EFFECT OF THE 2011 FUKUSHIMA DAIICHI NUCLEAR POWER PLANT ACCIDENT ON *CRYPTOMERIA JAPONICA* WOOD COMPONENTS

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Received January 17, 2023

Radiocontaminated *Cryptomeria japonica* from Iitate village in Fukushima was analyzed in terms of the bark, sapwood and heartwood. Both ^{134}Cs and ^{137}Cs activities were confirmed in these wood components. The sum of ^{134}Cs and ^{137}Cs activities was the highest in the bark, the lowest in the sapwood and that of the heartwood in between. Although the holocellulose isolated from the bark was contaminated by ^{134}Cs and ^{137}Cs activities, these radiocesiums were not detected in the holocelluloses from sapwood and heartwood. Similar results were obtained for dioxane lignins isolated from sapwood and heartwood. An examination of the FTIR spectra of holocelluloses and dioxane lignins from the bark, sapwood and heartwood of *C. japonica* originated from radiocontaminated Iitate, and radiocontamination-free Fuji areas revealed that the structures of the constitutions of the former wood were not altered by the radiocontamination. Furthermore, the FTIR spectra of the outer bark and inner bark holocelluloses from the artifically ^{137}Cs-impregnated Fuji tree were similar to those of the outer bark and inner bark holocelluloses of the radiocontaminated Iitate tree.

Keywords: *Cryptomeria japonica*, bark, sapwood, heartwood, lignin, holocellulose, ^{134}Cs, ^{137}Cs

INTRODUCTION

Residential areas, agricultural soils and forests in Fukushima prefecture and its surrounding prefectures were heavily radiocontaminated by the Fukushima Daiichi nuclear power plant (FDNPP) accident in March 2011. Although the contaminated soils from residential and agricultural areas were already removed, the decontamination of forests is still in waiting.

Kato and co-workers[1] observed that a coniferous forest located 150 km southwest of the FDNPP could selectively fractionate and then transfer the deposited radiocesium in the canopy to forest floor by throughfall and stemflow. The main composition of this forest was Japanese cedar (*Cryptomeria japonica*) and cypress (*Chamaecyparis obtusa*). They also found that more than 60% of the total radiocesium deposition remained in the canopy five months after the accident, suggesting that the canopy would be a secondary source of radiocesium contamination of the forest floor. Yoschenko and co-workers[2] studied the distribution of radiocesium in a plantation of *C. japonica* located approximately 34 km northwest of the FDNPP, from May 2014 to March 2015. They found that approximately 74% of the total radiocesium inventory was located in the forest soil, 20% in the litter and the remaining in the aboveground biomass. Their results implied that most of the radiocesium initially intercepted by the forest canopy was already transferred to the ground surface. From these works, it can be suggested that the soil in the forest floor is contaminated by the fallout radiocesium from the accident, which, in turn, may be the source of the radiocontamination of the forest trees.

In a study carried out six months after the accident on the distribution of radiocesium in the bark, sapwood and heartwood of *Quercus serrata* (a hardwood located approximately 138 km west of the FDNPP), *C. japonica* (a softwood sampled from three locations at approximately 138 km west, 65 km northwest and 26 km southwest of the FDNPP) and *Pinus densiflora* (a softwood grown approximately 138 km west of the FDNPP), Kuroda and co-workers[3] found that radiocesium existed in the bark, sapwood and heartwood of the three wood species. However, for the softwoods studied, the radiocesium activities in the bark were

much lower than those in the sapwood and heartwood. The authors concluded that direct radioactive deposition was the main reason of the observed results.

On the other hand, *Quercus serrata* and *C. japonica* were harvested from a location 60 km west of the FDNPP, approximately 18 months after the accident, in order to determine the ^{137}Cs activities in the woods.[4] For both wood species studied, the ^{137}Cs activities in sapwood were higher than those of heartwood and most of the radiocesiums in the tree rings was directly absorbed from the atmosphere via bark and leaves rather than via roots.

In order to study the cesium radioactivities in stem wood of softwoods, *C. japonica* (sampled from two locations at approximately 27 km and 30 km west of the FDNPP, respectively), *Chamaecyparis obtusa* (harvested from approximately 160 km southwest of the FDNPP), and *Larix kaempferi* (a larch wood harvested from approximately 30 km west of the FDNPP) were sampled three years and a half after the accident.[5] The results suggested that the distribution of ^{137}Cs in the sapwood of the three softwoods examined was relatively uniform, but the vertical and radial distribution in the heartwoods was heterogenous.

To verify whether ^{134}Cs and ^{137}Cs were absorbed through bark and then translocated into stem wood, Wang and co-workers[6] studied the movement of ^{133}Cs within the softwood *C. japonica* sampled two years and four months after the accident from two places located at 37°21’26.0’’N, 140°20’42.8’’E and 36°46’25.5’’N, 139°49’16.9’’E, respectively. They concluded that ^{133}Cs penetrated the bark to enter the wood and mostly accumulated in the heartwood. Furthermore, Iizuka and co-workers[7] studied the relationship between ^{137}Cs activities and potassium content in the stem wood of *C. japonica* one year and ten months after the accident. The wood sample was taken from a plantation located approximately 130 km southwest of the FDNPP. Their results showed that the ^{137}Cs activities peaked at the heartwood-sapwood boundary and gradually decreased toward the pith and that the heartwood-to-sapwood ratio of ^{137}Cs activities was significantly positively correlated with potassium content.

The research works mentioned above showed the contamination effect of the fallout radiocesiums from the FDNPP accident on the forest canopy, bark, sapwood and heartwood of softwoods, as well as hardwoods in the forest. However, the effects of the fallout radiocesiums on the main wood components, such as cellulose, hemicelluloses and lignin, were not mentioned in the cited literature. Furthermore, there is no work so far on the radiocontamination of cellulose, hemicelluloses and lignin. To partly fulfill the gap, in this study, the activities of ^{134}Cs and ^{137}Cs in the bark, sapwood and heartwood, as well as in the dioxane lignins and holocelluloses of these wood components from a *C. japonica* (Japanese name: sugi) tree grown in Iitate village, Fukushima prefecture, were determined and the influences of radiocesiums on the FTIR spectra of the isolated holocelluloses and lignins were examined. A *C. japonica* tree in the radiocontamination-free Fuji city, Shizuoka prefecture, was randomly felled and used for comparison with that from Iitate village.

EXPERIMENTAL

Tree samples

A *C. japonica* tree having 41 annual rings, from Iitate village, Fukushima prefecture, located approximately 33 km northwest of the FDNPP, was randomly felled on 10 July 2017 from a private plantation. This sample was designated as Iitate *C. japonica.* For comparison, a *C. japonica* tree with 56 annual rings in Fuji city, Shizuoka prefecture, located approximately 330 km southwest of the FDNPP, was cut down randomly on 3 December 2017, from another private plantation. Similar to the Iitate tree, this sample was named Fuji *C. japonica.* These two private plantations produce *C. japonica* for commercial purposes.

For both samples, the part below the height of 1.7 m and above 20 cm of the soil surface was used. The sizes of the sapwood and heartwood of each sample are shown in Figure 1, which presents photocopies of the lower surfaces of the sample trunks.

Each log sample was further cut off into round pieces of 4-6 cm thickness. For each wood sample, 12 round pieces were individually weighed and each piece was then separated into bark, sapwood and heartwood. The bark of the Fuji *C. japonica* was further separated into outer and inner barks. The bark of the Iitate *C. japonica* was composed of only one layer. The separated barks, sapwoods, and heartwoods were weighed and their compositions tabulated. The barks, sapwoods, and heartwoods were then milled in a Wiley mill and the wood meals in the 40-60 mesh interval were used for analytical determinations.

