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ORIGINAL ARTICLE

Decontamination of Cs from Japanese cedar (*Cryptomeria japonica*) via kraft cooking

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Abstract To investigate the possibility of decontaminating ^{137}Cs-contaminated *Cryptomeria japonica* wood, kraft pulping was conducted and the Cs behavior in the reaction process was examined. ^{133}Cs-treated or ^{137}Cs-contaminated bark, sapwood, and heartwood chips of *Cryptomeria japonica* were digested using an aqueous solution of NaOH and Na_2S. The pulp was washed with ultrapure water and filtered, after which the filtrate (black liquor) was collected. The black liquor was acidified to separate the supernatant and precipitation. The Cs (^{133}Cs and ^{137}Cs) concentrations in the chip and reaction products were measured. As for wood samples, the majority of Cs was present in black liquor, while only a minor amount of Cs was retained in the pulp (<1%). In the case of bark, although the majority of Cs was present in the black liquor, the proportion of Cs in the pulp was much higher than that in the wood pulp. In addition, the Cs in the precipitation of the bark was higher than that in the wood, possibly because the Cs in the bark was combined with some components, which is insoluble in alkaline solution. Our results suggest that ^{137}Cs-contaminated *Cryptomeria japonica* wood can be used in the pulp and paper industries.

Keywords Radiocesium · Stable cesium · Kraft pulping · Contaminated wood · Decontamination

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Introduction

The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident in March 2011 resulted in a release of large amounts of radionuclides into the Fukushima Prefecture [1]. Because forests cover approximately 70% of the entire prefectural area and are highly efficient at intercepting gaseous and particulate contaminants [2, 3], the radioactive contamination of forests and forest products is a serious problem in the Fukushima Prefecture.

Japanese cedar (*Cryptomeria japonica* D. Don) is one of the most abundant forest resources in Japan, comprising 43% of the country's artificial forests [4], and it is one of the most valuable plantations for the timber industry [5]. At present, in Japan, because there is no regulation in place for the radiation levels of building materials using wood from conifer forests [6], the risk of using ^{137}Cs-contaminated wood is difficult to evaluate and the use of *C. japonica* as timber is restricted without sufficient decontamination. To maintain the public function of the *C. japonica* forest and to revive the forest industry in the Fukushima Prefecture, it is important to

explore uses of *C. japonica* other than as timber. After being deposited onto forests, ^{137}Cs can be absorbed and translocated to wood through three main pathways: surface absorption through leaves [7, 8] and bark [9, 10], and root uptake from the soil. In the case of *C. japonica*, we confirmed that ^{133}Cs ions applied to bark were absorbed into the heartwood through the bark [10]. Therefore, a part of the ^{137}Cs in the wood of *C. japonica* is likely derived from bark absorption in an ionic form. In tree stem wood, ^{137}Cs may form ionic bonds with carboxylic groups, in cell walls or in the cytoplasm of living cells, and is very mobile across tree rings [11, 12], which indicates that ^{137}Cs could be removed from contaminated wood via chemical treatments. Therefore, one promising application of ^{137}Cs-contaminated *C. japonica* is to chemically convert ^{137}Cs-contaminated wood into pulp, which is then used to produce paper and related products. Kraft pulping is the most frequently used chemical method to transform wood into pulp [13]. A conventional kraft pulping process usually involves digesting the wood chips at elevated temperature and pressure with an aqueous solution of sodium hydroxide and sodium sulfide. Most of the lignin is dissolved and separated from the pulp via a washing process and filtration. The pulp is further processed until it is suitable for the manufacture of paper and other related products [14]. At present, because the behavior of ^{137}Cs during the kraft pulping of *C. japonica* wood is not well understood, the use of ^{137}Cs-contaminated *C. japonica* wood in pulp and paper industries is still in suspense.

Cesium is a metal element of group 1. ^{133}Cs is the only natural stable isotope, and ^{137}Cs is radioactive and has a half-life of ca. 30 years. It is extremely difficult to find the difficulties in chemical behavior between the two isotopes. In this study, laboratory kraft cooking of ^{133}Cs-treated and ^{137}Cs-contaminated wood is performed. The purposes of the study are to elucidate the behavior of Cs during the kraft pulping process and to evaluate whether ^{137}Cs-contaminated *C. japonica* wood can be used in pulp and paper industries. In addition, even though bark is usually removed from wood and is not used to produce pulp, the kraft cooking of bark is also conducted to study whether the behavior of Cs differs between the bark and wood during the kraft cooking process.

