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NOTE

Relationship between ^{137}Cs concentration and potassium content in stem wood of Japanese cedar (*Cryptomeria japonica*)

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Abstract To utilize forest resources in areas affected by fallout from the Fukushima Daiichi Nuclear Power Plant accident, it is important to understand the mechanisms of ^{137}Cs movement through the stem wood of contaminated trees. Understanding the mechanism of absorption and migration of ^{137}Cs to stem wood is necessary for clues to the future prediction of the transition of ^{137}Cs to xylem. In the present study, radial variations in ^{137}Cs concentration were investigated in Japanese cedar (*Cryptomeria japonica* D. Don) trees collected 1 year and 10 months after the accident. Additionally, the relationship between ^{137}Cs concentration and potassium (K) content was established. Trees with a higher moisture content and lower lightness value in heartwood tended to have a higher ^{137}Cs concentration in the heartwood. In these trees, ^{137}Cs concentration peaked at the heartwood–sapwood boundary and gradually decreased toward the pith. By contrast, K content within the heartwood remained nearly constant along the radial direction. The heartwood-to-sapwood ratio of ^{137}Cs concentration was significantly positively correlated with that of K content. Based on these results, we suggest that ^{137}Cs movement from sapwood to heartwood might be related to the K content ratio of heartwood and sapwood.

Keywords Japanese cedar · Stem wood · Heartwood · ^{137}Cs concentration · Potassium content

Introduction

Radiocesium (^{134}Cs and ^{137}Cs) was emitted into the atmosphere during the Fukushima Daiichi Nuclear Power Plant accident in March 2011, contaminating a large area of northeastern Japan. As the physical half-life of ^{137}Cs is 30.2 years, approximately 90% of the ^{137}Cs released remains 5 years after the accident. Thus, environmental contamination by ^{137}Cs deposition is still present in the forests in Fukushima Prefecture as well as in neighboring prefectures.

Japanese cedar (*Cryptomeria japonica* D. Don) is a native species of Japan and extensively used as a plantation forestry species. The wood from this species is utilized by the wood industry to produce construction lumber. The mobility of radiocesium emitted by the global fallout from atmospheric nuclear bomb testing or the Atomic bombs at Hiroshima and Nagasaki has been previously investigated in the stem wood of Japanese cedar [1–6]. Recently, it was reported that the radiocesium emitted by the accident in 2011 may be translocated to the stem wood of Japanese cedar via the foliar surface [7], bark surface [8], and root uptake [9]. Similar results have been obtained on the radioactive contamination of stem wood of other forest tree species [10–13]. Furthermore, Aoki et al. [14] suggested that stable cesium (^{133}Cs) was radially transported from the bark to stem wood via ray parenchyma cells in 3-year-old Japanese cedar seedlings. It is important, therefore, to clarify the within-tree distribution of ^{137}Cs in Japanese cedar from the initial period of contamination. In addition, the movement of ^{137}Cs in the stem wood should be determined to predict the future level of contamination

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of Japanese plantation forests and wood resources from the plantations.

Potassium (K), an alkali metal in the same group as cesium, is closely related to the blackening phenomenon in the heartwood of Japanese cedar. Heartwood with higher K contents have a lower lightness (*L**) value in the *L***a***b** system and a higher moisture content [15–20]. Okada et al. [21] reported that the K and other alkali metal contents in Japanese cedar showed an abrupt increase from sapwood to heartwood and that they showed almost constant values in the heartwood. They also found that K and other alkali metals are actively transported from the sapwood to the outer heartwood via the rays, resulting in their diffusion to and accumulation in the heartwood of Japanese cedar [22, 23]. Furthermore, the relationships between ^{137}Cs and ^{40}K in the wood of Japanese cedar have been investigated in several studies [1–3]. However, these were investigations conducted decades after the fallout event; moreover, the number of trees investigated was limited. Therefore, further research on these relationships at an early stage after fallout with a large number of trees is needed.

In the present study, ^{137}Cs concentration, K content, moisture level, and *L** of heartwood were investigated in Japanese cedar trees grown in a plantation located 130 km southwest of the Fukushima Daiichi Nuclear Power Plant. The movement of ^{137}Cs from sapwood to heartwood and its relationship to K content were investigated.