Analytical procedure

Wood meals from the bark, sapwood and heartwood were individually extracted with ethanol-benzene (1:2, v/v) in a Soxhlet apparatus for 16 hours. They were then filtered, washed successively with ethanol-benzene (1:2, v/v) and ethanol (95%) and air-dried. The extracted wood samples were used for the preparation of holocellulose, dioxane lignin and Klason lignin

determination.

Ash in the unextracted wood meals was determined at 525±25 °C for 3 hours (Tappi Standard T211 om). Klason lignin was determined using the 72% sulfuric acid method, as described in the literature.[8]

Holocellulose (cellulose and hemicelluloses) was prepared from the extractive-free wood meals, using the acid chlorite method described in the literature.[9]

Dioxane lignin was isolated from the extractive-free wood meals using the method described in the literature.[10]

Iron, silica, potassium magnesium, calcium in the bark, sapwood and heartwood of the Iitate *C. japonica* were determined by the X-ray fluorescence analytical technique using a ZSX Primus IV from Rigaku Corporation.

FTIR spectra of dioxane lignins and holocelluloses were recorded using a Jasco (Nihon Bunko) FT/IR-6100 Fourier transform infrared spectrometer.

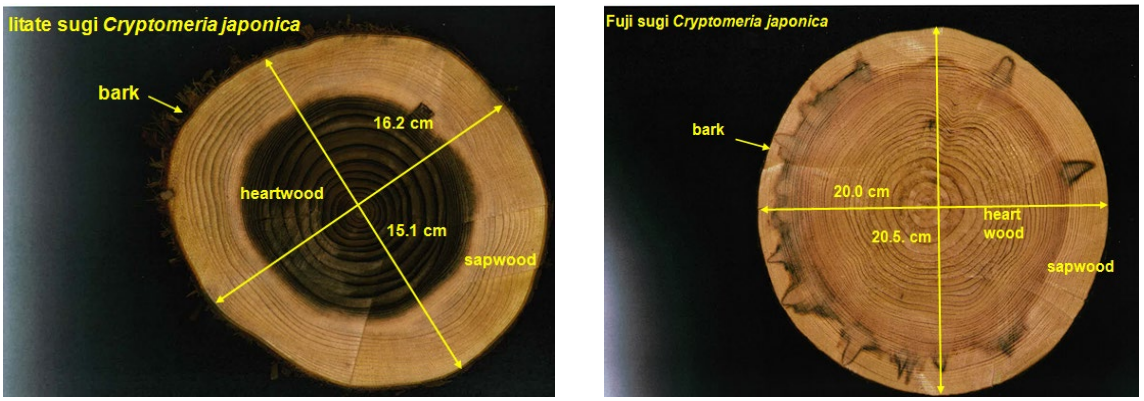

Figure 1: Surfaces of Iitate and Fuji *C. japonica* samples

Treatment of radiocontaminated paddy soil with sodium chlorite and 1,4-dioxane

Sodium chlorite treatment

A radiocontaminated paddy soil in Iitate village (0.5-1.0 mm soil, 40 g oven-dried) was mixed with 800 mL of distilled water in a glass flask. 5 mL of acetic acid was added and mixed. 8 g of sodium chlorite was added and mixed again. The mixture was then refluxed in a water bath at 70 °C for 1 hour. The step of addition of acetic acid and sodium chlorite was repeated at the second, third and fourth hour. Finally, the flask was cooled to room temperature, filtered on a no. 6 filter paper and washed well with water. The treated soil was first dried at room temperature then at 105 °C, prior to the determination of ^{134}Cs and ^{137}Cs.

1,4-Dioxane treatment

A similar soil sample as above (0.5-1 mm soil, 40 g oven-dried) was mixed with 200 mL of 1,4-dioxane. The mixture was acidified with 1.5 mL of 1N hydrochloric acid, then refluxed in a water bath at 70 °C for 2 hours. After cooling down to room temperature, the mixture was filtered on a 0.4 µm filter paper and washed well first with a dioxane-water mixture (1:2) and then with water. The treated soil was first dried at room temperature, then at 105 °C, before subjecting to the determination of ^{134}Cs and ^{137}Cs.

^{137}Cs absorption by Fuji *C. japonica* wood components

The following wood components of the Fuji *C. japonica* were employed in the absorption of ^{137}Cs: outer bark, inner bark, sapwood, heartwood, holocelluloses from these four wood components as well as dioxane lignins from sapwood and heartwood.

The ^{137}Cs isotope was purchased from Japan Radioisotope Association in chloride form in 0.1N HCl containing 100,000 Bq mL^{-1}. A solution of 18 Bq mL^{-1} was prepared and 120 mL of the resulting radiotracer solution were added to 8 g oven-dried of each wood component in 175 mL Corning graduated conical tubes. Due to the small amounts of dioxane lignin preparations, 3 g of lignins were used in the radiocontamination test. These tubes were shaken in an agitator at 25 °C for 24 h. They were then filtered on double Advantec filter papers No. 2 and No. 6. The radiocontaminated wood components were first air-dried, then at 105 °C overnight prior to the determination of their ^{137}Cs activities.

^{134}Cs and ^{137}Cs activities were determined using a Canberra coaxial Ge detector GC2020-7500SL-2002CSL, the software Spectrum Explorer (Genie 2K), as well as the guidance of the Japanese Ministry of Health, Labor and Welfare "Radiation Measurement Manual for Food in an Emergency" March 2002, and the Ministry of Education, Culture, Sports, Science and Technology, Measurement Series No. 7 "Gamma Ray Spectrometry with Germanium Semiconductor Detector". For Iitate *C. japonica*, ^{134}Cs and ^{137}Cs activities were corrected to the date of 15 March 2011.[11]

RESULTS AND DISCUSSION

Characteristics of Iitate *C. cryptomeria*

Visually, the color of the heartwood of Fuji *C. japonica* was light reddish, whereas that of Iitate *C.*

japonica black. *C. japonica* is an industrial tree used for housing in Japan. Thus, the black color of its heartwood would degrade its economic value. It was suggested that high potassium and moisture contents of the heartwood of *C. japonica* might be the cause of its black color.[12,13]

For Iitate *C. japonica*, the potassium content increased from bark towards heartwood (Table 1). A similar trend was observed for magnesium content. On the other hand, the contents of calcium, silica and iron decreased from bark to heartwood. It is wondered then whether, besides the effect of potassium, magnesium, calcium, silica and iron could be involved in the formation of the black color of heartwood of Iitate *C. japonica*. More works are required to clarify the cause of this black color.

Another characteristic of Iitate *C. cryptomeria* is that it had only outer bark, whereas Fuji *C. cryptomeria* did have both outer and inner barks. Outer and inner barks are formed by the vascular cambium. The outer bark is the outer layer of the phloem, where the proper arrangement of cells and the pattern of the hard bast, in particular, is disturbed by active dilatation growth.[14] The outer bark is mostly dead tissue. The inner bark is the living part of the bark inside the innermost cork and consisted of non-collapsed secondary phloem tissue. Since the inner bark was not observed in Iitate *C. cryptomeria*, it can be inferred that a part of its phloem was dead. However, whether the cause of this phenomenon was due to the radiocontamination in March 2011 or not is uncertain. Furthermore, it is also unknown whether the young age of the sampled tree (35-year rings in 2011), the cold climate of Iitate village and/or the soil properties of Iitate village etc. have any influence on the formation of the single bark of the Iitate *C. cryptomeria*. Any research on this subject would be very valuable for the plantation of *C. cryptomeria.*

Table 1
Inorganics in ashes from bark, sapwood, heartwood of Iitate *C. japonica*

Inorganics (%)	Bark	Sapwood	Heartwood
K (as K_2O)	0.032	0.120	0.491
Ca (as CaO)	0.787	0.280	0.177
Mg (as MgO)	0.032	0.018	0.057
Si (as SiO2)	0.096	0.011	0.016
Fe (as Fe2O3)	0.014	0.003	0.003

Analysis of Iitate and Fuji *C. cryptomeria*

As shown in Figure 1, the average ratio of the width of heartwood to sapwood in both vertical and horizontal directions of Iitate *C. japonica* was 0.64, whereas that from Fuji *C. japonica* – 0.83. The ratio of these values was 0.77, which, in turn, was similar to the ratio of the annual rings of Iitate *C. japonica* to Fuji *C. japonica* of 0.73. The results suggested that the smaller area of heartwood in Iitate *C. japonica* was due to its lower age.