Materials and methods

Sampling and sample preparation

The ^{133}Cs-treated wood was prepared by applying a ^{133}CsCl solution onto the bark of *C. japonica* at Nagoya University (Fig. 1, NU). On May 15, 2015, a 40-ml aliquot of 0.01 M ^{133}CsCl solution was spread uniformly on a paper towel. The

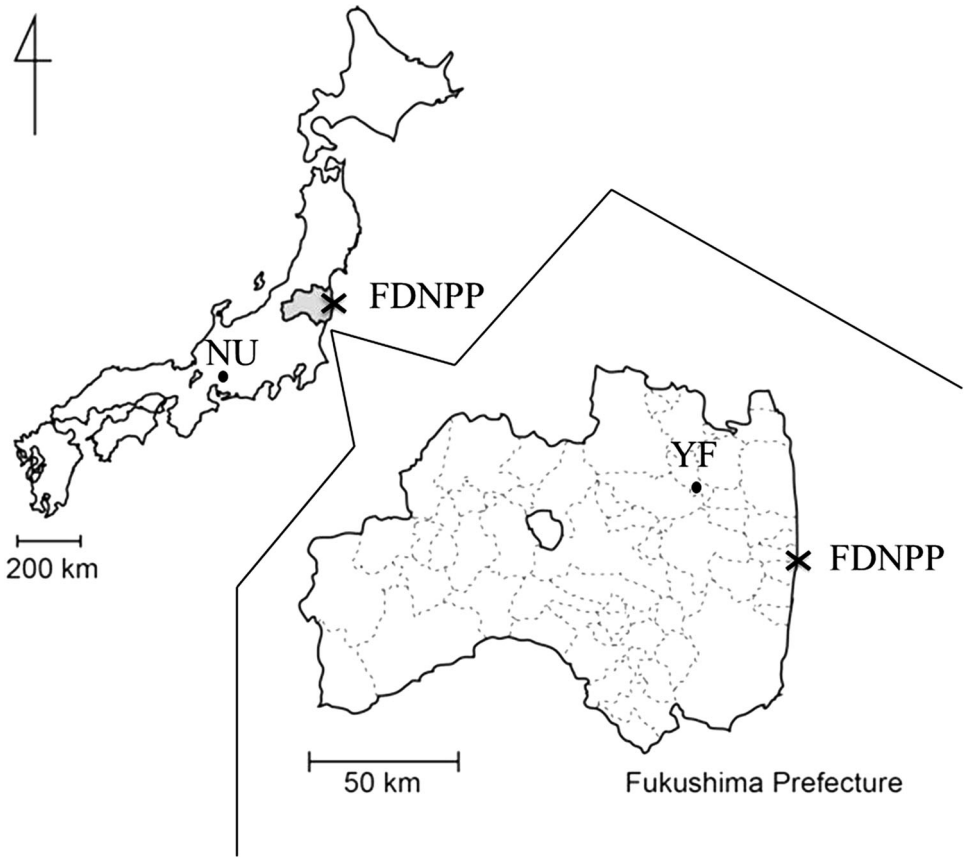

Fig. 1 A map of the sampling site; *YF* indicates the Yamakiya forest and *NU* indicates Nagoya University. *FDNPP* indicates the Fukushima Daiichi Nuclear Power Plant

