J Wood Sci (2011) 57:421–428
DOI 10.1007/s10086-011-1188-8

ORIGINAL ARTICLE

Naoki Okada · Yasuhiko Hirakawa · Yukio Katayama

Application of activable tracers to investigate radial movement of minerals in the stem of Japanese cedar (*Cryptomeria japonica*)

Received: September 6, 2010 / Accepted: March 16, 2011 / Published online: June 28, 2011

Abstract Two activable tracers, Rb and Eu, were injected into the sapwood of Japanese cedars (*Cryptomeria japonica* D. Don) to investigate the radial movement of minerals in their stems in the resting period. Eight trees of four cultivars, two of which genetically form wet heartwood, were treated near the end of the growing period. At 40 days after the treatment, Rb was detected in the outer heartwood, whereas Eu was not. Radial movement of Rb was more rapid in trees with wet heartwood than in those with normal heartwood. At 204 days after the treatment, more Rb was detected in the heartwood than was found on the first sampling, whereas no Eu was detected in the heartwood. The difference in radial movement between Rb and Eu was considered mainly to be the result of selective transport of beneficial minerals by Japanese cedar. The difference in the rate of radial movement of Rb between wet and normal heartwood became more conspicuous at 204 days after treatment. We concluded that the movement of Rb from the sapwood to the outer heartwood was by active transport through the rays, whereas that in the heartwood was by diffusion due to the gradient of Rb concentration.

Key words Rb · Eu · Neutron activation analysis · Radial transport · Heartwood formation

Introduction

Radial movement of materials in tree stems has not attracted much attention from researchers, whereas vertical movement has been extensively surveyed in relation to tree physiology.[1] Rays have been considered to play an important role in radial transport in many tree species,[2–4] but further effort to clarify the details is required.

In recent years, increased environmental concerns revived this topic among those who work with tree rings to elucidate pollution histories.[5] This approach assures correct results only when the pollutants are immobile in tree rings after being taken up by trees. For this reason, the validity of the tree ring method has been sometimes exposed to skepticism.[6–8] In fact, studies on artificial radionuclides ^{90}Sr and ^{137}Cs in tree rings[9–14] have revealed that the nuclides can be found in rings formed before 1945, and that ^{137}Cs has similar radial profiles to that of a natural radionuclide, ^{40}K. In addition, Lepp and Dollerd[15] have confirmed lateral movement of bark-applied ^{210}Pb to wood whether the above-ground parts of the plant are dormant or not. These observations provide direct evidence of radial movement of elements after they are taken up in tree stems or are even bark applied, and suggest that some elements absorbed in trees are not necessarily fixed in the ring of the respective growing season. However, the above studies have not specified the pathway and mechanism of movement of the radionuclides.

On the other hand, some tree species are reported to accumulate minerals in their heartwood;[16,17] Japanese cedar (*Cryptomeria japonica* D. Don) is one of these species. It accumulates a considerable amount of moisture in addition to minerals in its heartwood in the case of dark-colored heartwood.[18] This phenomenon is well known and is called black heartwood; it deteriorates wood quality and consequently results in a lower commercial value. It has been controversial as to what causes black heartwood in Japanese cedar. Abe and Oda[19] reported that K in heartwood as potassium hydrogen carbonate is one substance that causes black heartwood. However, it is still unresolved why and how K is accumulated in the black heartwood of Japanese cedar.

The difficulties of studying the radial movement of materials in a tree stem are ascribable to the lack of appropriate methods. Radioactive tracers were once a useful tool for

N. Okada · Y. Hirakawa
Forestry and Forest Products Research Institute, Tsukuba, Ibaraki 305-8687, Japan

N. Okada (✉) · Y. Katayama
Graduate School of Agriculture, Kyoto University, Kitashirakawa Oiwake-cho, Sakyo-ku, Kyoto 606-8502, Japan
Tel. +81-75-753-6097; Fax +81-75-753-6098
e-mail: okad@kais.kyoto-u.ac.jp

Y. Katayama
Division of Human Environment, University of Human Environments, Okazaki 444-3505, Japan

Table 1. Moisture content (%) in the heartwood of sample trees

Height above the ground[a] (m)	Higashishirakawa8		Minamiaizu5		Shimotakai7		Shimotakai27	
	HS8-1[b]	HS8-2[b]	MA5-1	MA5-2	ST7-1	ST7-2	ST27-1	ST27-2
1.5	293.8	199.1	89.6	89.5	158.1	189.8	89.7	95.8
3.5	208.2	177.3	69.4	74.2	175.3	143.2	89.2	81.9
5.5	261.7	174.5	70.1	68.1	156.5	122.9	68.7	143.5
7.5	251.5	224.3	65.6	71.1	164.8	148.7	–	126.2

Data were obtained by Hirakawa (unpublished data), one of the authors, for a wood quality study of Japanese cedar

[a] Activable tracers were injected into the stem at around 1.7 m above the ground

[b] Tree-1 indicates Rb injection and tree-2 indicates Eu injection

tracing the movement of substances, but nowadays application in the field is strictly restricted. Instead, activable tracers are expected to be a useful tool to overview the movement of materials in soil–plant systems.[20,21] We applied this method to Japanese cedar in a series of studies to investigate radial transport of minerals in the stem. In the present study, we first examined the potential of this method, and then used it to discuss the radial movement of minerals, especially of physiologically important K, in the resting period of tree growth.