For both Iitate and Fuji *C. japonica* trees, the compositions of heartwood and sapwood were higher than 50% and 30%, respectively, whereas that of the bark below 5%. Conversely, the ash content in the bark was higher than those in sapwood and heartwood (Table 2). Regardless of the origin of *C. japonica*, the content of ethanol-benzene extractives was highest in the heartwood, followed by the bark and sapwood in that order.

For both *C. japonica* trees, the holocellulose contents in sapwood and heartwood were higher than in the barks. Since the holocellulose contents in the bark, sapwood and heartwood of Fuji *C. japonica* were higher than those of the corresponding component from Iitate *C. japonica*, it is understandable that the Klason lignin contents of the components from the former tree were lower than those of the latter. The reason of the higher Klason lignin content in the bark, sapwood and heartwood of Iitate *C. japonica* is attributable to the higher extractive contents in these wood components of this tree. To overcome the interference of wood extractives, it is recommended that, prior to the determination of Klason lignin, the wood should be extracted firstly with ethanol-benzene (1/2, v/v), secondly with 95% ethanol and thirdly with hot water.[15] Because the ethanol-benzene extraction step was solely carried out in this study, the high Klason lignin contents of the wood components from Iitate *C. japonica* are understandable.

Compared to the Klason lignin contents in sapwood and heartwood from both Iitate and Fuji *C.*

japonica, those in the barks were much higher. This might be due to the higher extractive contents in the barks of both woods, which, in turn, cannot be extracted sufficiently and/or completely by the ethanol/benzene mixture alone. Krogell and co-workers[16] opinionated that, because the Klason lignin determination method is designed solely for the wood, it may not be suitable for the lignin in the bark.

To determine the Klason lignin and polysaccharide contents in the woody bark, a more stringent extraction regime is recommended.[17] The extraction protocol is composed of: 1) diethyl ether extraction to remove waxes, fatty acids, fats, resin acids, phytosterols, terpenes; 2) ethanol extraction to remove condensed tannins, flavonoids, phenolics; 3) hot water extraction to remove condensed tannins and water-soluble carbohydrates; and 4) 1% aqueous sodium hydroxide to remove phenolic acids, hemicelluloses, and suberin monomers. The effectiveness of this scheme should be confirmed by further research.

Radiocesium activities in the bark, sapwood and heartwood of Iitate *C. cryptomeria*

Determination of radiocesium activities showed that ^{134}Cs and ^{137}Cs activities were decreased from bark to heartwood and then to sapwood (Table 2). The results are different from those of Kuroda and co-workers,[3] who found that ^{137}Cs activities in the bark were much lower than those in sapwood and heartwood. They are also not in agreement with those of Mahara and co-workers,[4] who found ^{137}Cs activities in sapwood to be higher than in heartwood.

The ratios of ^{134}Cs to ^{137}Cs were 0.73, 0.77 and 0.68 for the bark, sapwood and heartwood, respectively. These values were much lower than the ratio of 1:1 from the fallout of the FDNPP accident on March 11, 2011, by reactor units 1, 2 and 3.[11] This suggested that ^{134}Cs and ^{137}Cs were metabolized in different ways in the bark, sapwood and heartwood of Iitate *C. japonica*.

A plot of the total activities of ^{134}Cs and ^{137}Cs against the content of ash, iron, and silica showed that they all had positive relationships (Figs. 2, 3, 4). On the other hand, a straight-line relationship was found for magnesium content and the ratio of ^{134}Cs to ^{137}Cs activities in the bark, sapwood and heartwood (Fig. 5). Due to the finding that the higher magnesium content the lower the ratio of ^{134}Cs to ^{137}Cs activities, it seemed that ^{134}Cs was more attractive to magnesium than was ^{137}Cs. However, neither the total activities of ^{134}Cs and ^{137}Cs, nor the ratio of ^{134}Cs to ^{137}Cs activities have any relationship with the content of potassium or calcium in the bark, sapwood and heartwood. It is believed that, since cesium and potassium are in the column 1A of the periodic table of elements, they may exchange ions to each other. Furthermore, there was no relationship between the potassium content and ^{134}Cs, ^{137}Cs activities in the bark, sapwood and heartwood. This is notably due to the lowest contents of potassium, as well as the lowest activities of ^{134}Cs and ^{137}Cs in the sapwood. The results suggest that the metabolism of potassium and radiocesiums in sapwood (living cells) is different from those in the bark and heartwood, both of which are of dead cells.

Table 2
Trunk weight composition and ^{134}Cs, ^{137}Cs activities of bark, sapwood and heartwood from Iitate and Fuji *C. japonica*

Cryptomeria japonica	Iitate	Iitate	Iitate	Fuji	Fuji	Fuji	Fuji
Component	Bark	Sapwood	Heartwood	Outer bark	Inner bark	Sapwood	Heartwood
Composition (%)	4.9	41.4	53.7	2.1	0.4	30.7	66.8
Ash (%)	1.60	0.65	1.08	1.00	3.18	0.54	0.45
EtOH/benzene extract. (%)	4.5	3.3	7.5	2.2	5.3	0.8	2.8
Holocellulose (%)	49.2	58.1	59.2	45.6	54.6	66.3	66.4
Klason lignin (%)	48.7	35.9	39.6	45.5	42.1	28.4	31.3
Dioxane lignin (%)	----	10.1	11.1	----	----	9.2	9.0
^{134}Cs (Bq.kg^{-1})	2,167 ±37	151±8	443±11	ND	ND	ND	ND
^{137}Cs (Bq.kg^{-1})	2,961±51	195±10	656±17	ND	ND	ND	ND
^{134}C/s^{137}Cs	0.73	0.77	0.68				

ND: not detected; radioactivity was corrected to the date of 15 March 2011

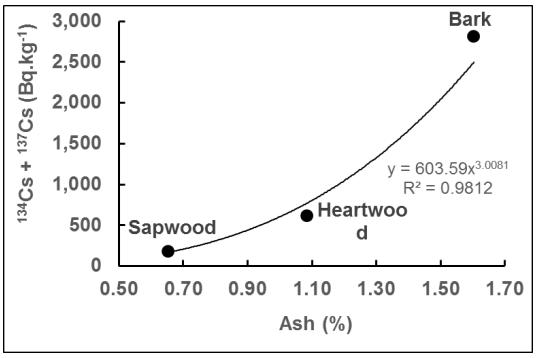

Figure 2: Relationship between cesium radioactivities and ash content in the bark, sapwood and heartwood of Iitate *C. japonica*

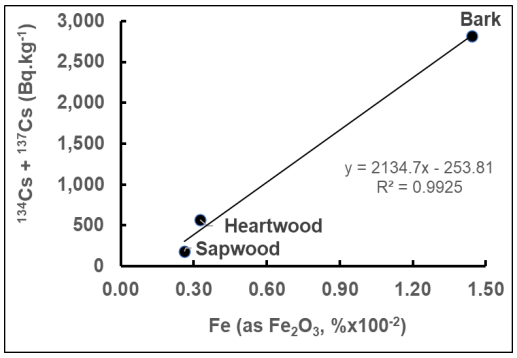

Figure 3: Relationship between cesium radioactivity and iron content in the bark, sapwood and heartwood of Iitate C. japonica