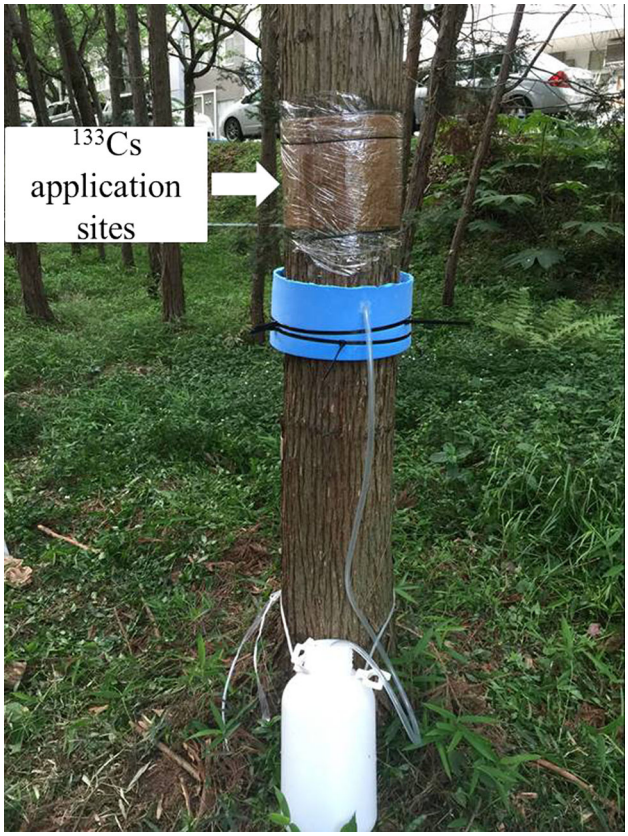

Fig. 2 Application of ^{133}Cs to Japanese cedar bark. A paper towel with 40 ml of a 0.01 M ^{133}CsCl solution was wrapped around the bark at 1.2 m. A urethane sheet and hose were used to direct the stemflow to the guttering

towel was then attached to the bark at a height of 1.2 m above the ground. To prevent the towel from drying, it was covered with plastic wrap and fixed using plastic wires. Further, to prevent soil contamination from ^{133}CsCl caused by stemflow, a 3-cm-thick urethane sheet was wrapped around the trunk below the paper towel, which was then fixed using plastic wires that also functioned as stemflow gutters. A hose was connected to the urethane sheet to direct the stemflow to a bucket. The juncture between the hose, bark, and urethane was filled with silicon resin to prevent leakage (Fig. 2). On June 4, 2015, the treated *C. japonica* was cut down, and 10-cm-thick disks were collected at heights of 1.2 m. As a control, untreated *C. japonica* was also cut and disks were collected at a height of 1.2 m. The surface soil (0–5 cm) was randomly collected at three points around each tree and assessed for whether the soil was contaminated with ^{133}Cs.

The ^{137}Cs-contaminated *C. japonica* disk was sampled from a *C. japonica* forest, located in Setohachiyama, Yamakiya district, Fukushima Prefecture, approximately 36 km northwest of the FDNPP (Fig. 1, YF). The stand age was 60 years, the air dose rates (μSv h^{-1}) was 1.5 μSv/h on September 24, 2015, which were measured with a scintillation survey meter (TCS-161, Aloka), about 1 m above ground level and were presented as an average from 10 different points. On that day, 10-cm-thick disks were collected at a height of 0.4 m for this study. The collected disks were divided into bark, sapwood, and heartwood. Wood and bark chips were prepared from each portion, and the average size of the wood and bark chips was 11 mm × 10 mm × 5 mm and 10 mm length × 10 mm width, respectively.

Kraft cooking

Wood and bark chips (1.5 g) were digested in a 10-ml high-pressure stainless steel tube, using "white liquor," which is an aqueous solution of 7.5 ml NaOH and $Na_2S{\cdot}9H_2O$. The active alkalinity and sulfidity of the alkaline solution was 20 and 28%, respectively. Kraft cooking was performed in an oven, the cooking temperature was gradually increased from 20 to 170 °C within 1.5 h, and this temperature was maintained for 2 h. After cooling, the kraft pulp was washed with 15 ml of Mill-Q water (with resistivity more than 18 MΩ) and filtered with 5-μm filter paper. The filtrate was collected and is referred as the 1st black liquor, which consisted primarily of water, dissolved wood components, and the spent digesting agents. The kraft pulp was further washed with 15 ml of ultrapure water and filtered an additional two times to collect the 2nd and 3rd black liquors. Then, the pulp was continuously washed until the filtrate became nearly colorless; the obtained pulp is referred to as the washed pulp. A portion of the black liquor (3 ml) was taken and acidified with 2 ml of 10% H_2SO_4 to adjust the pH to 2–3, resulting in the formation of precipitation, the main component of which was lignin. The precipitation and supernatant were then separated with a 0.45-μm membrane filter. The wood, bark chips not used for kraft cooking, washed pulp, and the precipitation were oven-dried at 80 °C for more than 48 h to a constant weight. A flow chart of the kraft cooking process is illustrated in Fig. 3.

