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**論文 (Original article)**

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**<sup>137</sup>Cs concentration observational errors in bark and wood caused by partial sampling from tree stems contaminated by the Fukushima nuclear accident**Shinta OHASHI<sup>1,\*</sup>, Katsushi KURODA<sup>1</sup>, Takeshi FUJIWARA<sup>2</sup> and Tsutomu TAKANO<sup>3</sup>**Abstract**

Long-term monitoring of radiocesium (<sup>137</sup>Cs) contaminations in the bark and wood caused by the Fukushima Dai-ichi Nuclear Power Plant accident in 2011 requires partial sampling from a standing tree without felling it. Because <sup>137</sup>Cs distributions within the bark and wood are assumed not to be uniform, it is necessary to understand the observational errors in <sup>137</sup>Cs concentration caused by partial sampling and to check the validity of this method. The objectives of this study are to examine 1) the circumferential distributions of <sup>137</sup>Cs concentration in the bark and wood and 2) the observational errors in <sup>137</sup>Cs concentration determined via partial sampling compared with bulk (felling) sampling. The circumferential distributions were investigated by dividing the tree stems collected in 2015 into eight directional segments (four segments for small trees). The relative standard deviations of the <sup>137</sup>Cs concentration among the directions (the mean of all trees) were 34% and 13% for the bark and wood, respectively. The patterns of the circumferential distributions were not biased toward a specific direction and were not species-dependent. Partial sampling was achieved by collecting bark pieces (3 cm × 3 cm) from four directions and wood cores (12 mm in diameter) from 1–2 directions during the period of 2016–2020. The observational errors caused by the partial sampling were estimated to be approximately 38% and 8%–18% for the bark and wood, respectively, and were considered random (unsystematic). These results indicate that, for example, to estimate the mean value of the <sup>137</sup>Cs concentration in the bark in a forest stand with the same accuracy as the bulk sampling with n = 3, n for the partial sampling should be increased to 6–8.

**Key words :** Fukushima Dai-ichi Nuclear Power Plant accident, radiocesium, circumferential distribution, increment core

**1. Introduction**

Radiocesium (<sup>137</sup>Cs) derived from the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident has been transferred into the stem wood of trees from foliage and bark surfaces (Masumori et al. 2015, Nishikiori et al. 2015, Wang et al. 2016, 2018) or from soil via roots (Komatsu et al. 2017). Accurate and prolonged monitoring of <sup>137</sup>Cs concentrations in the bark and wood of tree stems is critically important to make appropriate plans for forest management and wood use in radioactively contaminated areas. To obtain data concerning the <sup>137</sup>Cs concentration that is representative of an individual tree, bark samples should be collected from the entire circumference of the stem, and wood samples should be collected as disks by felling the tree (Photos 1a and 1b). However, because such bulk sampling decreases the number of trees and disturbs the environment in a forest stand, it is not suitable for decades-long monitoring programs. Partial sampling of bark and wood from a standing tree using a chisel

and an increment borer, respectively (Photos 1c and 1d), is required for sustainable monitoring. However, it is necessary to determine any observational errors caused by such partial and small-amount sampling. Accordingly, it is also essential to understand the variation in the vertical and circumferential distributions of the <sup>137</sup>Cs concentration in tree stems. Because the vertical distribution has been reported in several studies (Ohashi et al. 2014, 2020, Masumori et al. 2015, Ogawa et al. 2016, Yoschenko et al. 2017), we focus here on the circumferential distribution.

A few studies have investigated the circumferential distribution in stem wood (Mahara et al. 2014, Hirano et al. 2016). The data from these studies indicate that the relative standard deviation (RSD) of the <sup>137</sup>Cs concentration among four or eight evenly divided directions of a tree stem was approximately 10%–50% in Japanese cedar (*Cryptomeria japonica*) and konara oak (*Quercus serrata*) trees collected in 2012 (Mahara et al. 2014) and konara oak trees collected