Materials and methods

Materials

Four cultivars of Japanese cedar were selected from the clones planted in the National Forest Tree Breeding Center (now the Forest Tree Breeding Center, Forestry and Forest Products Research Institute), taking into consideration the moisture content of their heartwood, i.e., Higashishirakawa8 (HS8), Shimotakai7 (ST7), Minamiaizu5 (MA5), and Shimotakai27 (ST27), all of which were growing at the same site and were the same age (ca. 30 years old). A preliminary survey (Hirakawa, unpublished data) showed that the former two cultivars have wetter heartwood than the latter two.

Rubidium (Rb) and europium (Eu) were chosen as activable tracers for the reasons mentioned later. Chlorides of Rb and Eu were dissolved in pure water to prepare solutions of 100 and 5 mM, respectively. Five hundred milliliters of the solution in a plastic bottle was set on the stem of sample trees at around 1.7 m above the ground. The solutions were injected through tygon tube with a stainless tube (3 mm ϕ) at its end into the middle of the sapwood region on September 1, 1994, at nearly the end of the growing season. Each tree absorbed almost all the solution within 1 week.

Two cultivars, HS8 and MA5, were felled at 40 days after the treatment, at which time tree growth had almost ceased. Trees injected with Rb are designated “1” (e.g., HS8-1), and trees injected with Eu are designated “2” (e.g., HS8-2). After marking the point of injection, disks were removed from each tree at four stem heights: just above the injection point (+0 m), +1.5 m, +3.5 m, and +5.5 m. The two other cultivars, ST7 and ST27, were left for 204 days and felled before the beginning of the following growing season. Numbers 1 and 2 were also appended to the cultivar identifier, as above. Disks were removed from the trees also at four stem heights: just above the injection point (+0 m), +2 m, +4 m, and +6 m. The disk at +6 m of ST27-1 could not be sampled because of injury and rot caused by insect attack.

The moisture content in the heartwood of Japanese cedar clones in the National Forest Tree Breeding Center was measured by one of the authors for a wood quality study, and the data of the sample cultivars are shown in Table 1. HS8 and ST7 contained much more moisture in their heartwood than MA5 and ST27 did. The heartwood color of the former two cultivars became darker after felling. Thus, we confirmed that the sample trees fulfilled the requirements for a comparison between wet and normal heartwood.

After air-drying, wood strips of 10 mm (L) × 2 mm (T) were cut from four radii (injection side, I; opposite side, O; left side, L; and right side, R; see Fig. 1) from disks of two cultivars, HS8 and MA5, to determine the tracer concentrations. To obtain an overview of tracer movement in a cross section of each disk, tracer content of whole sapwood and outer heartwood (up to 20 mm inward from the sapwood–heartwood boundary) of four strips were investigated. Eu-injected trees (HS8-2 and MA5-2) were used as the controls for the Rb experiments on HS8-1 and MA5-1. Preliminary results of cultivars HS8 and MA5 indicated that tracers ascended the sapwood vertically from the point of injection and were not detected in the other three radii. Based on these results, wood strips of the other two cultivars, ST7 and ST27, were cut only from the radius starting at the injection point. In addition, wood strips were cut from the opposite side of the injection point of ST7-1 and ST27-1 as controls for the Rb content. To give a radial profile of tracer movement, each sapwood sample was evenly divided into three segments, and each outer heartwood sample was divided into four segments, each of which was 10 mm in radial length. The whole sampling procedure is shown in Fig. 1.

During the drying of sample disks, redistribution of tracers might occur, but this should not seriously influence the results. Since water in wood is vaporized in drying, the movement of liquid water including minerals (i.e., mass flow) should not occur in the sample disks. In addition, the movement of water is mostly along the tangential direction in the disks. Thus, we judged that the influence of possible redistribution of tracers, if any, was negligible in discussing the radial movement from sapwood to heartwood.