^{133}Cs and ^{137}Cs determination

^{133}Cs determination: The chips, washed pulp, and precipitation were digested with HNO_3 using a graphite block acid digestion system (EcoPre; ODLAB). The concentrations of ^{133}Cs in the abovementioned components, as well as in the supernatant, were determined using an inductively coupled plasma mass spectrometer (ICP-MS) (iCAPQc, Thermo Scientific) with indium (In) as the internal standard. The ion-exchangeable ^{133}Cs in the soil samples (2 g) were extracted using 1 M NH_4AC (40 ml, pH 7) and its concentration was determined with ICP-MS.

^{137}Cs determination: The chip, pulp powder, black liquor, and supernatant were placed in plastic containers (Ø 56 mm × H 35 mm; PP-U9, Sekiya Rika), and the ^{137}Cs activities were measured using a high-purity germanium

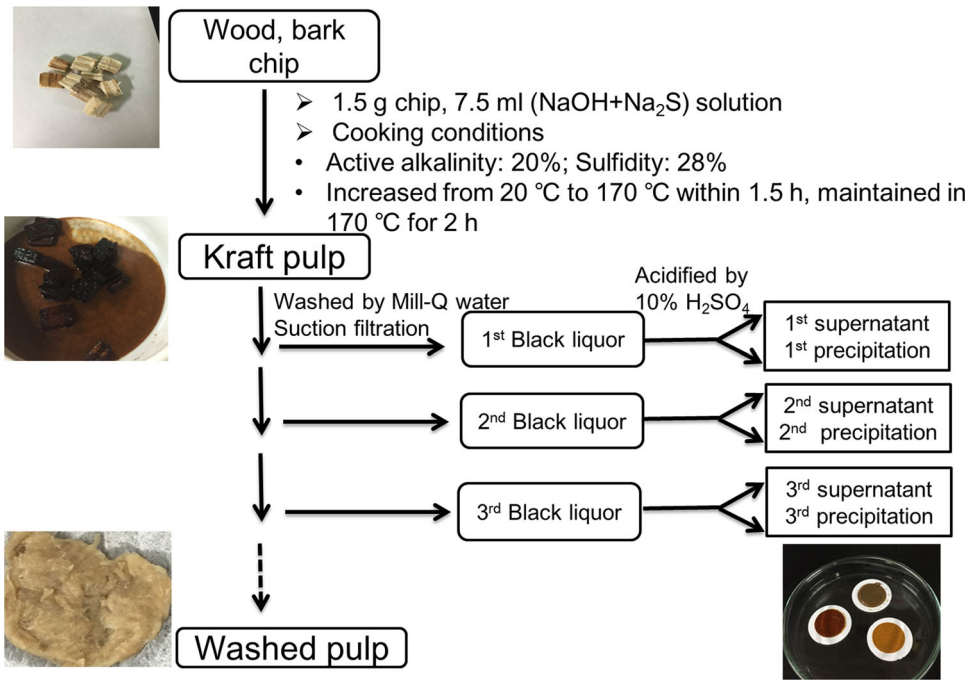

Fig. 3 Flow-chart of the kraft pulping process

(HPGe) detector (GC3520, Canberra; GEM-30195-P, Seiko EG&G Ortec). Standard ^{137}CsCl solutions (Japan Radioisotope Association) diluted with ^{133}CsCl were used as standard sources for the HPGe detector. Measurements were continued for 80,000 s or until a counting error of less than 5% of the ^{137}Cs peak was achieved. The radioactive concentration of ^{137}Cs (Bq/kg dry weight) was calculated based on the gamma ray spectrum using the Gamma Explorer software (Canberra Japan KK, Tokyo). The activities were corrected for decay back to the sampling date. The ^{137}Cs activity for the precipitation was calculated from the difference between the ^{137}Cs activities in the corresponding black liquor and supernatant.

Statistical analysis

The Shapiro–Wilk test was employed to check the normality of the data set, and the Mann–Whitney *U* test (significance level of 5%) was used to compare the ^{133}Cs concentration between the treated samples and the control samples.