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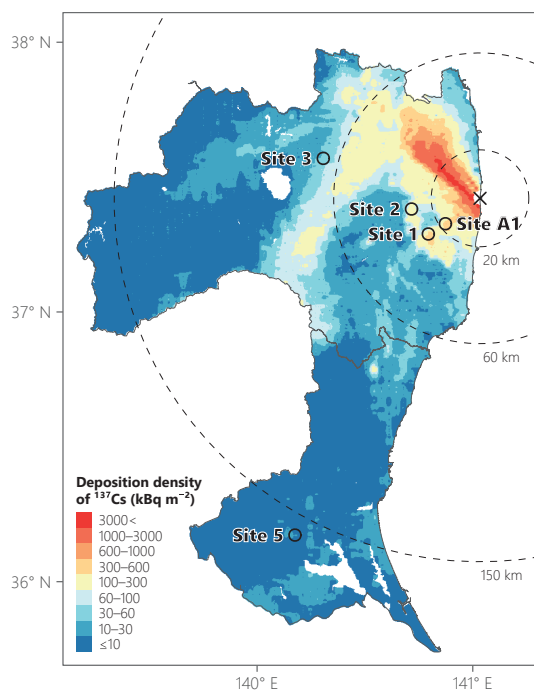
**Photo 1. Sampling of (a) bark from the entire circumference of a tree stem, (b) a wood disk, (c) a bark piece, and (d) a wood core.**

in 2014 (Hirano et al. 2016). On the other hand, the circumferential distribution in bark has not been reported for trees affected by the FDNPP accident. Even though studies on the circumferential distribution in the bark are scarce for the trees affected by the Chernobyl Nuclear Power Plant (ChNPP) accident, one study showed that the RSD of the  $^{137}\text{Cs}$  concentration in bark among four directions of a tree stem was 33%–47% at some sites even nearly 30 years after the accident (Cosma et al. 2016). Therefore, the circumferential distributions in bark and wood are assumed to be highly heterogeneous in some individual trees. To reduce the  $^{137}\text{Cs}$  concentration observational errors associated with partial sampling from a tree stem, it is crucial to determine whether there is a tendency for the  $^{137}\text{Cs}$  concentration to be high in a specific direction and whether the presence or absence of such a tendency differs among tree species. Additionally, it is also important to determine whether there is a correlation between the circumferential distributions in bark and wood (i.e., whether the  $^{137}\text{Cs}$  concentration in wood is high in the same direction that the concentration in bark is high) to understand the characteristics of  $^{137}\text{Cs}$  contamination in tree stems.

In addition to heterogeneities in the circumferential distribution of  $^{137}\text{Cs}$ , the shape of a core sample likely affects  $^{137}\text{Cs}$  concentration observational errors in the stem wood.

Because the radial distribution of the  $^{137}\text{Cs}$  concentration in the stem wood is often heterogeneous (e.g., Mahara et al. 2014, Ohashi et al. 2014, 2020), stem wood should be sampled as a circular sector in cross-section. However, because the shape of a core is columnar, sampling with an increment borer extracts too much wood from near the pith. This may result in an underestimation of  $^{137}\text{Cs}$  concentration in the stem wood in some cases because the concentration tends to be lower on the pith side than on the bark side (Mahara et al. 2014, Ohashi et al. 2014, 2020). Therefore, it is necessary to check whether there is a systematic error in the  $^{137}\text{Cs}$  concentration in wood cores compared with that in wood disks.

In this study, we investigated the circumferential distributions of  $^{137}\text{Cs}$  concentration in stem bark, sapwood, and heartwood collected in 2015 and examined 1) the variability of concentration among different directions, 2) the presence of a directionality that is common among individual trees, and 3) the correlation between the stem parts. Additionally, we investigated the  $^{137}\text{Cs}$  concentrations in partial samples collected without felling trees and those in bulk samples collected by felling trees in the period of 2016–2020. Finally, we discuss the validity of partial sampling for a long-term monitoring survey of  $^{137}\text{Cs}$  concentrations in a forest.



**Fig. 1. Locations of the sampling sites and deposition density of  $^{137}\text{Cs}$  (as of December 28, 2012) in Fukushima and Ibaraki Prefectures, Japan.**

Open circles indicate sampling sites, and a cross mark indicates the Fukushima Dai-ichi Nuclear Power Plant. Data on the deposition density were provided by MEXT (2013).