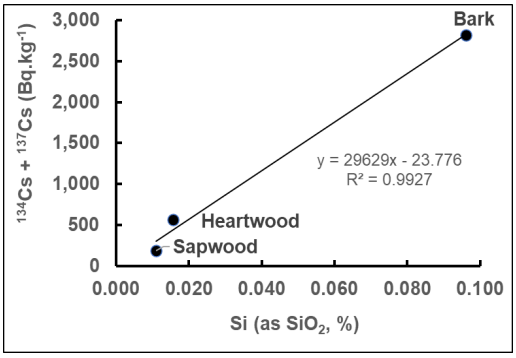

Figure 4: Relationship between cesium radioactivities and silica content in the bark, sapwood and heartwood of Iitate *C. japonica*

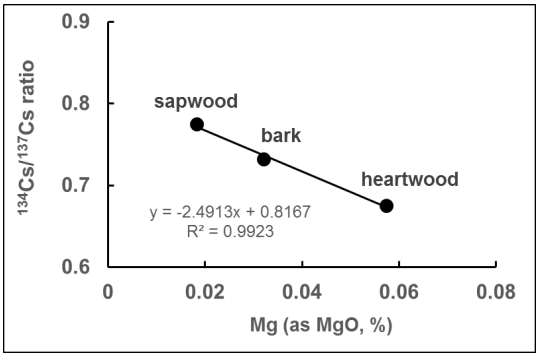

Figure 5: Relationship between $^{134}Cs/^{137}Cs$ ratios and magnesium content in the bark, sapwood and heartwood of Iitate *C. japonica*

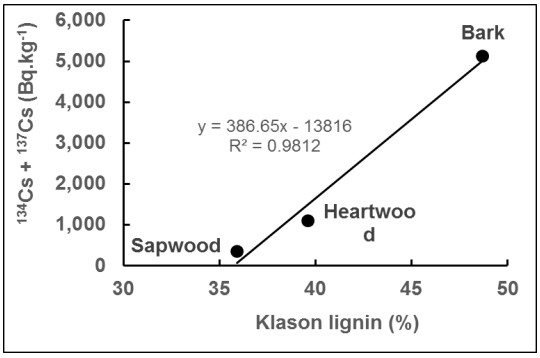

Figure 6: Relationship between cesium radioactivities and Klason lignin content in the bark, sapwood and heartwood of Iitate *C. japonica*

Radiocesium activities in the holocellulose and lignin of Iitate *C. cryptomeria*

Prior to the isolation of holocellulose and dioxane lignin or the determination of Klason lignin, bark, sapwood and heartwood of both Iitate and Fuji *C. japonica* were extracted with an ethanol/benzene mixture.

Since the acid ionization constant K_a (1×10^2) of sulfuric acid is low (*i.e.* as compared to $K_a = 2\times10^9$ of hydrochloric acid), it is a weak acid. Therefore, it is expected that only a small part of minerals, such as potassium, calcium, magnesium, silica and iron, would be soluble in 72% sulfuric acid used in the Klason lignin determination. In other words, potassium, calcium, magnesium, silica and iron existing in the bark, sapwood and heartwood of Iitate *C. japonica* would be retained, to a great extent, in the lignins and holocelluloses of these components. Metal ions, such as iron, copper, zinc, manganese, calcium, barium, magnesium, potassium, sodium *etc*., are able to sorb to the bark, sapwood and heartwood of spruce wood through ion exchange by complexion with the functional groups, such as carboxyl groups and phenolic

hydroxyl groups in the wood.[18,19] Furthermore, our previous results[20] also suggested that radiocesiums could exchange cations with calcium, magnesium, potassium, copper and barium in radiocontaminated paddy soil. Thus, these minerals would also be able to exchange cations with ^{134}Cs and ^{137}Cs activities in the wood components (Figs. 2-5). Because Ca^{2+} and Mg^{2+} were found to form lignin-metal complexes,[21] ^{134}Cs and ^{137}Cs are expected to be bound to the lignin of Iitate *C. japonica* through its potassium, calcium, magnesium, silica and iron. This would explain the results of Figure 6, showing the relation between the Klason lignin content and the sum of ^{134}Cs and ^{137}Cs activities, specifically, the higher the Klason lignin content, the higher the sum of ^{134}Cs and ^{137}Cs activities of the wood component.

Table 3 shows that, for Iitate *C. japonica*, the ethanol-benzene extraction removed more ^{134}Cs than ^{137}Cs activities from the bark, sapwood and heartwood. In particular, ^{134}Cs activities in sapwood were completely removed. The results suggest that ^{134}Cs and ^{137}Cs activities in the living cells of sapwood are more susceptible to ethanol-benzene extraction than those in dead cells of heartwood and bark.

The acidic chlorite treatment used in the isolation of holocellulose removed approximately 29% of ^{134}Cs and 40% of ^{137}Cs activities in the extractive-free bark. However, ^{134}Cs and ^{137}Cs activities in extractive-free sapwood and heartwood were completely removed by the acidic chlorite treatment. As suggested above, minerals such as potassium, calcium, magnesium, silica and iron are present in lignin. Thus, they may also exist in the holocellulose and exchange ions with ^{134}Cs and ^{137}Cs activities in these wood components. Recent research[22] indicated that cesium chloride was able to form complexes with D-arabinose, L-arabinose, and lactose at the hydroxyl groups of these sugars. Suppose that ^{134}Cs and ^{137}Cs were to form complexes with the hydroxyl groups of arabinose, xylose, mannose, galactose and glucose in hemicelluloses and cellulose of Iitate *C. japonica*, the weak acidity of the chlorite solution used in the extraction of holocellulose would not be able to break ^{134}Cs and ^{137}Cs apart from those complexes. Thus, our hypothesis that the cation exchange of minerals, such as potassium, calcium, magnesium, silica and iron, in holocellulose with ^{134}Cs and ^{137}Cs left these radiocesiums vulnerable to be removed by the acidic chlorite treatment. Similar conclusions can be applied to the undetected ^{134}Cs and ^{137}Cs activities in the dioxane lignins isolated from sapwood and heartwood of Iitate *C. japonica*.

Whereas holocelluloses and dioxane lignins from extractive-free sapwood and heartwood contained no radiocesium activities, holocellulose from extractive-free bark was still radiocontaminated. The results suggest that these observations may be due to the lower radiocesium activities in the holocelluloses and dioxane lignins of sapwood and heartwood, or to the insufficient amount of the acidic chlorite used for the bark and/or the radiocesiums were bound more tenaciously to the bark than to sapwood and heartwood.

Table 3
Effects of ethanol/benzene, sodium chlorite and dioxane on the removal of radiocesium from the components of Iitate *C. japonica*

Radioactivities ($Bq.kg^{-1}$)	^{134}Cs	^{137}Cs	$^{134}Cs+^{137}Cs$	$^{134}Cs/^{137}Cs$
Bark before EtOH/φ extraction	2,167±37	2,961±51	5,128±82	0.73
after EtOH/φ extraction	1548±33	2,385±48	3,933±82	0.65
radiocesium removal (%)	28.6	19.5	20.3	
Sapwood before EtOH/φ extraction	151±33	195±10	346±17	0.77
after EtOH/φ extraction	ND	157±10	157±10	------
radiocesium removal (%)	100	19.5	54.6	------
Heartwood before EtOH/φ extraction	443±11	656±17	1,099±30	0.68
after EtOH/φ extraction	418±11	653±11	1,072±30	0.64
radiocesium removal (%)	5.6	0.0	2.5	
Holocellulose from extractive-free bark	1,104±28	1,421±31	2,562±50	0.78
Radiocesium removed by $NaOCl_2$ (%)	28.7	40.4	34.9	
Holocellulose from extractive-free sapwood	ND	ND	ND	
Holocellulose from extractive-free heartwood	ND	ND	ND	
Dioxane lignin from extractive-free sapwood	ND	ND	ND	
Dioxane lignin from extractive-free heartwood	ND	ND	ND	