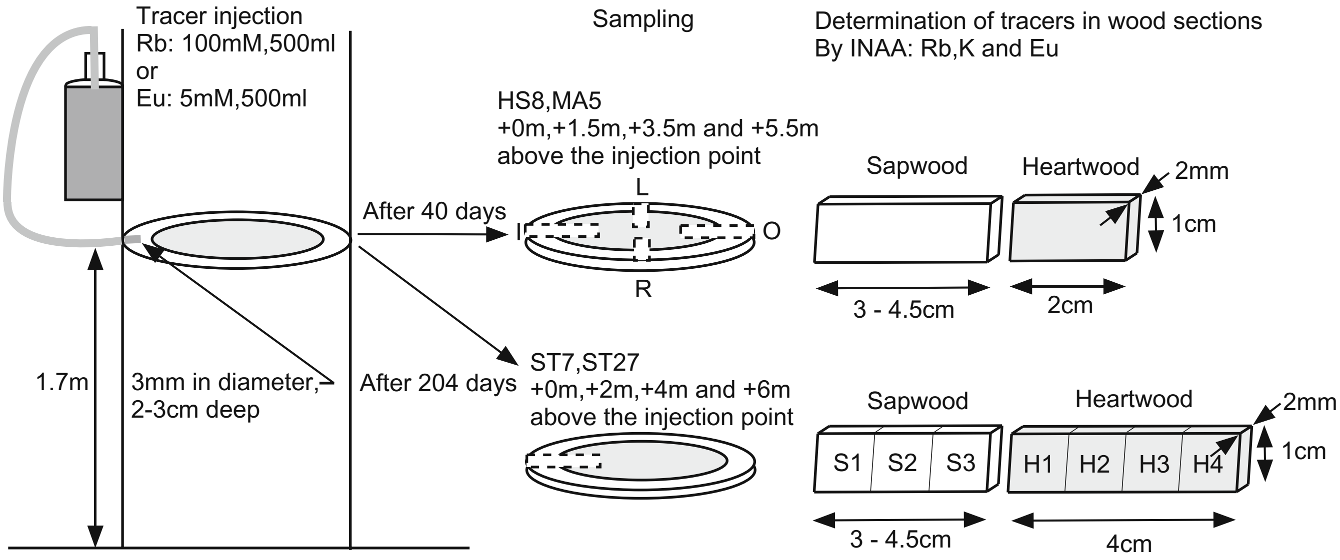

Fig. 1. Tracer injection and preparation of analytical samples. *I*, injection side; *O*, opposite side; *L*, left side; *R*, right side; *INAA*, instrumental neutron activation analysis

Determination of activable tracer concentrations

Activable tracers are nonradioactive elements that can be easily made radioactive by neutron irradiation. They are usually detected by means of instrumental neutron activation analysis (INAA) and are useful tools for field research where the use of radioactive tracers is not allowed.

Rubidium and europium used in this study become radioactive by irradiation with thermal neutron as follows:

$$^{85}\mathrm{Rb}(\mathrm{n},\gamma)\,^{86}\mathrm{Rb}$$

$$^{151}\mathrm{Eu}(\mathrm{n},\gamma)\,^{152}\mathrm{Eu}$$

The former formula indicates that ^{85}Rb, the most common rubidium isotope, absorbs a neutron to become ^{86}Rb, which decays with the emission of γ-rays of characteristic energy. This transformation enables us to quantitatively determine the tracer in materials by measuring the characteristic γ-rays.

Rubidium is an alkali metal and accordingly has similar chemical properties to other members of the group, especially K, because of their similar ion radii and ionization potentials. This property makes it easier for us to trace the behavior of K in a tree stem. On the other hand, Eu, one of the rare earth elements, is scarcely detected in Japanese cedar. This element has a large cross section for neutron activation and is, therefore, very sensitive in terms of INAA. These are the reasons that we choose these two elements as activable tracers in this study.

Rubidium and europium in wood were determined by means of INAA. Potassium was also determined for reference purposes. About 150–300 mg of wood specimens were irradiated by thermal neutrons, typically for 2 min, at the facility of the Reactor Research Institute, Kyoto University, or the Institute of Atomic Energy, Rikkyo University. After one day of cooling, specimens were measured by using a Ge solid-state detector with a multichannel pulse-height analyzer for 10–20 min to determine the K content. For the determination of Rb and Eu, wood specimens were irradiated for 60 min and measured for 20–60 min after 1 week of cooling. The determination of tracers was conducted within 1 year after felling each tree. The details of activation analysis are described in an earlier report.[22]

Results

Estimation of the background Rb content in the stem of Japanese cedar

Rubidium is usually detected in tree stems in small amounts; the concentrations in sap and wood substances of Japanese cedar are reported to be less than 5 ppm.[23] Before discussing the movement of injected Rb in Japanese cedar, it is necessary to estimate the background content. Two Eu-injected trees, HS8-2 and MA5-2, were used as control trees. Table 2 summarizes Rb and K contents and their ratio (Rb/K) in sapwood and outer heartwood of the control trees. Rubidium could not be determined in the injection sides at the lower three heights in HS8-2 and at the lowest in MA5-2 because of the interference from high-yield Eu-induced γ-rays. Concentrations of Rb and K varied in the ranges 1.9–28 ppm and 590–8100 ppm, respectively, in all the samples analyzed. Despite the considerable differences in element contents, the values of Rb/K were quite similar (0.0024–0.0049), even between different cultivars. In addition, when Rb/K values at different stem heights were grouped for sapwood and heartwood on the injection and opposite sides (i.e., IS, IH, OS and OH), no significant differences were detected among the four groups by ANOVA (HS8-2: $F = 0.091$, $P = 0.963$, MA5-2: $F = 0.716$, $P = 0.563$). Thus, we judged that the mean values of Rb/K (HS8 = 0.0039, MA5 = 0.0031) can be used as the background in discussing the movement of Rb in stems.

Table 2. Background Rb and K contents and Rb/K ratio at four different heights in control trees, i.e., Eu-injected trees not injected with Rb

	+0 m[a]			+1.5 m			+3.5 m			+5.5 m		
	Rb (ppm)	K (ppm)	Rb/K	Rb (ppm)	K (ppm)	Rb/K	Rb (ppm)	K (ppm)	Rb/K	Rb (ppm)	K (ppm)	Rb/K
HS8-2												
IS	–	–	–	–	–	–	–	–	–	4.1	1100	3.7E-03
IH	14	3600	3.9E-03	17	4400	3.9E-03	15	3800	3.9E-03	28	8100	3.5E-03
OS	3.8	780	4.9E-03	5.0	1200	4.2E-03	3.6	890	4.0E-03	2.5	890	2.8E-03
OH	21	5900	3.6E-03	25	5800	4.3E-03	27	6300	4.3E-03	22	6400	3.4E-03
MA5-2												
IS	–	–	–	1.9	730	2.6E-03	2.2	690	3.2E-03	2.1	680	3.1E-03
IH	8.0	2500	3.2E-03	6.8	2800	2.4E-03	8.1	2600	3.1E-03	7.2	2400	3.0E-03
OS	2.0	620	3.2E-03	1.9	590	3.2E-03	2.6	820	3.2E-03	2.6	960	2.7E-03
OH	7.7	2200	3.5E-03	6.9	2200	3.1E-03	11	3600	3.1E-03	8.7	2800	3.1E-03