## 2. Materials and Methods

### 2.1 Samples for investigating the circumferential distribution of $^{137}\text{Cs}$

Samples were collected from sites 1 and 2 (Kawauchi Village, Fukushima Prefecture, Japan; Fig. 1) in August, 2015. Three individuals each of 47-year-old cedar (*Cryptomeria japonica* [L.f.] D.Don), 30-year-old cypress (*Chamaecyparis obtusa* [Siebold et Zucc.] Endl.), and 30-year-old oak (*Quercus serrata* Murray) at site 1 and 61-year-old cedar at site 2 were selected from three different diameter classes, i.e., large (L), medium (M), and small (S) diameters. The diameter at breast height (DBH) of each tree is shown in Fig. 2. Before felling the trees, the north (magnetic north) sides of the tree stems were marked with a permanent marker and the stems near breast height were wrapped in tarpaulin to prevent  $^{137}\text{Cs}$  contamination by litter and soil. Then, the stem disks were collected from the stems near breast height after felling the trees using a chain saw, and the magnetic north direction was again marked on the upper side of each disk. Later, during data processing, the ordinal directions of the stem disks were determined by assuming a magnetic declination of  $-7.5^\circ$  at the sampling sites.

To divide the disk into eight directions, we drew lines on the disk surface, making the center angle of each circular sector  $45^\circ$  (here we treated the pith as the center), except in

the cases of two small-diameter trees. The disks of the small-diameter trees, the M- and S-sized oaks from site 1, were divided into four directions. The wood disks were split using a bandsaw after collecting bark samples from each direction using a chisel. Each split disk sample was further divided into sapwood and heartwood using a chisel and a hammer. Then, all samples were chipped using a cutting mill with a 6-mm sieve and oven-dried at  $75^\circ\text{C}$  for 48 h.

### 2.2 Samples for estimating the $^{137}\text{Cs}$ concentration observational errors via partial sampling

We compared the  $^{137}\text{Cs}$  concentration in the stem wood determined from partial sampling (core sampling) with that determined from bulk sampling (disk sampling) to estimate the observational error. First, to check whether the observational error is affected by the number of cores collected from a tree, we investigated the  $^{137}\text{Cs}$  concentration determined from a single core and that determined by averaging the concentrations of two cores. Three individuals each of 48-year-old cedar (*Cryptomeria japonica*; DBH = 20.7–33.1 cm), 31-year-old cypress (*Chamaecyparis obtusa*; DBH = 14.9–20.6 cm), and 31-year-old oak (*Q. serrata*; DBH = 9.4–12.3 cm) grown at site 1 were used for this investigation. Wood cores were collected from two directions ( $180^\circ$  opposite) on each tree stem at a height of 1 m using a 12-mm-diameter increment borer in August 2016. After the core sampling, the trees were felled with a chain saw, and wood disks were collected from near the locations where the cores were collected. After separating the wood samples into sapwood and heartwood, the core and disk samples were chipped using a cutting mill with 2-mm and 6-mm sieves, respectively.

Second, to check the observational error caused by the partial sampling that we expected to use in practice, we investigated the  $^{137}\text{Cs}$  concentration in the stem wood determined from two cores roughly chipped using pruning shears instead of a cutting mill (i.e., the cores were less homogenized than cores chipped normally using a cutting mill). Using the same method previously described, core and disk samples were collected from six pines (*Pinus densiflora* Siebold et Zucc.) and three chestnuts (*Castanea crenata* Siebold et Zucc.) at site A1 (Kawauchi Village, Fukushima Prefecture, Japan) in August 2017 and 2019. The trees were approximately 50 years old and had DBH values of 17.9–38.1 cm. The samples were separated into sapwood and heartwood. Here, two cores collected from different directions on the same tree were treated as a single sample and chipped into semicircles with a thickness of 1–2 mm using pruning shears, while the disk samples were processed in the same way as described previously.