Materials and methods

Wood samples from Japanese cedar were collected from a plantation in the Funyu Experiment Forest, Utsunomiya University, Tochigi, Japan (36°46′N, 139°49′E). The University Forest is located at about 130 km southwest of the nuclear plant which caused the accident. According to the results of aircraft monitoring by the Ministry of Education, Culture, Sports, Science and Technology, the soil pollution degree of Funyu Experimental forest is estimated to have been 30–100 kBq/m^2 as of July 16, 2011 [24].

Disks (5 cm in thickness) were collected from 0.3 to 0.4 m above the ground for twenty 40-year-old Japanese cedar trees in January 2013. To measure the moisture content, blocks of heartwood and sapwood were prepared from the disks. Moisture contents were measured by the oven-drying method. The *L** of heartwood in the air-dry condition was measured using radial boards (approximately 30 mm longitudinal × 50 mm radial plane × 20 mm tangential plane) prepared from the sample disks. The *L** of the heartwood in both radial boards was measured using a chroma meter (CR-400, Konica Minolta, Tokyo, Japan) by the *L***a***b** system.

The disks were divided into fan shaped parts, and then heartwood and sapwood were divided. In addition, heartwood was divided into three parts, namely, H1, H2, and H3, from the pith to the bark. Sapwood was divided into two parts, namely, S1 for the heartwood side and S2 for the bark side. Each sample (the total seven samples in the parts of heartwood, sapwood, H1, H2, H3, S1, and S2) was ground using a mill (IFM-S10G, Iwatani, Tokyo, Japan) to prepare wood powder for measuring ^{137}Cs concentrations. The wood meal was oven-dried and then packed in a U-8 container (100 mL). The concentration of ^{137}Cs was measured with a germanium (Ge) semiconductor detector (Seiko EG&G, Ortec, Tokyo, Japan). Measurement conditions were as follows: measurement duration, 6000 s or longer; gamma-ray peaks, 661.64 keV.

K content (g/kg dry weight) in the heartwood and sapwood from 12 selected individuals was measured by atomic absorption spectroscopy. Oven-dried wood meal, used for measuring ^{137}Cs concentration, was also used for this experiment. To obtain ash, the wood meal was placed in an electric muffle furnace (FUL220FA, Advantec, Osaka Yakken, Osaka, Japan) at 500 °C for 24 h. The ash was suspended in 0.1 M hydrochloric acid. K content was determined with an atomic absorption photometer (Z-2310, Hitachi, Tokyo, Japan).

Results and discussion

The mean moisture contents of heartwood and sapwood were 152 and 247%, respectively (Table 1). Based on these results, the sampled trees were categorized into three groups: the six with the highest moisture contents (HMC), another six with the lowest ones (LMC), and the remainder. The trees in the HMC group had lower *L** values in their heartwood than those in the LMC group (Table 1). However, there were no significant differences in mean sapwood moisture contents between the HMC and LMC groups (Table 1). The relationship between *L** value and moisture content in heartwood is shown in Fig. 1. A significant, highly negative correlation ($r = -0.839$, $p < 0.01$) was found between these traits. Similar correlations between *L** and moisture content in heartwood of Japanese cedar have been reported previously [15, 16, 18, 20].

Changes in K content from pith to bark in HMC and LMC groups are shown in Fig. 2. Black and white columns indicate HMC and LMC groups. K contents in both groups gradually increased from S2 to H1. The mean contents in heartwood ranged from 3.05 to 3.65 g/kg in the HMC group and from 1.17 to 1.28 g/kg in the LMC group. In sapwood, the contents ranged from 0.90 to 1.33 g/kg and from 0.55 to 0.85 g/kg in the HMC and LMC groups, respectively. The contents in the HMC group were threefold and 1.5-fold higher than those in the LMC group. Oda et al. [16] reported that K content in the heartwood of Japanese cedar