[a] Height above the injection point

IS, sapwood of the injection side; IH, heartwood of the injection side; OS, sapwood of the opposite side; OH, heartwood of the opposite side

Rb content and Rb/K in Rb-injected trees at 40 days after the treatment

Rubidium content increased both in the sapwood and the heartwood of the injection side of HS8-1 (Fig. 2, left). The concentrations in the sapwood were from 45 to 60 ppm at the four stem heights, nearly 10 times the values for the other three sides. The concentrations in the heartwood of the injection side were from 45 to 125 ppm at the four stem heights, 2–5 times the values for the other three sides. The value of Rb/K also indicated that Rb injected in the middle of sapwood region at a height of +0 m moved upward and inward in the stem.

The results for MA5-1 were similar to those for HS8-1, but the concentration of Rb was much lower than the latter (Fig. 2, right). At a height of +5.5 m, there were no clear differences in Rb content among the four radii. However, the Rb/K value indicated the movement of Rb into outer heartwood at the lower three heights.

Eu content in Eu-injected trees at 40 days after the treatment

Europium was detected only in the sapwood of the injection side of the two cultivars (Table 3). The concentration drastically decreased upward from 2100 ppm at +0 m to 0.59 ppm at +5.5 m in HS8-2, and from 204 ppm at +0 m to 0.33 ppm at +1.5 m in MA5-2. No Eu was detected at +3.5 m and +5.5 m in MA5-2. The great vertical differences in Eu content compared with Rb content were probably based on the differences in their chemical properties. As was also the case for Rb, the Eu content was greater in HS8-2 than in MA5-2.

Table 3. Eu content (ppm) in stems of two Eu-injected Japanese cedars

Sample	+0 m[a]	+1.5 m	+3.5 m	+5.5 m
HS8-2				
IS	2100	500	100	0.59
IH	nd	nd	nd	nd
OS	nd	nd	nd	nd
OH	nd	nd	nd	nd
MA5-2				
IS	240	0.33	nd	nd
IH	nd	nd	nd	nd
OS	nd	nd	nd	nd
OH	nd	nd	nd	nd

[a] Height above the injection point

IS, sapwood of the injection side; IH, heartwood of the injection side; OS, sapwood of the opposite side; OH, heartwood of the opposite side; nd, not detected

Table 4. Background contents of Rb and K and Rb/K in two cultivars of Japanese cedar

Radial position[a]	ST7-1			ST27-1		
	Rb (ppm)	K (ppm)	Rb/K	Rb (ppm)	K (ppm)	Rb/K
S1	3.6	340	1.1E-02	2.1	260	8.1E-03
S2	8.2	890	9.2E-03	3.1	370	8.4E-03
S3	9.1	1200	7.6E-03	6.1	830	7.3E-03
H1	15	2900	5.2E-03	5.9	1400	4.2E-03
H2	13	3100	4.2E-03	6.9	1400	4.9E-03
H3	18	3900	4.6E-03	7.1	1600	4.4E-03
H4	23	4200	5.5E-03	–	–	–

[a] S1, the outermost sapwood; S3, the innermost sapwood; H1, the outermost heartwood; H4, the innermost section analyzed

Rb content and Rb/K in Rb-injected trees at 204 days after the treatment

There was not much difference in base level Rb/K among the four stem heights (Table 2), and the injected Rb did not move to the opposite side of the stem (Fig. 2). In examining the trees at 204 days after treatment, therefore, the opposite side of the lowest height (+0 m) of Rb-injected trees was analyzed as the control. The Rb contents in the opposite side from the injection point were 3.6–23 ppm in ST7-1 and 2.1–7.1 ppm in ST27-1 (Table 4), which are almost as same as the values of the two cultivars given in Table 2. As already seen in Table 2, the cultivar with wetter heartwood (ST7) contains more Rb, especially in heartwood, than the normal heartwood (ST27) does in this determination. Despite the differences in Rb and K contents, the values of Rb/K of the two cultivars are quite similar (Table 4).

Radial profiles of Rb content and Rb/K were quite similar at the four stem heights in ST7-1 (Fig. 3, left). The concentration of Rb increased inward in the sapwood