The observational error of the  $^{137}\text{Cs}$  concentration in the bark determined from partial sampling (piece sampling)

was also investigated by comparing it with that determined from bulk sampling (entire circumference sampling). Twelve pines (*P. densiflora*) and nine chestnuts (*Castanea crenata*) at site A1; six cedars (*Cryptomeria japonica*), three pines (*P. densiflora*), and three oaks (*Q. serrata*) at site 3 (Otama Village, Fukushima Prefecture, Japan); and three cypresses (*Chamaecyparis obtusa*) at site 5 (Ishioka City, Ibaraki Prefecture, Japan) were used for this investigation. The trees were approximately 50 years old and had DBH values of 16.6–38.1 cm. In July–September of 2017–2020, we collected bark pieces (approx. 3 cm in the axial and tangential directions) from four directions (approx. 90° apart from each other) of each tree stem at a height of 1 m using a 3-cm-wide chisel and a hammer. We then felled the trees and collected bark from the entire circumference of the tree stems using the same method as in the monitoring survey by the Forestry and Forest Products Research Institute (FFPRI; Kuroda et al. 2013). The bark pieces collected from different directions on the same tree were treated as a single sample and cut into approximately 5-mm pieces in the axial and tangential directions using pruning shears, while the bark samples collected from the entire circumference were chipped using a cutting mill with a 6-mm sieve.

The samples were oven-dried at 75 °C for 48 h, and the sample weights were converted to that dried at 105 °C by multiplying by 0.98 for the bark samples and by 0.99 for the wood samples (Ohashi et al. 2017).

### 2.3 Radioactivity measurements

The samples were packed into 2-L or 0.7-L Marinelli containers (MAX-Y20 or MAX-Y07, Sugiyama-gen, Tokyo, Japan), U-8 containers (No. 3-20, Umano Kagaku Youki, Osaka, Japan), or #737 containers (Kartell, Milan, Italy) depending on the sample amount. The radioactivity of <sup>137</sup>Cs in the sample was determined via gamma-ray spectrometry with a high-purity Ge detector (GEM20, GEM40, or GWL-120, ORTEC, Oak Ridge, TN). Peak efficiency calibrations for the Marinelli, U-8, and #737 containers were done using standard sources: MX033MR, MX033U8PP (Japan Radioisotope Association, Tokyo, Japan), and EG-ML (Eckert & Ziegler Isotope Products, Valencia, CA), respectively. Each measurement was continued until the counting error dropped to 5% or less; however, for some wood core samples with small amounts and low <sup>137</sup>Cs concentrations, we allowed an error of up to 10%. For measurements combining the well-type Ge detector (GWL-120) and the #737 container, the gamma-ray self-absorption was corrected by assuming that the bulk densities of the samples were the same as that of the standard source (Ohashi et al. 2021).

### 2.4 Estimation of <sup>137</sup>Cs concentration observational errors via partial sampling

To evaluate the difference between the <sup>137</sup>Cs concentrations determined from partial sampling ( $X$ ) and those determined from bulk sampling ( $Y$ ), the mean absolute percentage error (MAPE) was calculated according to the following equation:

$$\text{MAPE} = \frac{100}{n} \sum_{k=1}^n \left| \frac{X_k - Y_k}{Y_k} \right|$$

where  $n$  is the number of samples.