Results

^{133}Cs and ^{137}Cs concentrations in the bark and wood of *C. japonica*

The ^{133}Cs and ^{137}Cs concentrations in the bark, sapwood, and heartwood are shown in Fig. 4a for ^{133}Cs-treated trees and in Fig. 4b for the trees from Fukushima. The ^{133}Cs concentrations in the sapwood and heartwood of the ^{133}Cs-treated trees were considerably higher than those of the control trees, even though there were no significant differences between them. Because no significant differences were observed between the average ^{133}Cs concentrations in the soils under the treated trees (0.097 ± 0.021 μg g^{-1}) and those under the control trees (0.075 ± 0.029 μg g^{-1}), this indicates that the influence of root-uptake caused by the leakage of the ^{133}CsCl solution was negligible, and that the ^{133}Cs entered the wood through the bark and was translocated into the heartwood. For the ^{137}Cs-contaminated trees from Fukushima, the ^{137}Cs concentration was highest in the bark, and the concentration in the heartwood was higher than that in the sapwood (Fig. 4b).

Pulp yields of the bark and wood

The pulp yields of the bark, sapwood, and heartwood of ^{133}Cs-treated and ^{137}Cs-contamianed *C. japonica* are given in Table 1. The pulp yields of the sapwood and heartwood were nearly the same and the pulp yield of the bark was the lowest of the three parts of the wood for both the ^{133}Cs-treated and ^{137}Cs-contamianed *C. japonica.* The lower pulp yields in the bark compared to the wood may occur because the amounts of polyphenols with high molecular weight, extractives, and ash in bark are higher than those in wood, which will lead to a lower pulp-yield in bark than in wood.

Proportion of Cs in each component after kraft cooking

To investigate the behavior of Cs during the kraft cooking process, we calculated the amount of ^{133}Cs (μg) and the

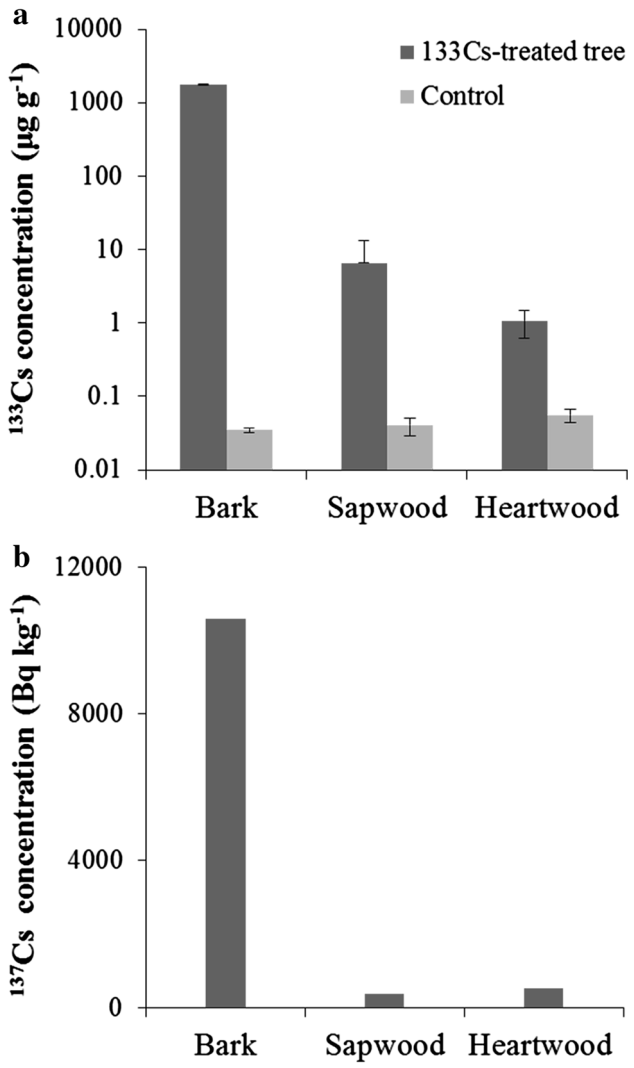

Fig. 4 **a** ^{133}Cs and **b** ^{137}Cs concentrations in the bark, sapwood, and heartwood. The *Y*-axis of **a** is in logarithmic units. The *error bars* indicate the standard deviations of three replicates

Table 1 Pulp yields (%) of the ^{133}Cs-treated and ^{137}Cs contaminated bark and wood ($n = 3$)

	^{133}Cs-treated	^{137}Cs-contaminated
Bark	34 ± 2.1^{a}	29 ± 0.58^{a}
Sapwood	38 ± 3.6^{ab}	36 ± 4.2^{b}
Heartwood	41 ± 0.58^{b}	34 ± 2.1^{ab}