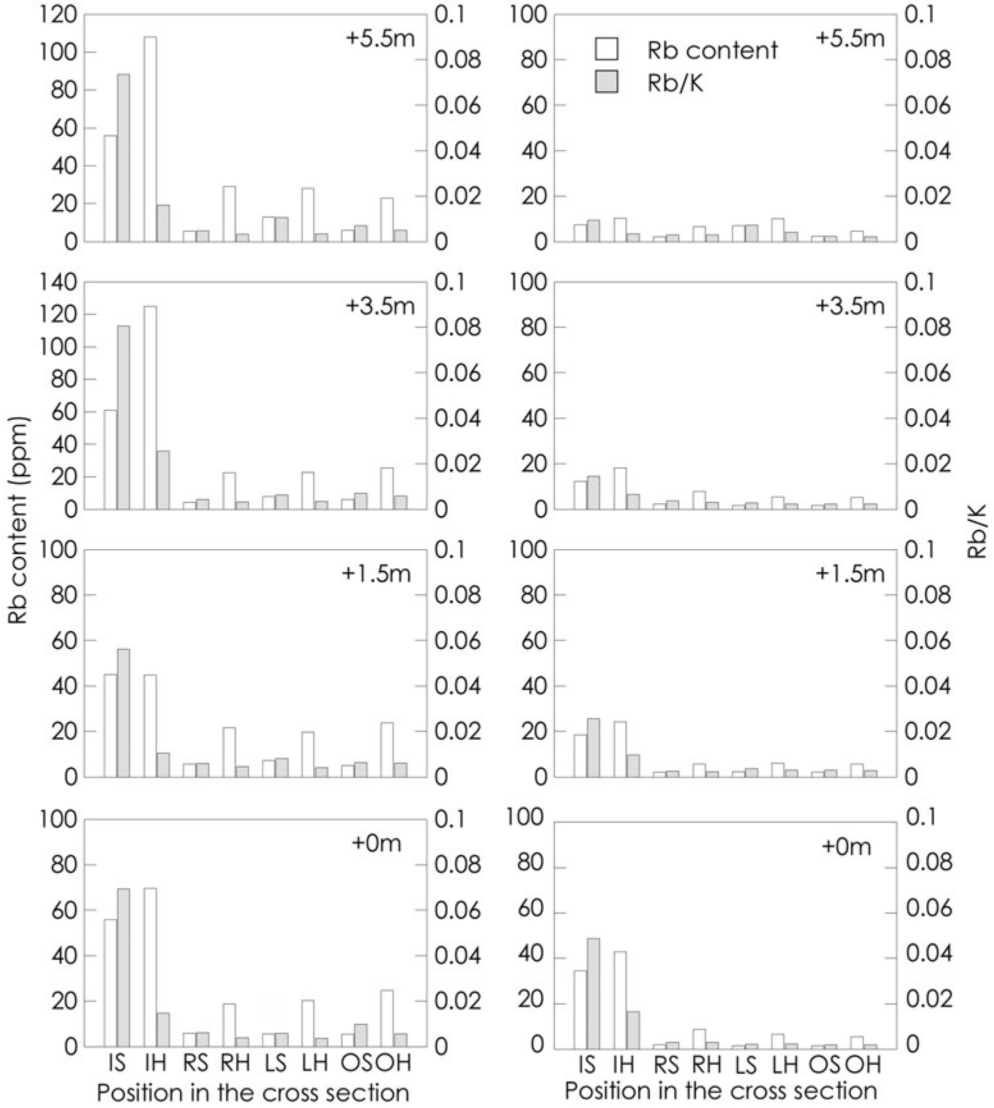

Fig. 2. Rb content and Rb/K ratio in Rb-injected trees at 40 days after the treatment. HS8-1 with wet heartwood (*left*) and MA5-1 with normal heartwood (*right*). *IS*, injection-side sapwood; *IH*, injection-side heartwood; the same pattern is followed for O, L, and R

region and became highest in the outermost heartwood; it gradually decreased inward in the heartwood region. The value at each radial position was greater than that of the control samples (Table 4). The values of Rb/K were, at all stem heights, also greater than those of the control samples and decreased from outermost sapwood to inner heartwood. All these results indicated that Rb injected in the sapwood at a height of +0 m moved into the heartwood and ascended in the conducting area of the sapwood.

The results of ST27-1 were similar to those of ST7-1. There was not much difference in Rb content in the radii from the lower two stem heights, but it became highest at the innermost sapwood or the outermost heartwood (Fig. 3, right). The Rb/K values decreased inward from the outermost sapwood at all heights, as in ST7-1. As mentioned above, specimens could not be sampled at +6 m because of wounded tissue induced by insect attack. The influence of the injury may have reached the lower part of the stem and caused high Rb contents and thereby high Rb/K values at +4 m. Even though it was not as conspicuous as for ST7-1, a comparison with the results of control samples in Table 4 also indicated the radial movement of Rb into heartwood in ST27-1.

Europium content in Eu-injected trees at 204 days after the treatment

Europium was not detected in the heartwood of either ST7-2 or ST27-2, even at 204 days after treatment (Fig. 4). The difference in Eu content between the two cultivars was much smaller than that for Rb. The concentration at 204 days after the treatment was clearly higher than that at 40 days (Table 3), especially at upper stem heights and in the cultivar with normal heartwood, ST27-2. The concentration along radii was often highest in the middle of the sapwood region and was not detected in the innermost sapwood. Such a difference in the radial profile of Eu content in sapwood compared to that of Rb is likely to be related to their different movement processes.