Notes: 1) ND: not detected; 2) Radioactivities were corrected to the date of 15 March 2011

Table 4
Effects of sodium chlorite and dioxane treatments on the removal of ^{134}Cs and ^{137}Cs from Iitate radiocontaminated paddy soil

Radioactivities (Bq.kg^{-1})	^{134}Cs	^{137}Cs	$^{134}Cs + ^{137}Cs$
Original soil	1,233±13	14,317±62	15,550 ± 64
Sodium chlorite treatment	1,251±15	14,139±69	15,390 ± 70
Radiocesium removal (%)	----	1.2	1.0
Dioxane treatment	1,302±15	15,363±76	16,665 ± 76
Radiocesium removal (%)	----	----	----

Note: Radioactivities were not corrected to the date of 15 March 2011

In order to confirm whether the acidic chlorite and dioxane treatments would remove radiocesium activities from the holocellulose and dioxane lignin preparations, a radiocontaminated paddy soil from Iitate village was extracted with acidic chlorite and dioxane. The results are given in Table 4.

Compared to the original soil, the chlorine dioxide treatment did not affect ^{134}Cs and ^{137}Cs activities of the soil to any extent. However, the acidic dioxane did increase the apparent activities of ^{134}Cs and ^{137}Cs of the soil. Thus, it can be expected that the solvents used in the isolations of holocelluloses and dioxane lignins would not affect the ^{134}Cs and ^{137}Cs activities in the bark, sapwood and heartwood of Iitate *C. japonica*. These results would further support our hypothesis that minerals, such as potassium, calcium, magnesium, silica and iron, in holocellulose and lignin would exchange cations with ^{134}Cs and ^{137}Cs in these wood components and the ionized ^{134}Cs and ^{137}Cs would be displaced by the acidic chlorite and dioxane used.

FTIR analysis of Iitate and Fuji *C. cryptomeria* holocelluloses and lignins

FTIR spectra of bark holocelluloses

The assignments of the absorption bands found in the FTIR spectra of holocelluloses obtained from the barks of Iitate and Fuji *C. japonica* are shown in Table 5. A large and broad band at 3375-3377 cm^{-1} was present in the spectra of the bark of the Iitate, as well as in both inner and outer bark of the Fuji *C. japonica*. This band is assigned to the O-H stretching vibration. The band at 2921 cm^{-1} in the bark of the Iitate tree and the shift of the band to the 2895 cm^{-1} in the inner and outer barks of the Fuji tree correspond to the C-H stretching vibration.

Whereas there was no band at 2360 cm^{-1} in the inner and outer barks of the Fuji tree, the bark of the Iitate tree did have such a band, which, in turn, was similar to the band at 2350 cm^{-1} in the spectrum of *Leucaena leucocephala* bark. This band is assigned to carbon dioxide.[23]

The band at 1727 cm^{-1} in the spectrum of the Iitate tree bark was much stronger than those in the inner and outer bark of the Fuji tree. This band is assigned to the C=O stretch of acetyl and carbonyl groups in hemicelluloses.[24] Both holocelluloses from outer and inner barks of the Fuji tree contained a band at 1643 cm^{-1}, which is assigned to the O-H bond of adsorbed water.[23,25] This band might be displaced to 1626 cm^{-1} in the spectrum of the Iitate tree.

The holocelluloses from both the inner and outer barks of the Fuji tree contained a band at 1427 cm^{-1}. However, this band was a shoulder in the spectrum of holocellulose from the bark of Iitate tree. Both the holocelluloses from the barks of Fuji and Iitate trees also possessed a band at 1372-1374 cm^{-1}. These two bands at 1427 cm^{-1} (or 1409 cm^{-1}) and 1372-1374 cm^{-1} are assigned to the asymmetric and symmetric C-H deformation, respectively.[25] The bands at 1158 cm^{-1} and 1159 cm^{-1} in the respective holocelluloses from the barks of the Iitate and Fuji tress are assigned to the C-O-C symmetric stretching of cellulose and hemicelluloses.[24] Both holocelluloses from the barks of the Iitate and Fuji trees contained a band at 1063 cm^{-1}, which is the C=O stretching in both cellulose and hemicelluloses.[24,25] All the holocelluloses of the barks from the Iitate and Fuji trees contained a band at 898 cm^{-1}. Whereas this band is assigned to the C=O groups relating to the β-glycosidic linkage,[23] it is also assigned to the stretching of glucose ring with C_1-H deformation.[24,25] Therefore, the assignment of this band should be studied further.

Since the literature on the FTIR spectra of holocelluloses from bark is scarce, these spectra from the original and ^{137}Cs-impregenated outer and inner barks of the Fuji *C. japonica* are given in Figures 7 and 8, respectively.

Table 5
FTIR absorption bands of holocelluloses isolated from barks, sapwoods and heartwoods of Iitate and Fuji *C. japonica*

Wood component / Functional groups	FTIR bands (cm^{-1})						
	Iitate bark	Fuji outer bark	Fuji inner bark	Iitate sapwood	Fuji sapwood	Iitate heartwood	Fuji heartwood
O-H stretching vibration	3375	3377	3377	3355	3355	3379	3352
C-H stretching vibration	2921	2895	2895	2898	2897	2896	2898
CO_2	2360						
C=O stretching of acetyl and carbonyl groups in hemicelluloses	1727	1727	1727	1727	1727	1727	1727
O-H bond of adsorbed water	1626	1643	1643	1636	1636	1635	1612
Asymmetric and symmetric C-H deformation	1409	1427	1427	1427	1427	1427	1427
Unknown				1260	1259	1261	1258
C-O-C symmetric stretching	1158	1159	1159	1160	1160	1159	1159
C=O stretching of cellulose and hemicelluloses	1063	1063	1063	1062	1061	1062	1060
Unknown	898	898	898	898	898	898	898

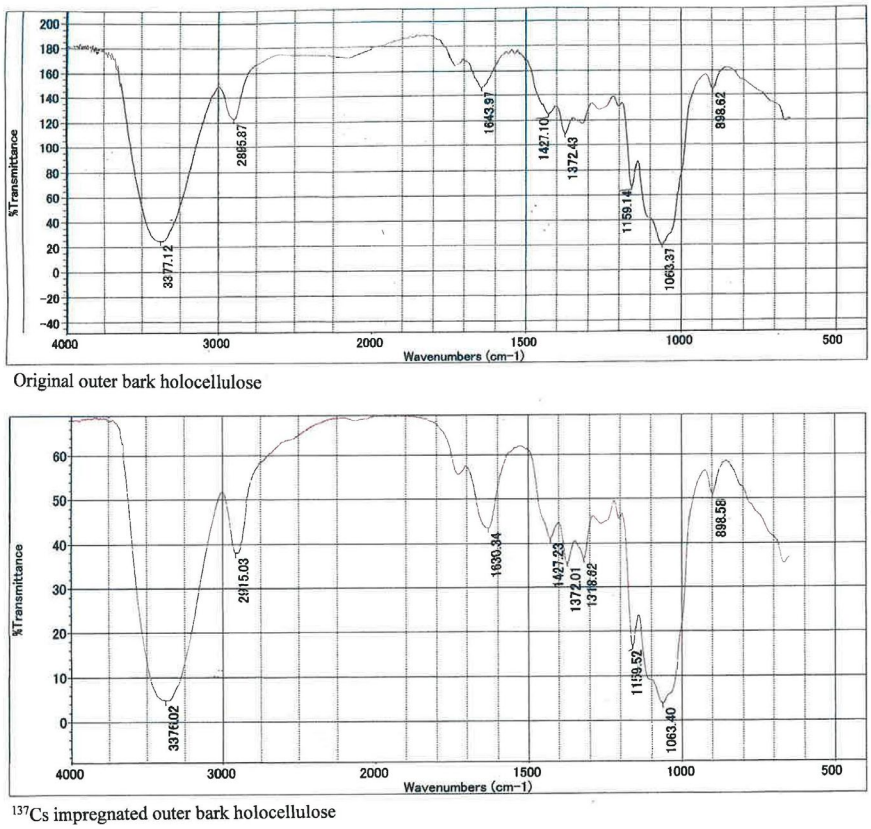

Figure 7: FTIR spectra of holocelluloses from the original and ^{137}Cs-impregnated outer barks of Fuji *C. japonica*