Table 1 Moisture content and L^* value in heartwood of 40-year-old trees

Category	*n*	Moisture content (%)				L^*	
		Heartwood		Sapwood			
		Mean	SD	Mean	SD	Mean	SD
Total	20	152	57	247	29	62.16	5.54
HMC	6	197	20	232	25	56.11	2.50
LMC	6	76	17	244	21	68.62	1.65
Others	8	176	33	261	34	61.86	3.08

HMC and LMC indicate six highest heartwood moisture-content individuals and six lowest heartwood moisture-content individuals, respectively

n number of samples, *SD* standard deviation

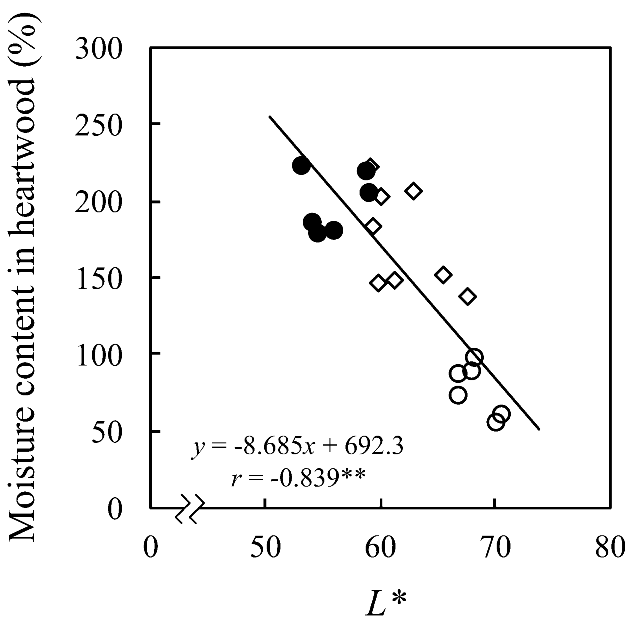

Fig. 1 Relationship between L^* value and moisture content in heartwood in 40-year-old Japanese cedar trees. *r*, correlation coefficient; **, significant at 1% level. Samples were categorized into three groups based on the results of moisture content measurements. The six individuals with the highest and the six individuals with the lowest moisture content were categorized in the HMC (closed circles) group and the LMC (open circles) group, respectively. The remaining eight individuals were classed as "others" (open diamonds)

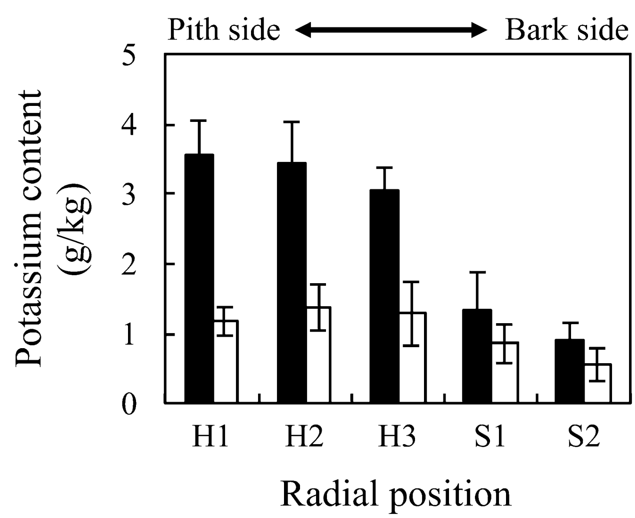

Fig. 2 Radial variation in potassium content in HMC and LMC groups. Black and white columns indicate HMC and LMC groups, respectively. Bars indicate standard deviations. Abbreviations for radial positions are defined in Fig. 1