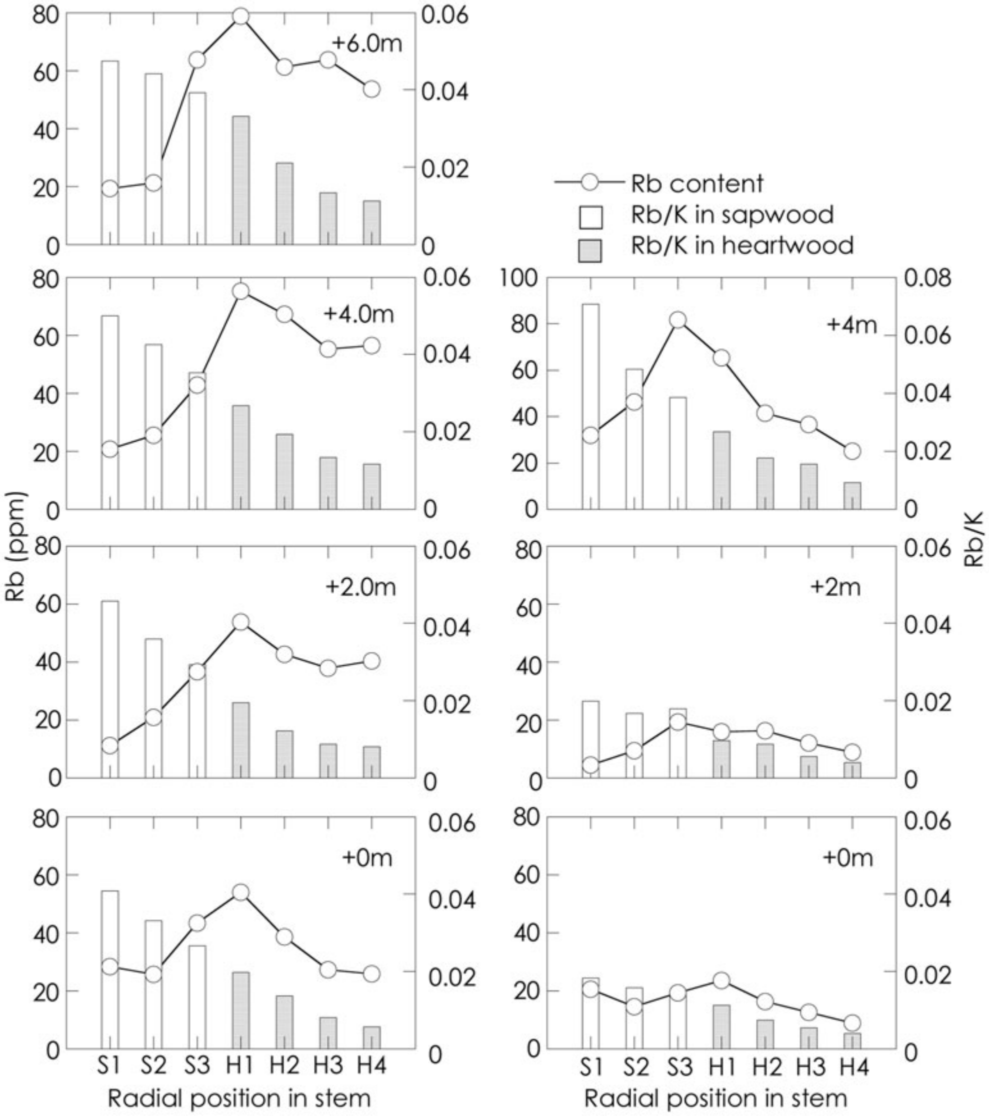

Fig. 3. Rb content and Rb/K in Rb-injected trees at 204 days after the treatment. ST7-1 with wet heartwood (*left*) and ST27-1 with normal heartwood (*right*)

Discussion

Discrimination of elements by Japanese cedar

It is evidence of the selective absorption of elements by plants that the elemental composition of plants is not the same as that of substrates where the plants grow. Our results imply that Japanese cedar discriminates beneficial elements from nonbeneficial elements not only in absorption but also in transport in its stem, especially in the radial direction. The pathway of minerals when moving up the stems of Japanese cedar is tracheids in the sapwood region, in which there is no living organ directly connecting the roots and the leaves. The process of vertical mineral transport is, therefore, not strictly regulated by the tree. Mass flow of sap and adsorption/desorption of each mineral to cell walls are the decisive factors in vertical transport. In contrast, sapwood and heartwood are connected by rays that include living parenchyma that can regulate the radial transport. Stewart[3] has pointed out an important role of rays in radial transport of materials in tree stems. Since the white zone between the sapwood and the heartwood disconnects the pathway of free water by the extremely low moisture content in tracheids, it is unlikely that minerals easily diffuse in tracheids from the sapwood to the heartwood. Ziegler[2] confirmed radial movement of ^{14}C sugar and $^{35}SO_4^{2-}$ via rays in broad-leaved tree species. He mentioned that the symplastic route was the main pathway of radial movement, and the velocity was greater than that driven by normal diffusion alone. Chaffey and Barlow[4] also claimed that ray cells were important symplastic pathways between xylem and phloem in two angiosperm trees. In our results, it is much more likely that rays play an important role in radial transport in the stem of Japanese cedar.

Japanese cedar is reported to accumulate some minerals such as alkali metals in its heartwood.[16,24] Although the reason has not yet been clarified, the minerals accumulated