The values are the mean ± standard deviation of three replicates

Different letters mean significant difference at $p < 0.05$

activity of ^{137}Cs (Bq) in the chips and each component after kraft cooking; the results are shown in Tables 2 and 3, respectively. Then, we calculated the proportion of Cs in each component based on these results. The proportion was calculated to be the percent ratio of ^{133}Cs or ^{137}Cs in each component to the total amount of ^{133}Cs or ^{137}Cs in all the components after kraft cooking. The results are shown in Fig. 5. For sapwood, approximately 99% of the ^{133}Cs was present in the supernatant, while that in the pulp (0.59%) and precipitation (0.1%) was less than 1%. The ^{137}Cs in the sapwood was detected in the 1st and 2nd black liquors, which include the supernatant and precipitation, and the ^{137}Cs proportion of the 1st black liquor (68%) was greater than that of the 2nd black liquor (32%). The proportion of Cs in the heartwood was similar to that in the sapwood in which the majority of the ^{133}Cs (approximately 99%) was retained in the supernatant while that in the pulp (0.84%) and precipitation (0.31%) were less than 1%, and the distribution of ^{137}Cs was greater in the 1st black liquor (56%) than in the 2nd (27%) and 3rd (17%) black liquors. In the case of bark, even though the majority of the ^{133}Cs (95%) and ^{137}Cs (86%) was present in the supernatant, the proportions of the ^{133}Cs (4.2%) and ^{137}Cs (5.9%) in the pulp were higher than those in the sapwood and heartwood. In addition, the proportions of ^{133}Cs and ^{137}Cs in the precipitation of the bark were also higher than those in the sapwood and heartwood.

Discussion

Use of ^{137}Cs contaminated *C. japonica* wood in pulp mills

We explored the possibility of using radionuclide-contaminated woods as resources for pulp in this study. To use the contaminated wood, at least the following two requirements should be fulfilled: (1) the ^{137}Cs activity in the pulp should be under the detection limit or below the safety level and (2) the ^{137}Cs in the black liquor should be properly processed.

In this study, because ^{137}Cs was not detected (Fig. 5d, f) and only a minor amount of ^{133}Cs was present (Fig. 5c, e) in the wood pulp, this suggests that the ^{137}Cs-contaminated *C. japonica* wood could potentially be used to produce paper and related products via the kraft pulping process. The existence of minor amounts of ^{133}Cs in the wood pulp may be due to the insufficient washing process because only 45 ml of ultrapure water in total was used. In industrial pulping processes, the pulp obtained after kraft cooking will undergo several further processes, such as bleaching and washing, before it can be used to produce paper. It is expected that the amount of Cs retained in the wood-pulp will be further reduced after such processes.

The majority of the ^{137}Cs was present in the black liquor (Fig. 5d, f), which consists primarily of dissolved wood components, spent digesting agents, and other impurities. In a pulp mill, to improve the economic benefits and to minimize waste, the digesting agents are recycled. Because

Table 2 The amount of ^{133}Cs (μg) in each component after kraft cooking 1.5 g of chips

	Chip	Pulp	1st black liquor		2nd black liquor		3rd black liquor	
			1st supernatant	1st precipitation	2nd supernatant	2nd precipitation	3rd supernatant	3rd precipitation
Bark	2690	128 ± 95	216 ± 1450	11 ± 8.8	522 ± 390	1.4 ± 1.3	227 ± 168	0.94 ± 0.97
Sapwood	6.2	0.033 ± 0.025	2.7 ± 0.38	0.0026 ± 0.0006	1.7 ± 0.1	0.0021 ± 0.0005	1.1 ± 0.4	0.00060 ± 0.00043
Heartwood	1.6	0.014 ± 0.003	0.96 ± 0.12	0.0053 ± 0.0009	0.39 ± 0.07	ND	0.33 ± 0.09	ND

The values are the mean ± standard deviation of three replicates

The bark and wood chips were thoroughly homogenized and only one sample was analyzed

ND not detected

Table 3 The activity of ^{137}Cs (Bq) in each component after kraft cooking 1.5 g of chips