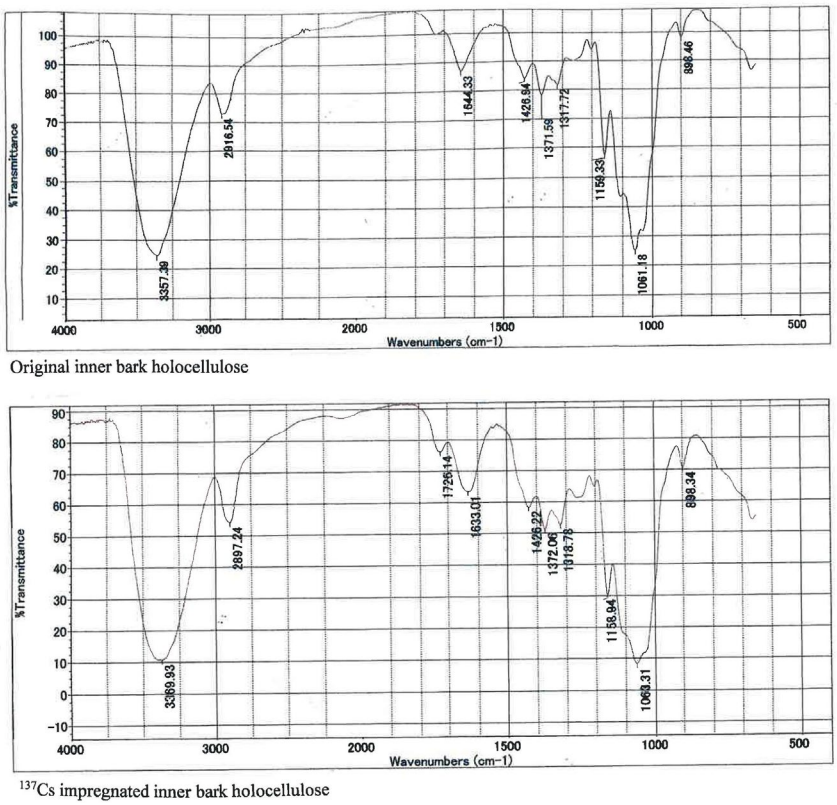

Figure 8: FTIR spectra of holocelluloses from the original and ^{137}Cs-impregnated inner barks of Fuji *C. japonica*

FTIR spectra of sapwood and heartwood holocelluloses

The assignments of the absorption bands found in the FTIR spectra of the holocelluloses obtained from sapwoods and heartwoods of Iitate and Fuji *C. japonica* are shown in Table 5. A comparison with the FTIR spectrum of the holocellulose from the bark of Iitate tree reveals that the broad band at 3375 cm^{-1} in the spectrum of bark holocellulose was shifted to the band at 3355 cm^{-1} in the sapwood holocellulose and then to 3379 cm^{-1} in the spectra of heartwood holocellulose. However, for the Fuji tree, the broad band at 3377 cm^{-1} in the spectra of both holocelluloses from inner and outer barks were shifted to the band at 3355 cm^{-1} in the spectrum of sapwood holocellulose and that at 3352 cm^{-1} in the spectrum of heartwood holocellulose.

Similarly, the band at 2921 cm^{-1} in the spectrum of Iitate bark holocellulose was gradually shifted to 2898 cm^{-1} and 2896 cm^{-1} in the sapwood and heartwood holocelluloses, respectively. Meanwhile, the band at 2895 cm^{-1} in the spectrum of holocellulose from the bark of the Fuji tree was gradually shifted to 2897 cm^{-1} and 2898 cm^{-1} in the sapwood and heartwood holocelluloses, respectively.

The band at 1727 cm^{-1} was found in all the spectra of the bark, sapwood and heartwood holocelluloses of both Iitate and Fuji trees.

Concerning the Iitate tree holocellulose, the band at 1643 cm^{-1} in the bark was shifted to 1636 cm^{-1} in the sapwood and 1635 cm^{-1} in the heartwood. On the other hand, the band at 1643 cm^{-1} in the Fuji tree bark holocellulose was shifted to 1636 cm^{-1} in the sapwood and 1612 cm^{-1} in the heartwood.

The shoulder at 1427 cm^{-1} in the spectrum of the bark holocellulose from the Iitate tree was observed as weak bands in the spectra of holocelluloses prepared from its sapwood and heartwood.

The band at 1427 cm^{-1} appeared in all the spectra of holocelluloses from bark, sapwood and heartwood of both Iitate and Fuji trees. Similarly, holocelluloses in the bark, sapwood and heartwood of both Iitate and Fuji trees contained a band at 1371-1374 cm^{-1}. Likewise, the bands at 1160 cm^{-1}, 1061-1063 cm^{-1}, 898 cm^{-1} existed in all the spectra.

As discussed above, the main absorption bands in the FTIR spectra of holocelluloses isolated from the bark, sapwood and heartwood of Fuji *C. japonica* are confirmed in the FTIR spectra of holocelluloses in the corresponding component of Iiate *C. japonica*. Thus, it may be concluded that the fallout cesium radioactivities did not affect the composition and/or structure of cellulose and

hemicelluloses of *C. japonica*.

FTIR spectra of dioxane lignins

Table 6 shows the FTIR absorption bands of dioxane lignins isolated from the bark, sapwood and heartwood, respectively, of Iitate and Fuji trees, as well as their assignments. Regarding the Iitate tree, the O-H stretching band in the broad region of 3000-3500 cm^{-1} is observed at 3413 cm^{-1} in the bark and moved gradually to 3420 cm^{-1} in sapwood and 3432 cm^{-1} in heartwood. A similar trend was found for the Fuji tree as well. Namely, the band at 3396 cm^{-1} in the bark shifted to 3426 cm^{-1} in the sapwood and then to 3436 cm^{-1} in the heartwood.

For the Iitate tree, the C-H stretching band at 2924 cm^{-1} in the bark lignin moved to 2937 cm^{-1} in both sapwood and heartwood lignins. Similarly, the band at 2923 cm^{-1} in the Fuji tree bark lignin shifted to 2937-2938 cm^{-1} in sapwood and heartwood lignins. Whereas the C-H stretching band at 2852 cm^{-1} was a peak in the bark lignin, it was a shoulder in both sapwood and heartwood lignins from both Iitate and Fuji trees.

The unconjugated C=O group band at 1714 cm^{-1} in the Iitate bark lignin remained at the same position in both lignins of its sapwood and heartwood. However, for the Fuji tree, the same band appeared at 1709-1710 cm^{-1} in the bark, but shifted to 1716 cm^{-1} in sapwood and to 1713 cm^{-1} in heartwood.