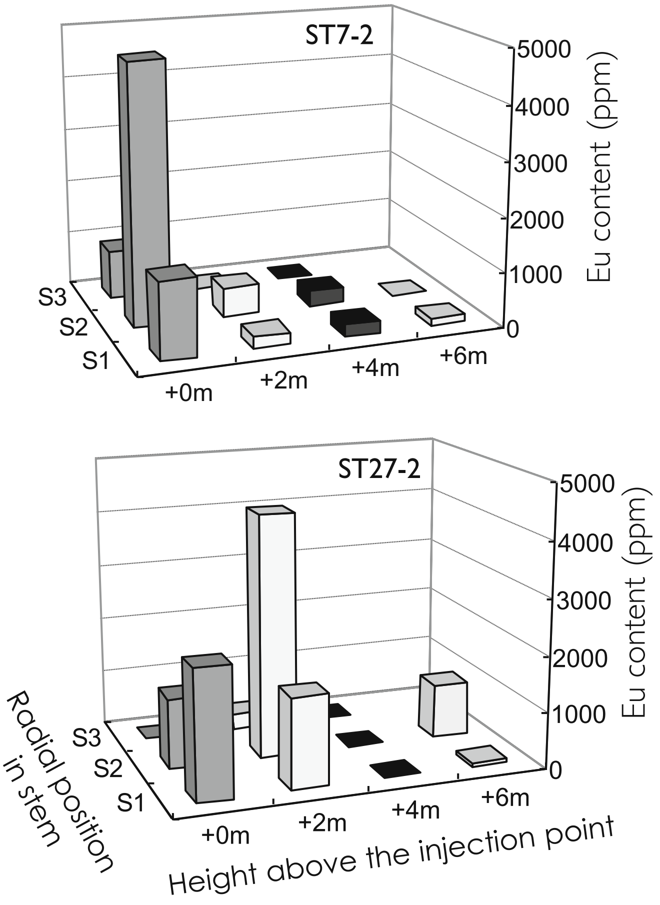

Fig. 4. Eu content in Eu-injected trees at 204 days after the treatment. ST7-2 with wet heartwood (*upper*) and ST27-2 with normal heartwood (*lower*)

in the heartwood are considered to have a close relation with heartwood formation or regulation of pH.[24] It is, therefore, reasonable that Rb, an alkali metal species, easily moved into the heartwood of Japanese cedar after it was injected in the sapwood. In contrast, no Eu was detected in the heartwood even at 204 days after the injection. The difference implies that not all minerals, but only beneficial elements, are easily transported to heartwood.

Genetic difference in radial transport of minerals

As previously mentioned, it is well known that black heartwood of Japanese cedar accumulates much more moisture and minerals than normal heartwood.[18,25] Although several hypotheses have been proposed, such as soil conditions, injuries, and the effects of microorganisms, it is still controversial as to what is the main cause of black heartwood. Genetics is no doubt an important factor because some cultivars of Japanese cedar are reported to often have black heartwood.[26] In fact, Morikawa et al.[27] reported the differences in mineral content in heartwood among three cultivars of Japanese cedar, concluding that especially high potassium content causes black heartwood. In this study, we designed the experiment to examine whether the genetic characteristics of accumulating moisture and minerals in heartwood have a certain relationship with the radial transport of minerals in stems.

Rubidium injected in sapwood moved into heartwood more rapidly in cultivars with wetter heartwood (HS8 and ST7) than in those with normal heartwood (MA5 and ST27). There are considerable differences in the background K and Rb contents in the heartwood between the former two cultivars and the latter two (Tables 2 and 4). These results indicate that individual trees genetically predisposed to accumulating more minerals in their heartwood will radially transport minerals much more rapidly than the others.

Active transport or not?

It is impossible to explain the movement of injected Rb into the heartwood by diffusion alone. To make the discussion easier, we should separate the process into two steps: (1) from the sapwood to the outer heartwood, and (2) from the outer heartwood to the inner heartwood.

At 204 days after injection, Rb content was highest at the outermost heartwood in ST7-1 (Fig. 3, left). Rb could not have diffused from the sapwood to the outermost heartwood because of the barrier of the white zone and the reverse gradient of Rb concentration. Therefore, active transport through rays is the only explanation of the movement of Rb in this region. Once Rb is transported into the heartwood, however, there are no living cells that regulate mineral transport. Simple diffusion processes resulting from the concentration gradient will dominate the distribution of Rb in the heartwood. The monotonous decreases of both Rb content and Rb/K ratio from the outer heartwood to the inner heartwood suggest this explanation. In short, the radial movement of injected Rb in Japanese cedar proceeds by two steps: active transport from the sapwood to the outermost heartwood through rays, and thereafter diffusion to the inner heartwood.

It is obvious that the above transport process continues in the resting period because a greater amount of Rb was detected in the outer heartwood of ST7-1 at 204 days after the treatment than in that of HS8-1 at 40 days after the treatment. Thus, the radial transport of some minerals in stems of Japanese cedar is probably related to certain physiological processes in the heartwood other than tree growth. In this context, the formation of black heartwood, which is associated with the accumulation of alkali metals and moisture, is likely to have a close relation with what we have discussed in this article. Further work should be oriented toward answering basic questions such as (1) what is the advantage for Japanese cedar to accumulate K in its heartwood, and (2) is water movement necessary for K transport to heartwood?

Acknowledgments We would like to thank members of the Wood Quality Laboratory, Forestry and Forest Products Research Institute (FFPRI), and Dr. Y. Fujisawa and Dr. R. Nakada, the National Forest Tree Breeding Center (now the Forest Tree Breeding Center, FFPRI), for their assistance in the field experiment in this study. We also thank Dr. Y. Kawabata, Tokyo University of Agriculture and Technology; Dr. S. Koh, Kyoto University; and the staff of the Reactor Research Institute, Kyoto University, the Institute of Atomic Energy, Rikkyo University, and the Radioisotope Research Center, Kyoto University for their assistance in the activation analysis. We also thank Dr. K. Rupa Kumar, Indian Institute of Tropical Meteorology, and Dr. L. Xiong, FFPRI, for their critical reading of an earlier draft. This study was financially supported by the Science and Technology Agency (now the Ministry of Education, Culture, Sports, Science and Technology), the Government of Japan.