	Chip	Pulp	1st black liquor		2nd black liquor		3rd black liquor	
			1st supernatant	1st precipitation	2nd supernatant	2nd precipitation	3rd supernatant	3rd precipitation
Bark	15.9	1.2 ± 0.1	13 ± 1.8	1.1 ± 1.2	2.5 ± 1.1	0.30 ± 0.09	1.3 ± 0.5	0.33 ± 0.17
Sapwood	0.49	ND	0.29 ± 0.04		0.14 ± 0.01		ND	
Heartwood	0.78	ND	0.42 ± 0.01		0.20 ± 0.04		0.12 ± 0.02	

The values are the mean ± standard deviation of three replicates

The bark and wood chips were thoroughly homogenized and only one sample was analyzed

The ^{137}Cs in the supernatant and precipitation of the sapwood and heartwood was not detected

ND not detected

most of the ^{137}Cs was concentrated and retained in the black liquor (Fig. 5d, f) and the recovery process for the digesting agents would considerably enhance the average concentration of radionuclides compared to those in the wood [15, 16], developing methods to remove ^{137}Cs from black liquor is necessary to reduce not only the exposure to workers in pulp plants but also the discharge of ^{137}Cs into aquatic ecosystems. The application of a Cs-selective absorbent used to remove ^{137}Cs from radioactive water appears to be a promising choice [17–20].

Our results show that only a minor amount of Cs was present in the precipitation (Fig. 5c, e). The main component of the precipitation is lignin, and this means that the lignin could be further converted to added-value products, such as biosorbents of trace metals from wastewater [21, 22], precursors for carbon fibers [23], or amphiphilic derivatives with high surface activity [24], as well as several other applications [25, 26].

Implications for ^{137}Cs speciation in bark

Understanding the chemical speciation of ^{137}Cs is essential to evaluate its formation process and behaviors in the environment [27]. After its release into the atmosphere, ^{137}Cs was transported to the northwestern region of FDNPP [28] in the form of aerosols [29], including small particles such as Cs balls [30], or associated with sulfate salts and organic matter [27, 28]. Radionuclides can be trapped by tree bark directly via atmospheric deposition, stemflow, and resuspended soil particles resulting from wind erosion [31]. The chemical speciation of ^{137}Cs in bark plays a significant role in its behavior, such as its penetration into the wood through the bark [10] and its degradation.

In this study, we found that the proportions of ^{133}Cs and ^{137}Cs in the bark pulp were higher than those in the wood pulp (Fig. 5), suggesting that part of the ^{133}Cs and ^{137}Cs in the bark were not dissolved in the kraft pulping process. The insolubility of ^{133}Cs and ^{137}Cs may have two causes: (1) the ^{133}Cs and ^{137}Cs absorbed by the bark combined with unknown components have low solubility or cannot dissolve in the kraft pulping process and/or (2) the ^{137}Cs was deposited in an insoluble form such as a Cs-ball [30]. In addition, we found that the proportions of ^{133}Cs and ^{137}Cs in the precipitation of the bark (Fig. 5) were higher than those in the wood (Fig. 5); this can be explained in two ways: (1) ^{133}Cs and ^{137}Cs dissolved during the kraft pulping process but co-precipitated together with specific components in the bark via acidification and/or (2) the pore sizes of the filter paper (5 μm) used to separate the pulp and the black liquor and the membrane filter (0.45 μm) used to separate the supernatant and the precipitation were different. Therefore, it is likely that Cs-bearing particles