Of the symmetric aromatic skeletal vibration bands at 1602 cm^{-1}, 1513 cm^{-1} and 1427 cm^{-1}, the band at 1602 cm^{-1} in the bark lignin moved to 1598 cm^{-1} in both sapwood and heartwood lignins from the Iitate tree. For the Fuji tree, this band was found at 1607-1608 cm^{-1} in the bark lignin, but shifted to 1597 cm^{-1} in both sapwood and heartwood lignins. The asymmetric aromatic skeletal vibration band at 1513 cm^{-1} was found in the lignins of the bark, sapwood and heartwood of the Iitate tree. Similarly, this band was confirmed at 1512 cm^{-1} in the lignins of the bark, sapwood and heartwood of the Fuji tree. The aromatic skeletal vibration band at 1427 cm^{-1} was found in the lignins prepared from the bark, sapwood and heartwood of the Iitate tree. The same band was also found in the lignins from the outer bark, but not in the inner bark of the Fuji tree. However, the band appeared clearly at 1426 cm^{-1} in the spectra of lignins in both sapwood and heartwood of the Fuji tree.

Table 6
FTIR absorption bands of dioxane lignins isolated from barks, sapwoods and heartwoods of Iitate and Fuji *C. japonica*

Wood component / Functional groups	FTIR bands (cm^{-1})						
	Iitate bark	Fuji outer bark	Fuji inner bark	Iitate sapwood	Fuji sapwood	Iitate heartwood	Fuji heartwood
O-H stretching vibration	3413	3396	3396	3420	3426	3432	3436
CH stretching vibration	2924	2923	2923	2937	2938	2937	2937
C-H stretching of O-CH_3 group	2852	2852	2852	2852^{sh}	2852^{sh}	2852^{sh}	2852^{sh}
C=O unconjugated	1714	1710	1709	1714	1716	1714	1713
Aryl ring stretch symmetric	1602	1607	1608	1598	1597	1598	1597
Aryl ring stretch asymmetric	1513	1512	1512	1513	1512	1513	1512
C-H deformation asymmetric	1461	1461	1455	1458	1461	1461	1461
Aromatic skeletal vibration	1427	1417	----	1427	1426	1427	1426
C=O stretch in aryl ring	1269	1269	1268	1270	1270	1269	1269
C-C, C-O, C=O stretches	1223	1223	1223	1223	1223	1223	1223
Aromatic C-H in plane deformation	1153	1153	1153	1141	1141	1141	1141
Aromatic C-H in plane deformation	1033	1033	1032	1032	1032	1033	1032
C-H deformation out-of-plane, aryl ring	856	----	856	857	857	856	857
C-H deformation out of plane, aryl ring	816	819	818	817	8157	817	816

sh: shoulder

The asymmetric C-H deformation band at 1461 cm^{-1} was found in the lignins from bark and heartwood, but at 1458 cm^{-1} in the sapwood of the Iitate tree. This 1461 cm^{-1} band was also confirmed in the lignins from the outer bark, sapwood and heartwood of the Fuji tree. However, it appeared at 1455 cm^{-1} in the inner bark lignin of the same wood. The C=O stretching band in the aromatic rings at 1268-1270 cm^{-1}, 1223 cm^{-1}, the aromatic C-H deformation band at 1141-1153 cm^{-1}, as well as the aromatic C-H deformation band at 1032-1033 cm^{-1}, all appeared in the lignins of the bark, sapwood and heartwood of both Iitate and Fuji trees. The bands at 856-857 cm^{-1} and 816-819 cm^{-1} were all found in the lignins from the bark, sapwood and heartwood of both Iitate and Fuji trees. They are assigned either to the C-H out-of-plane or to the C-H stretch vibration.

The above assignment of FTIR bands was described in the literature.[26] Similarly to the results of the holocelluloses discussed above, based on the examination of the FTIR spectra of dioxane lignins isolated from the bark, sapwood and heartwood of Iitate and Fuji *C. japonica*, it may also be concluded that cesium radioactivities did not alter the composition and/or structure of lignin of *C. japonica*.

Impregnation of Fuji *C. japonica* with ^{137}Cs solution

Table 2 shows that the components of Iitate *C. japonica*, such as bark, sapwood, heartwood and holocellulose from extractive-free bark, were radiocontaminated with ^{134}Cs and ^{137}Cs. It is curious then how the corresponding wood component in the non-radiocontaminated Fuji *C. japonica* would respond when they were impregnated with a ^{137}Cs solution. The results of the experiment are given in Table 7.

The bound ^{137}Cs activities were highest in the outer bark, followed by heartwood, inner bark and sapwood. The results are similar to those of the Iitate tree, thus suggesting that radiocesium is readily bound to the tree components, such as bark, heartwood and sapwood, as well as to holocelluloses and lignins of these tree components.

Table 7
Impregnation of wood components from Fuji *C. japonica* with ^{137}Cs solution

Wood component	Initial ^{137}Cs ($Bq.mL^{-1}$)	Bound ^{137}Cs ($Bq.kg^{-1}$)	Bound ^{137}Cs (%)
Outer bark	246,436±861	189,670±663	77.0
Inner bark	196,681±842	108,834±466	55.3
Sapwood	113,786±592	51,105±266	44.9
Heartwood	126,878±539	82,140±349	64.7
Holocellulose of outer bark	417,687±2,108	29,403±663	31.0
Holocellulose of inner bark	852,166±3,546	308,054±1,282	33.6
Holocellulose of sapwood	195,854±859	97,118±426	49.6
Holocellulose of heartwood	175,895±783	94,961±424	54.0
Dioxane lignin of sapwood	482,682±6,053	17,225±216	3.6
Dioxane lignin of heartwood	476,295±5,779	15,330±186	3.2

The average ^{137}Cs activities in holocelluloses from outer and inner barks are approximately one half of those of the outer and inner barks. These results are similar to those for the Iitate tree. The lignins in both sapwood and heartwood of the non-radiocontaminated Fuji *C. japonica* contained very few bound ^{137}Cs activities, around 3%, thus suggesting that the radiocesiums ^{134}Cs and ^{137}Cs existing in sapwood and heartwood were readily removed by the dioxane used in the isolation of dioxane lignins. This is also the reason why dioxane lignins from the extractive-free Iitate *C. japonica* contained no ^{137}Cs activities, as shown in Table 3. Attempts were made to scan the components, such as holocelluloses from bark, sapwood, heartwood, and dioxane lignins from sapwood and heartwood of the radiocesium-impregnated Fuji *C. japonica* using the FTIR technique. However, due to the small amounts of the prepared ^{137}Cs-impregnated components of this wood, only ^{137}Cs-impregnated holocelluloses from the outer and inner barks could be used for this investigation. A comparison of the absorption bands in the FTIR spectra of the holocelluloses from original non-radiocontaminated outer bark and the ^{137}Cs-impregnated one is summarized in Table 8. The OH stretching vibration band at 3377 cm^{-1} in the original outer

bark holocellulose appeared at a similar position of 3376 cm^{-1} in the ^{137}Cs-contaminated one. However, the C-H stretching vibration band at 2895 cm^{-1} in the original outer bark holocellulose moved up to 2915 cm^{-1} in the ^{137}Cs-contaminated one. The C=O stretching band at 1727 cm^{-1} of acetyl and carbonyl groups in hemicelluloses remained at the same wavelength in both non-radiocontaminated and radiocontaminated outer bark holocelluloses. However, the O-H bond of adsorbed water moved down from 1643 cm^{-1} in the original uncontaminated outer bark holocellulose to 1630 cm^{-1} in the ^{137}Cs-contaminated one.