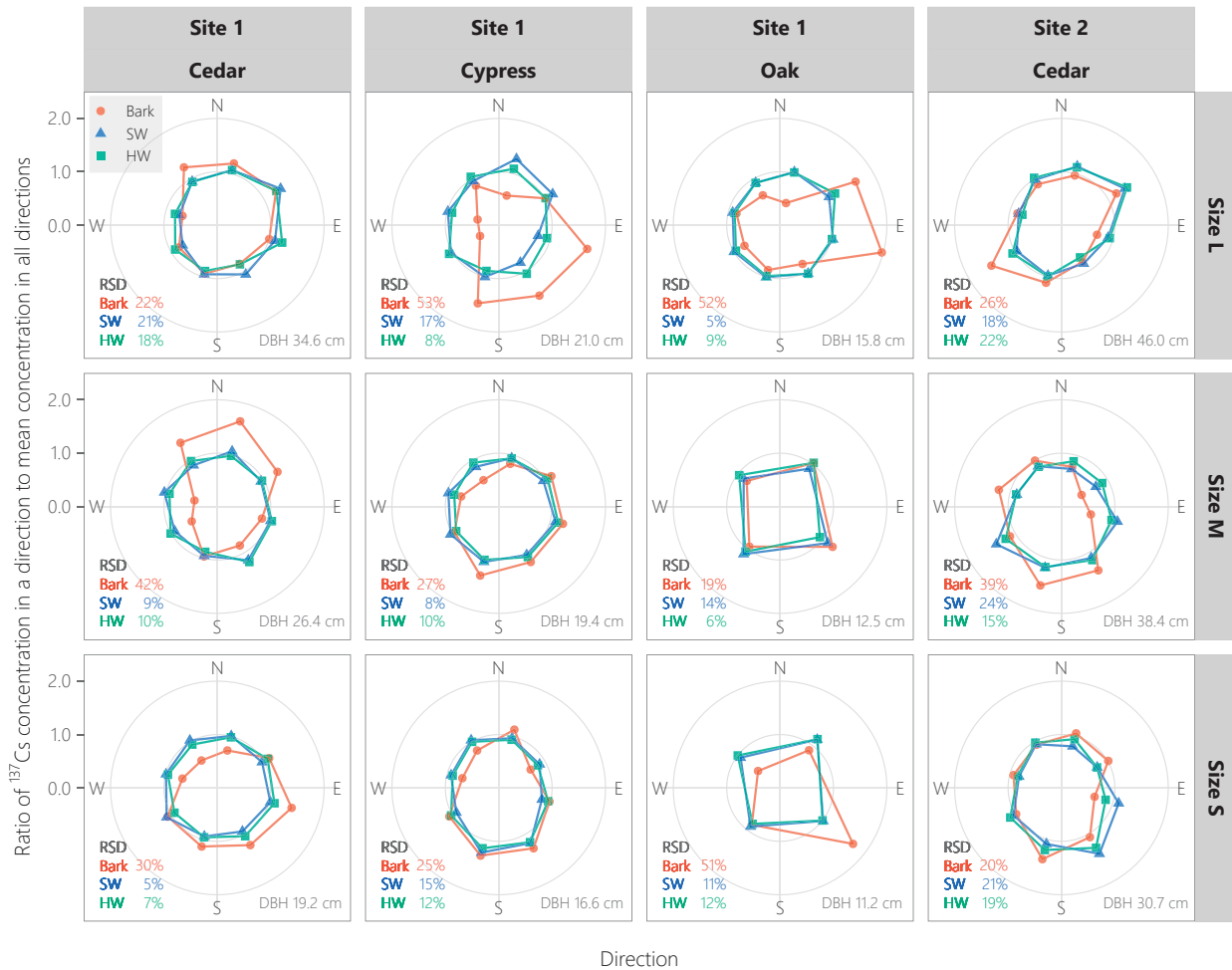
Because not only  $X$  but also  $Y$  contains the observational error (at least the error caused by the radioactivity measurement), we estimated the observational error in  $X$  by simulating MAPE with the assumption that the observational error in  $Y$  was 5% (counting error of the radioactivity measurement). The simulation was done by generating 100,000 random numbers from a normal distribution (mean = 100, standard deviation = 5) for  $Y$  and 100,000 random numbers from the normal distribution (mean = 100, standard deviation =  $\sigma$ ) for  $X$  using R version 4.0.4 (R Core Team 2021). Various values of  $\sigma$  were tested sequentially to search for the optimum value that derives the nearest MAPE to the observed MAPE. We determined the optimum value of  $\sigma$  to be the observational error in  $X$ .

## 3. Results and Discussion

### 3.1 Circumferential distribution of <sup>137</sup>Cs

The circumferential distributions of the <sup>137</sup>Cs concentration in the bark, sapwood, and heartwood collected in 2015 are shown in Fig. 2. The mean RSD values of the <sup>137</sup>Cs concentration among the directions were 34%, 14%, and 12% in the bark, sapwood, and heartwood, respectively. The relatively large RSD in the bark is likely due to the heterogeneous deposition of accident-derived <sup>137</sup>Cs (Tanaka et al. 2013), the non-uniform removal of <sup>137</sup>Cs by stemflow with preferential flow paths (Imamura et al. 2017a), and the random exfoliation of bark via aging and weathering. The RSD values in the sapwood and heartwood observed in this study are relatively small compared with previous studies that showed an approximately 10%–50% RSD among the directions (Mahara et al. 2014, Hirano et al. 2016).