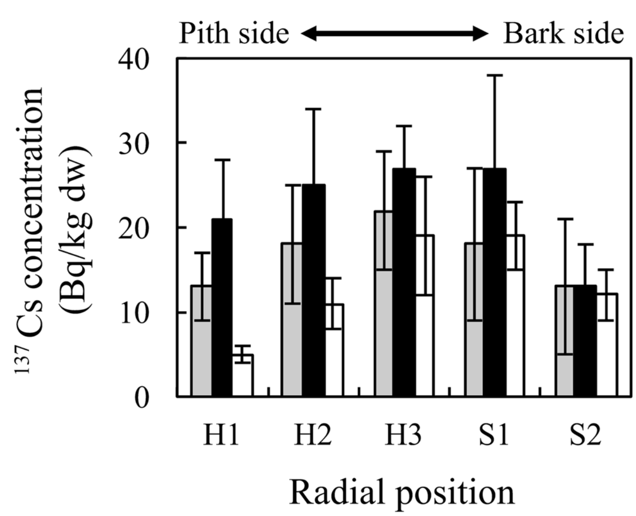

Fig. 3 Radial variation in ^{137}Cs concentration in the three groups. Gray, black, and white columns indicate total number, HMC, and LMC groups, respectively. Bars indicate standard deviations. Abbreviations for radial positions are defined in Fig. 1

was higher than that in sapwood. In addition, Ishiguri et al. [20] reported a significant positive correlation between the K and moisture contents of heartwood in Japanese cedar. The results obtained in the present study are similar to those reported elsewhere [2, 16, 19, 21].

Radial variation in the ^{137}Cs concentration in each group is shown in Fig. 3. Gray, black, and white columns indicate total number, HMC and LMC groups. ^{137}Cs concentration ratios in the H1–H3 positions in HMC group were lower (22%) than those in the LMC group (74%). These results suggested that Japanese cedar trees with higher heartwood moisture contents tended to accumulate more ^{137}Cs in the

Table 2 ^{137}Cs concentration, potassium content, and moisture content of heartwood and sapwood

Factor	Unit	*n*	Heartwood (H)		Sapwood (S)		H/S	
			Mean	SD	Mean	SD	Mean	SD
^{137}Cs concentration	Bq/kg dw	12	20	8	16	5	1.4	0.8
Potassium content	g/kg	12	2.31	1.13	0.91	0.37	2.64	1.16
Moisture content	%	12	137	66	238	23	0.59	0.30

n number of samples, *SD* standard deviation, *H/S* heartwood/sapwood

heartwood than trees with lower heartwood moisture contents. When rubidium (Rb), an alkali metal in the same group as K and Cs, was injected into the stem of several Japanese cedar cultivars with different heartwood moisture contents, Rb and K concentrations in the heartwood were higher in cultivars with higher heartwood moisture contents than in those with lower heartwood moisture contents [22]. Our results for ^{137}Cs and K are similar to those reported by Okada et al. [22]. Okada et al. [21] reported that the concentration of Cs and other alkali metals in Japanese cedar heartwood remained nearly constant along the radial direction. However, in the present study, the radial variation in ^{137}Cs concentration differed from that of K content (Fig. 2). Katayama et al. [2] examined radial variations of ^{137}Cs and ^{40}K concentrations in Japanese cedar by analysis of 115 annual rings harvested in 1979. They found that ^{137}Cs was detected in annual rings formed before 1945, suggesting that ^{137}Cs might move within the stem wood. A similar result was obtained by Kudo et al. [3].

Okada et al. [21] reported that abrupt changes were observed in metal element concentrations near the sapwood–heartwood boundary, although most elements were evenly distributed in the heartwood. They also concluded that physiological processes, such as heartwood formation, might exert an important influence on the distribution of trace elements under ordinary growing conditions. Therefore, the diffusion of ^{137}Cs and K within heartwood might be related to physiological changes related to heartwood formation.

Bruce and Richard [25] reported that the movement of elements in the xylem is based on the moisture content of the heartwood, the permeability, and the nature of sapwood–heartwood transition, and then K is an element showing high mobility. In order to discuss the movement of ^{137}Cs from sapwood to heartwood, Table 2 shows measurement results in ^{137}Cs concentration, K content and moisture content of a total of 12 individuals categorized into HMC and LMC. K content was calculated from the average of the measured values of three parts in the heartwood and two parts in the sapwood, respectively. The Heartwood (H)/Sapwood (S) of ^{137}Cs concentration, K content, and moisture content were 1.4, 2.64 and 0.59, respectively. The correlation coefficient between H/S of the 3 factors is shown in Table 3. A significant, highly