Table 8
Comparison of FTIR absorption bands of the original outer and inner bark holocelluloses with those from the corresponding ^{137}Cs-impregnated components of Fuji *C. japonica*

Chemical functional groups	Original	^{137}Cs-impregnated	Original	^{137}Cs-impregnated
O-H stretching vibration	3377	3376	3357	3369
C-H stretching vibration	2895	2915	2916	2897
C=O stretching of acetyl and carbonyl groups in hemicelluloses	1727	1728	1726	1726
O-H bond of adsorbed water	1643	1630	1644	1633
Asymmetric C-H deformation	1427	1427	1427	1426
Symmetric C-H deformation	1372	1372	1371	1372
Unknown	1318	1318	1318	1318
C-O-C symmetric stretching	1159	1159	1159	1159
C=O stretching of cellulose and hemicellulose	1063	1063	1061	1063
Unknown	898	898	898	898

Other absorption bands at 1427 cm^{-1} (asymmetric C-H deformation), 1372 cm^{-1} (symmetric C-H deformation), 1318 cm^{-1}, 1159 cm^{-1} (C-O-C symmetric stretching), 1063 cm^{-1} (C=O stretching of cellulose and hemicelluloses), and 898 cm^{-1} all appeared in both original and ^{137}Cs-impregnated outer bark holocelluloses. Similar results were obtained for the original and ^{137}Cs-impregnated inner bark holocelluloses, except that the C-H stretching vibration at 2916 cm^{-1} moved down to 2897 cm^{-1} (Figs. 7, 8).

From the above results, it can be concluded that artificial ^{137}Cs radiocontamination of non-radiocontaminated bark holocelluloses would not alter the composition and/or the structure of holocelluloses of *C. japonica.* Similar results are also expected for the holocelluloses, as well as the lignins from sapwood and heartwood.

Possible radiocontamination pathway of vascular plants

The bark of a vascular plant is directly exposed to wetting during rainfall and may exchange water with the atmosphere through absorption and desorption of water vapor. Llek and co-workers[27] suggested that the ability of the bark to absorb water vapor during non-rainfall season, *i.e.* hygroscopicity, leads to partial saturation of bark tissues during dry season that may alter the rate of saturation during rainfall, and that bark hygroscopicity may constitute an average of 30% of total bark water storage capacity.

Although bark does absorb water, most of the water absorbed would not be available to the plant, but returned to the air by evaporation. As such, only after the bark is moistened thoroughly to the phloem layer lying behind will water be available for the living cells in the trunk to uptake. Due to its relatively low boiling point (671 °C), ^{137}Cs is volatilized easily when it is released suddenly at high temperature, as in the FDNPP accident. ^{137}Cs and other released radioactive materials in the atmosphere are transported as gaseous substances or particulate matter, and finally land by dry deposition on earth surface (vertical transportation due to gravity drop and turbulence) and wet deposition due to precipitation. On the day of March 11, 2011, it was raining in Fukushima from 3 PM. The rain fall was 0.5 mm. The released ^{137}Cs might then be carried away with the rainfall and thus may retained in the canopies of the forest.

A part of the radiocontaminated rain water would pass through the canopies by dripping from leaves and branches (throughfall) or flowing down the stems of trees (stemflow), and eventually reach the forest floor.[28] The other part of the radiocontaminated rain water would eventually

evaporate from the canopies and be lost to the atmosphere. The radiocontaminated stemflow takes place on the bark of the tree. As mentioned above, the radiocontaminated water absorbed by the bark will eventually be returned to the air by evaporation. Suppose that if only the water is evaporated, this would result in the retention of radiocesium on/in the bark, which, in turn, would not be available to the plant, but to the bark. This may be one reason of the high radiocesium activities in the bark of the Iitate *C. japonica* mentioned above. These reasonings are contrary to the conclusions that radiocesium moves radially from the bark to the inner parts of the tree, such as sapwood and heartwood.[6,29]

Xylem and phloem are complex vascular tissues of vascular plants and together they form vascular bundles. Xylem exists at the center of the vascular bundle and can be also found in roots, stem and leaves. It transports water and minerals from roots to aerial parts of the plant.[30] The movement of the xylem is unidirectional, *i.e.* moving up the stem of the plant. On the other hand, phloem occurs on the outer side of the vascular bundle just behind the bark. Its function is to transport food, nutrients, such as sugars, *e.g*. sucrose, and amino acids from leaves to growing parts, such as roots and shoots, as well as to storage regions, like seeds and fruits. This movement of substances is the translocation within the plant. The movement of the phloem is bidirectional, *i.e.* moving up or down the stem of the plant from source to sink.

Once the upward water inside the xylem reaches the leaf surface, it evaporates through the stomata (singular: stoma) on the leaf surface. This loss of water is the transpiration of plant. The cohesion–tension theory assumes that the water ascent in trees is exclusively due to the transpirational pull from continuous water columns in the xylem conduit running from roots to leaves.[31] Each stoma is surrounded by two guard cells, which open and close in response to light intensity and quality, leaf water conditions and carbon dioxide concentration. Stomata must open to let the air containing carbon dioxide and oxygen diffuse into the leaf for photosynthesis and respiration. Simultaneously, water inside the plant is lost to the outside environment, resulting in an increase of transpiration. Thus, plants must maintain a balance between water loss and efficient photosynthesis.

The products of photosynthesis are simple sugars, such as sucrose, which are transported through the plant via the phloem. This is the other translocation process of the plant. The points of sugar delivery are the growing parts, such as roots, young shoots, which, in turn, are nearest to the location where the photosynthesis takes place.

From the facts considered above, it is rational to propose that: 1) the radiocesium vapor and/or particulate matter from the FDNPP accident were retained in the forest canopies and deposited on the leaves surfaces; 2) these radioactive materials enter the plant through the open stomata during daytime, where the plant transpiration and photosynthesis take place; 3) once entering the stomata, the radioactive materials go to the phloem stream, contaminate the sapwood, and then are carried away together with food, nutrients (sugars, amino acids *etc*.) to roots, shoots, flowers, seeds, fruits *etc*.; 4) simultaneously, radiocesium deposited in the soil (dry deposition) as well as through-fall radiocesium, together with water and minerals in the soil enter the roots through the diffusion process called osmosis.

Osmosis in roots leads water moving into root hair cells. Once inside the root hair cells, water, food, nutrients and radiocesium enter the root cortex, then passes through the endodermis and from there they can access the xylem tubes in the center of the plant and allow for water transportation in plant. This pathway of radiocesium movement would contaminate the heartwood as well.

Since stomata are the only ports for both the water from plant transpiration going out and carbon dioxide, oxygen in the air entering the plant, it is expected that the radiocesium activities deposited on the leaves could only partly reach the phloem stream through the stomata route. As a result, the radioactivities in the contaminated sapwood would be lower than in the bark.

As mentioned above, the phloem stream distributes food, nutrients (sugars, amino acids *etc*.) to roots. Therefore, it is expected that this is where the going down phloem stream meets the food, minerals, nutrients and soluble radiocesium entering the roots through the osmosis process. The mixture would then meet the xylem stream after passing the endodermis. Consequently, the radioactivities of the xylem stream would be higher than that of the phloem stream. Therefore, the radioactivities in heartwood would be higher compared to those in sapwood.

CONCLUSION

Analysis of the bark, sapwood, heartwood from a radiocontaminated *C. japonica* confirmed the presence of ^{134}Cs and ^{137}Cs in these wood

components. The total of ^{134}Cs and ^{137}Cs was the highest in the bark and the lowest in the sapwood, while that of heartwood was in between. ^{134}Cs and ^{137}Cs were found in the holocelluloses isolated from the bark, but not in those from the sapwood and heartwood. The results were probably due to the high ^{134}Cs and ^{137}Cs activities in the bark. Similarly, dioxane lignins from sapwood and heartwood were also free of ^{134}Cs and ^{137}Cs activities. Because minerals, such as calcium, magnesium, potassium, silica and iron, were all present in the bark, sapwood and heartwood, it was thought that the holocellulose and dioxane lignins in these wood components also contained these minerals. Due to the ion exchange capabilities of radiocesiums with these minerals, the holocelluloses and lignins would be contaminated by these radiocesiums. However, an examination of FTIR spectra of the holocelluloses and lignins from radiocontaminated and non-radiocontaminated *C. japonica* suggested that the chemical components of both holocellulose and lignin were not affected by the radiocontamination. Similar results were obtained for a *C. japonica* artificially radiocontaminated with ^{137}Cs.

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