the forest ecosystem. Tree contamination following the Chernobyl accident was researched extensively (Tikhomirov and Shcheglov, 1994; Fesenko et al., 2001; Yoshida et al., 2004; Calman et al., 2009; Zhiyanski et al., 2010). Some studies documented ^{137}Cs distribution among annual rings of the trees at the steady phase (Soukhova et al., 2003; Yoshida et al., 2011).

The distribution of the radioactive materials in the wood of Japanese cedar trunks has been examined in annual rings for the purpose of understanding changes caused by the fallout derived from the atomic bomb in Japan and experiments with nuclear weapons (Kudo et al., 1993; Momoshima et al., 1995; Mahara et al., 1995; Kagawa et al., 2002). In the reports, ^{137}Cs existed in annual

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rings formed before the fallout, which suggested that ^{137}Cs moves easily within the wood. Kudo et al. (1993) found that 40 years after local fallout, ^{137}Cs concentration in heartwood of Japanese cedar was higher than that in sapwood, and similar results were reported by others (Momoshima et al., 1995; Mahara et al., 1995; Kagawa et al., 2002).

However, most studies of the Chernobyl accident and the local fallout in Japan were performed at least 10 years after the fallout; thus, ^{137}Cs had already moved in the wood and the early distribution might have changes and ^{137}Cs distribution have reached equilibrium state in the wood of trees. Only Garrec et al. (1995) examined ^{137}Cs distribution of white fir (*Abies alba*) in France 3 years after the Chernobyl accident and it differed from that at the steady phase. Following the accident at Fukushima, some studies have investigated tree and forest contamination (Kato et al., 2012; Hashimoto et al., 2013; Kuroda et al., 2013). However, there are few studies regarding ^{137}Cs distribution in the wood. Only Mahara et al. (2014) reported on Japanese cedar and Konara oak (*Quercus serrata*), and found similar results as those by Garrec et al. (1995). These ^{137}Cs distributions reported by the Garrec et al. (1995) and Mahara et al. (2014) were the initial state during the early phase.

^{137}Cs distribution in the wood of whole tree during the early phase is important for understanding the process of ^{137}Cs transportation. In previous studies, a single sample obtained from the ground height of a tree was examined, which is not sufficient for the analysis. Furthermore, no study has addressed the detailed changes in ^{137}Cs distribution during the early phase. The objective of this study was to evaluate the characteristics of ^{137}Cs distribution in the wood of Japanese cedars and to document changes in distribution during the early phase. This knowledge is important for the management of contaminated forests and for the estimation of future contamination of trees.

2. Materials and methods

From 2011 to 2013, 13 Japanese cedar trees were felled in 6 forests of 2 regions in the Fukushima prefecture. Details regarding the study areas and trees used in the experiments are shown in Table 1. In the Kawamata area, 2 Japanese cedar trees (Table 1; No.1–No.10) were felled in each of 5 forests in May 2012. Two or three disks were obtained from each tree at heights of 0.5, 8.5, and 16.5 m above the ground, depending on the height of the tree. The Yamakiya area was highly contaminated and off-limits, and only 1 tree was felled each year in 2011, 2012, and 2013. In November 2011, 3 disks were obtained from tree No. 11 at heights of 0.2, 4.2, and 8.2 m from the ground. In December 2012, 4 disks were obtained from tree No. 12 at heights of 0.5, 5, 10, and 15 m from the ground. In August 2013, 3 disks were obtained from tree No. 13 at heights of 0.5, 3, and 6 m from the ground.

The maximum and the minimum diameter of the disk were measured, and the overall diameter was determined by the average of the maximum and minimum values. Rectangular wood samples were cut from the disks along a straight line via the pith. In the Kawamata area, two sapwood samples (2–3 cm wide × 2–3 cm long × 5 cm thick) were obtained from the both sides of the disk, and one heartwood sample was obtained at the center of the disks. In the Yamakiya area, the rectangular wood samples (1 cm wide × 3 cm long × 5 cm thick) were obtained from the edge to the center of the disk in succession. Samples were dried at 105 °C for 24 h, and ^{137}Cs concentration per kilogram of dry weight of wood samples were measured with a Germanium semiconductor detector (SEG-EMS: SEIKO EG&G Inc., GC2513: CANBERRA) using a 100-ml container, and measured with a NaI Spectrometer (2480WIZARD2: Perkin Elmer) using a 20-ml container with the accumulation time from 3600 to 80,000 s. The measurement error by the Germanium semiconductor detector was less than 3–5 Bq kg^{-1}, and the relationships of ^{137}C concentration measurement by the Germanium semiconductor detector (C_G) and by the NaI Spectrometer (C_N) were shown as a linear equation with slope of unity (linear approximation: $C_N = 1.0183 \times C_G$, $R^2 = 0.9387$). ^{137}Cs concentration was determined after decay compensation on March 15, 2011.

The wood samples were measured as rectangular and powder samples. The precision of the measurement of rectangular samples was confirmed by the following method. Firstly, ^{137}Cs concentrations from the rectangular samples (C_A) and that of the upside down rectangular samples (C_B) were compared (linear approximation: $C_B = 1.0012 \times C_A$, $R^2 = 0.9946$). Secondly, the average of ^{137}Cs concentrations of the rectangular samples (C_{Ave}) and the powder samples (C_P) were compered (linear approximation: $C_p = 0.9862 \times C_{Ave}$, $R^2 = 0.9969$). The relationships for both were shown as a linear equation with slope of unity and strong positive correlation. Therefore, it was confirmed that the measurement of a rectangular sample was sufficient to accurately evaluate ^{137}Cs concentration in the wood in this study.

Table 1
Characterization of Study area and Japanese cedar trees used for the experiments.

Study area	Deposition[a] ^{137}Cs [kBq/m^2]	Distance from FDNP [km]	Sampling year [Year]	Tree no.	Tree age[b] [Year]	Diameter[c] [M]	Tree height[d] [M]
Kawamata	**101**	**50.3**	**2012**	**No. 1**	**48**	**0.36**	**19.4**
				No. 2	**38**	**0.19**	**17.4**
	251	**49.1**	**2012**	**No. 3**	**47**	**0.22**	**13.6**
				No. 4	**41**	**0.15**	**12.2**
	133	**42.3**	**2012**	**No. 5**	**56**	**0.35**	**22.5**
				No. 6	**53**	**0.26**	**24.0**
	112	**44.7**	**2012**	**No. 7**	**49**	**0.30**	**22.7**
				No. 8	**39**	**0.22**	**19.5**
	139	**45.3**	**2012**	**No. 9**	**38**	**0.19**	**17.7**
				No. 10	**30**	**0.23**	**14.9**[e]
Yamakiya	**570**	**34.2**	**2011**	**No. 11**	**35**	**0.24**	15.0
			2012	**No. 12**	**36**	0.25	21.2
			2013	**No. 13**	**37**	0.26	12.7

[a] Total deposition of ^{137}Cs estimated by the exchange equation (NRA, 2012) from air dose rate obtained by 3rd airplane measurement (NRA, 2011).
[b] Age of tree evaluated by the number of annual rings at 0.5 m height.
[c] Diameter of tree evaluated at 1.2 m height.
[d] Height of tree from the ground surface to the crown top.
[e] The tip of tree was broken before the fallout.