Table 3 Correlation coefficient between the three factor of H/S

Factor	Code	X1	X2
H/S of ^{137}Cs concentration	X1	1.000	
H/S of potassium content	X2	0.879**	1.000
H/S of moisture content	X3	0.504 ns	0.544 ns

H/S heartwood/sapwood, ** significant at 1% level, *ns* no significance

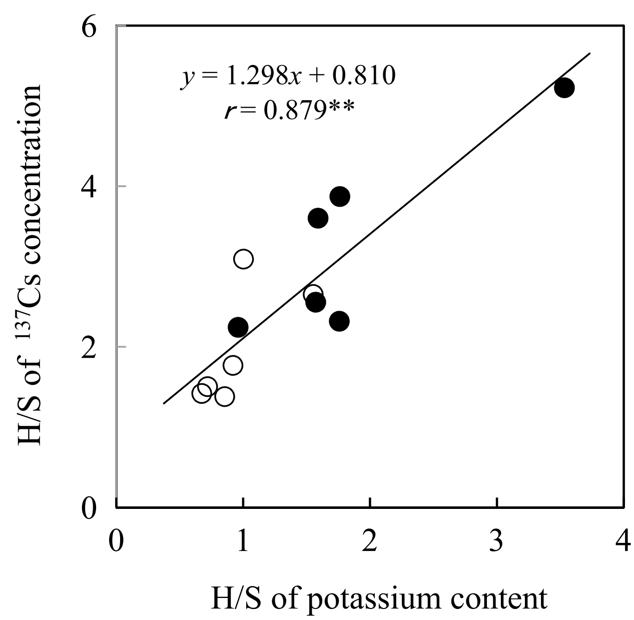

Fig. 4 Relationship between heartwood (H) / sapwood (S) with respect to potassium content and ^{137}Cs concentration. Filled circles and open circles indicate HMC and LMC groups, respectively. **, significant at 1% level

positive correlation coefficient ($r = 0.879$, $p < 0.01$) was obtained between H/S of ^{137}Cs concentration and that of K content (Fig. 4). As reported in Bruce and Richard [25], it was confirmed that the movement of ^{137}Cs from sapwood to heartwood was related to the H/S of K content. A correlation coefficient of 0.5 or more was obtained between H/S of ^{137}Cs concentration and that of moisture content and between H/S of moisture content and that of K content, but there was no significance. Cs is a Group I alkali metal with chemical properties similar to K and it is present in solution as the movement cation Cs^+ [26]. Potassium supply exerts the great influence on Cs uptake from solution. It appears that the uptake of radiocesium is operated mainly by two transport pathways on plant root cell membranes, namely the K^+ transporter and the K^+ channel pathway [27]. Based on these reports, a continued investigation on the relation between Cs, K, and moisture content in the xylem is necessary.

In the present study, the trees with higher moisture content and lower lightness value in heartwood tended to have higher ^{137}Cs concentrations in the heartwood. In the trees with higher heartwood moisture content, the peak values of ^{137}Cs concentration were observed at the heartwood–sapwood boundary, and ^{137}Cs concentration gradually decreased toward the pith. On the other hand, K content was nearly constant along the radial direction within the heartwood. The heartwood-to-sapwood ratio of ^{137}Cs concentration was significantly and positively correlated with that of K content. Based on these results and the already reported findings, we suggest that ^{137}Cs movement from sapwood to heartwood might be related to the K content ratio of heartwood-to-sapwood.

Conclusion

Japanese cedar trees with higher moisture content and lower L^* value in the heartwood tend to have a higher concentration of ^{137}Cs. In trees with higher heartwood moisture content, ^{137}Cs concentration gradually decreased from the heartwood–sapwood boundary to the pith, although K content in heartwood was nearly constant. A significant, highly positive correlation was found between H/S of K content and that of ^{137}Cs concentration. Therefore, based on the present study and the already reported findings, we suggest that the movement of ^{137}Cs from sapwood to heartwood might be influenced by the content gradient in K from sapwood to heartwood.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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