References

1. Tyree MT, Zimmermann MH (2002) Xylem structure and the ascent of sap. Springer-Verlag, Berlin
2. Ziegler H (1965) Use of isotopes in the study of translocation in rays. In: International Atomic Energy Agency (ed) Isotopes and radiation in soil-plant nutrition studies. Scientific Technical Information, Publication 108, IAEA, Vienna, pp 361–370
3. Stewart CH (1966) Excretion and heartwood formation in living trees. Science 153:1068–1074
4. Chaffey N, Barlow P (2001) The cytoskeleton facilitates a three-dimensional symplasmic continuum in the long-lived ray and axial parenchyma cells of angiosperm trees. Planta 213:811–823
5. Watmough SA (1999) Monitoring historical changes in soil and atmospheric trace metal levels by dendrochemical analysis. Environ Pollut 106:391–403
6. Hagemeyer J (1993) Monitoring trace metal pollutions with tree rings: a critical reassessment. In: Markert B (ed) Plants as a biomonitors. VCH Weinheim, New York, pp 541–563
7. Bindler R, Renberg I, Jonatan K, Ove E (2004) Tree rings as Pb pollution archives? A comparison of ^{206}Pb/^{207}Pb isotope ratios in pine and other environmental media. Sci Total Environ 319: 173–183
8. Martin RR, Naftel SJ, Macfie SM, Jones KW, Feng H, Trembley C (2006) High variability of the metal content of tree growth rings as measured by synchrotron micro X-ray fluorescence spectrometry. X-Ray Spectrom 35:57–62
9. Brownridge JD (1984) The radial distribution of ^{137}Cs and ^{40}K in tree stems. J Plant Nutr 7:887–896
10. Katayama Y, Okada N, Ishimaru Y, Nobuchi T, Aoki A (1986) Behavior of radioactive nuclides in the radial direction of the annual rings of sugi (in Japanese). Radioisotopes 35:636–638
11. Kohno M, Koizumi Y, Okumura K, Mito I (1988) Distribution of environmental cesium-137 in tree rings. J Environ Radioact 8:15–19
12. Chigira M, Saito Y, Kimura K (1988) Distribution of ^{90}Sr and ^{137}Cs in annual tree rings of Japanese cedar, *Cryptomeria japonica* D. Don. J Radiat Res 29:152–160
13. Momoshima N, Bondietti EA (1994) The radial distribution of ^{90}Sr and ^{137}Cs in trees. J Environ Radioact 22:93–109
14. Kagawa A, Aoki T, Okada N, Katayama Y (2002). Tree-ring strontium-90 and cesium-137 as potential indicators of radioactive pollution. J Environ Qual 31:2001–2007
15. Lepp NW, Dollard GJ (1974) Studies on lateral movement of ^{210}Pb in woody stems. Oecologia 16:179–184
16. Taneda K, Ota M, Nagashima M (1986) The radial distribution and concentration of several chemical elements in woods of five Japanese species. Mokuzai Gakkaishi 32:833–841
17. Okada N, Katayama Y, Nobuchi T, Ishimaru Y, Aoki A (1993) Trace elements in the stems of trees V. Comparison of radial distributions among softwood stems. Mokuzai Gakkaishi 39:1111–1118
18. Oda K, Matsumura J, Tsutsumi J, Abe Z (1994) Black-heartwood formation and ash contents in the stem of sugi (*Cryptomeria japonica* D. Don) (in Japanese). Sci Bull Fac Agric Kyushu Univ 48:171–176
19. Abe Z, Oda K (1994) The color change of sugi (*Cryptomeria japonica*) heartwood from reddish brown to black II. Identification of potassium hydrogen carbonate as one of the causative materials (in Japanese). Mokuzai Gakkaishi 40:1126–1130
20. Gladney ES, Hakonson TE, Muller N (1984) Cesium as an activatable tracer for studying biological intrusion of plant roots into waste burial areas. J Radioanal Nucl Chem 84:423–429
21. Yamashita M, Takyu T, Saba T (1995) Influences of root pruning on reproduction and activity of roots in mature tea plants. Jpn J Crop Sci 64:740–746
22. Katayama Y, Okada N, Ishimaru Y, Nobuchi T, Yamashita H, Aoki A (1986) Determination of trace elements in annual rings of Yaku sugi by thermal neutron activation analysis (in Japanese). Radioisotopes 35:577–582
23. Okada N, Katayama Y, Nobuchi T, Ishimaru Y, Aoki A (1990) Trace elements in the stems of trees III. Element content in the sap and the wood substance of sugi (*Cryptomeria japonica*). Mokuzai Gakkaishi 36:1–6
24. Okada N, Katayama Y, Nobuchi T, Ishimaru Y, Yamashita H, Aoki A (1987) Trace elements in the stems of Trees I. Radial distribution in sugi (*Cryptomeria japonica* D. Don). Mokuzai Gakkaishi 33:913–920
25. Kubo T, Ataka S (1998) Blackening of sugi (*Cryptomeria japonica* D. Don) heartwood in relation to metal content and moisture content. J Wood Sci 44:137–141
26. Miyajima H (1989) Heartwood color (in Japanese). In: Kyushu no sugi to hinoki. Kyushu University Press, Fukuoka, pp 164–166
27. Morikawa T, Oda K, Matsumura J, Tsutsumi J (1996) Black-heartwood formation and ash contents in the stem of sugi (*Cryptomeria japonica* D. Don) II: heartwood properties of three sugi cultivars (in Japanese). Bull Kyushu Univ For 74:41–49