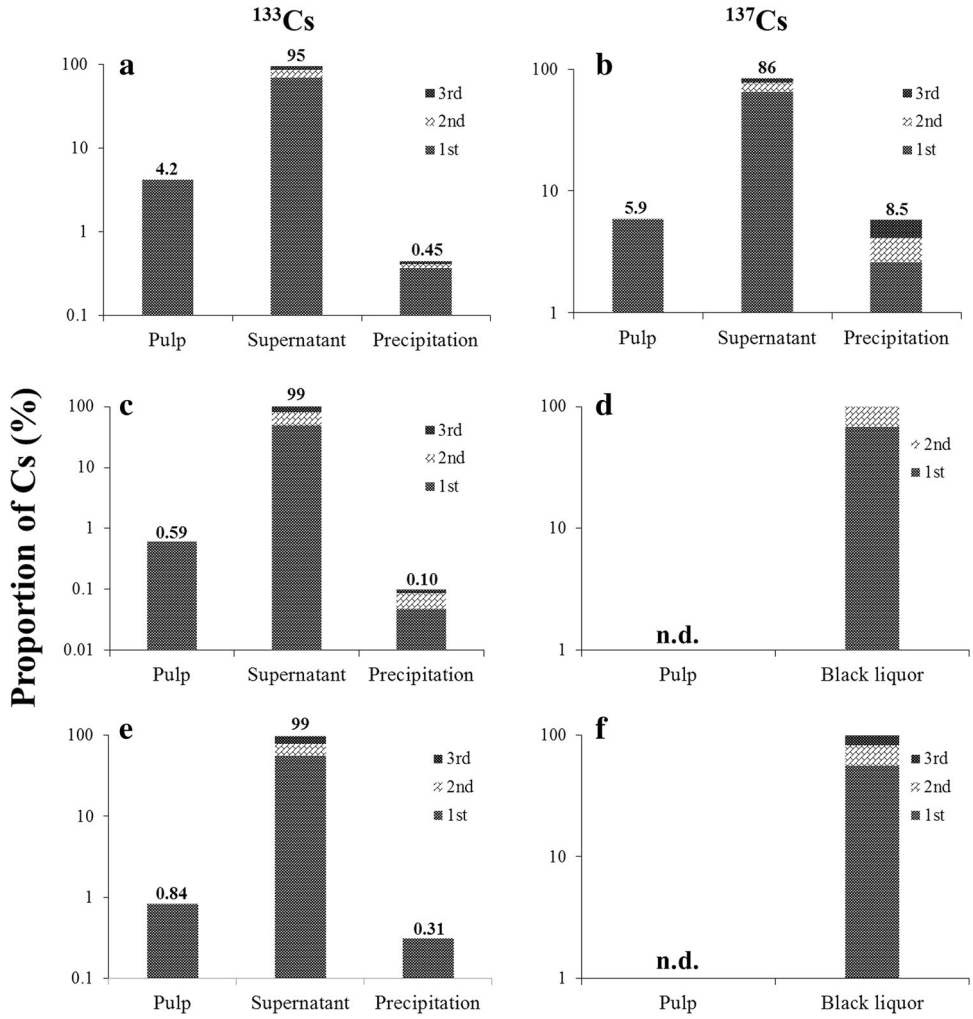

Fig. 5 Proportion of ^{133}Cs and ^{137}Cs in each component after kraft cooking: **a**, **b** bark; **c**, **d** sapwood; and **e**, **f** heartwood. The proportion was calculated as the percent ratio of the ^{133}Cs or ^{137}Cs in each component to the total amount of ^{133}Cs or ^{137}Cs in all the components (chips were not included). The ^{137}Cs radioactivity in the sapwood and heartwood was expressed in the form of black liquor because the ^{137}Cs radioactivity was not detected in the supernatant and precipitation. *n.d.* indicates not detected. The *Y*-axis of **a** is in logarithmic units

with diameters smaller than 5 μm but larger than 0.45 μm passed through the filter paper but were retained on the membrane filter. One possible Cs-bearing particle is a ^{137}Cs-ball, the diameter of which has been reported to be approximately 2 μm [30]. Previous studies have also reported that ^{137}Cs absorbed on bark likely exists in a stable chemical form and is not easily washed off the bark by weak acid solutions [32]. Further investigations are required to fully understand the speciation of ^{137}Cs in bark.

Conclusions

In this study, laboratory kraft cooking of ^{133}Cs-treated and ^{137}Cs-contaminated *C. japonica* wood and bark was conducted to investigate the behavior of Cs during the kraft cooking process and to evaluate whether the ^{137}Cs-contaminated *C. japonica* could be used in pulp and paper industries. Most of the Cs was transferred into the black liquor, while only a minor amount of the Cs was retained in the pulp. A minor amount of the Cs was also contained in the precipitation yielded by the acidification of the black liquor. From these results, it is suggested that the pulp from ^{137}Cs-contaminated woods be used if this minor amount of Cs retained in the pulp can be removed. The digested agents in black liquor can be recovered if the ^{137}Cs can be removed by a ^{137}Cs-special absorbent. In addition, the lignin could be used for further applications. The different behavior of Cs in the kraft cooking of bark and wood suggests that ^{137}Cs is bonded with unknown components in the bark that are difficult to dissolve in an alkaline solution. Further study at the pulp mill-scale would be interesting and is required for the forest industry in the Fukushima Prefecture to recover.

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Compliance with ethical standards

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

Human and animal rights statement This article does not contain any studies with human participants or animals performed by any of the authors.

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