The RSD of the <sup>137</sup>Cs concentration among the directions did not vary significantly among the tested species and between the sites. Even though the <sup>137</sup>Cs concentration in the bark showed biases in the circumferential distributions in some trees (e.g., the M-size cedar, L-size cypress, and L-size oak at site 1 and L-size cedar at site 2), the directions with high <sup>137</sup>Cs concentrations were not the same for all trees. In contrast to our



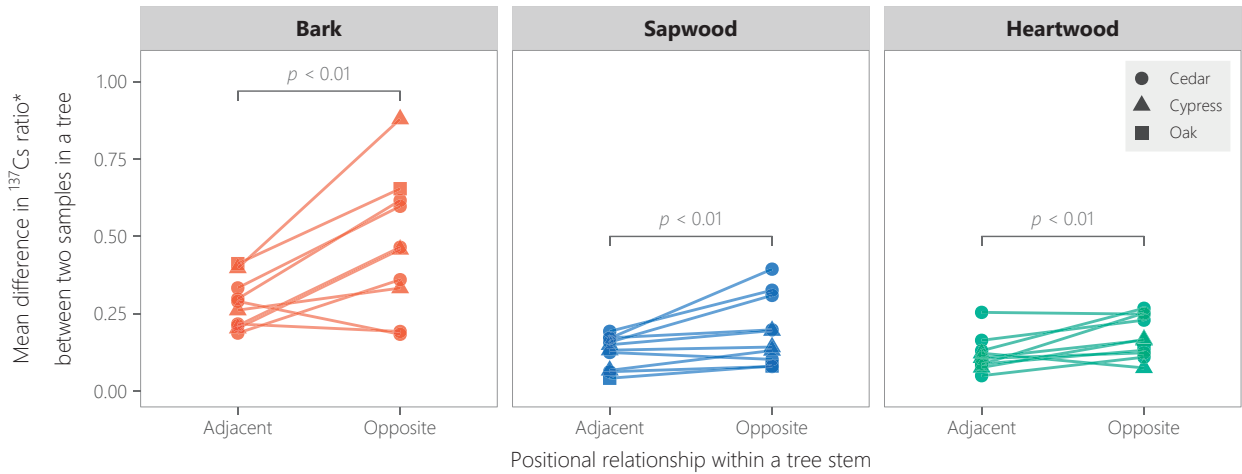
**Fig. 2. Circumferential distributions of the <sup>137</sup>Cs concentrations in the bark and wood disk samples in 2015.**  
*SW sapwood, HW heartwood, RSD relative standard deviation (among the directions), DBH diameter at breast height*

results, Mahara et al. (2014) reported that <sup>137</sup>Cs concentration in the stem wood of an oak (*Q. serrata*) collected in 2012 was high in the direction facing the wind direction during the main <sup>137</sup>Cs deposition event. They inferred that such anomalous directionality may appear in deciduous tree species such as oak, which did not absorb <sup>137</sup>Cs via their leaves in 2011, and predicted that this directionality would soon disappear as a consequence of the transport and diffusion of <sup>137</sup>Cs. Therefore, directionality might have also existed at our study sites shortly after the accident; however, four years after the accident, we could not find any evidence of directionality. The relatively small RSD in this study also indicates that the circumferential distribution of <sup>137</sup>Cs concentration in the stem wood may have been homogenized with time. Similarly, directionality was not found in oaks (*Q. petraea*) contaminated by the ChNPP accident 24 years after the accident (Kilic 2012).

Although there was no common directionality in the <sup>137</sup>Cs concentration among the trees, there was a spatial autocorrelation in the <sup>137</sup>Cs concentration within a tree stem. The difference in the <sup>137</sup>Cs concentrations between two adjacent directions tended to be smaller than between two opposite

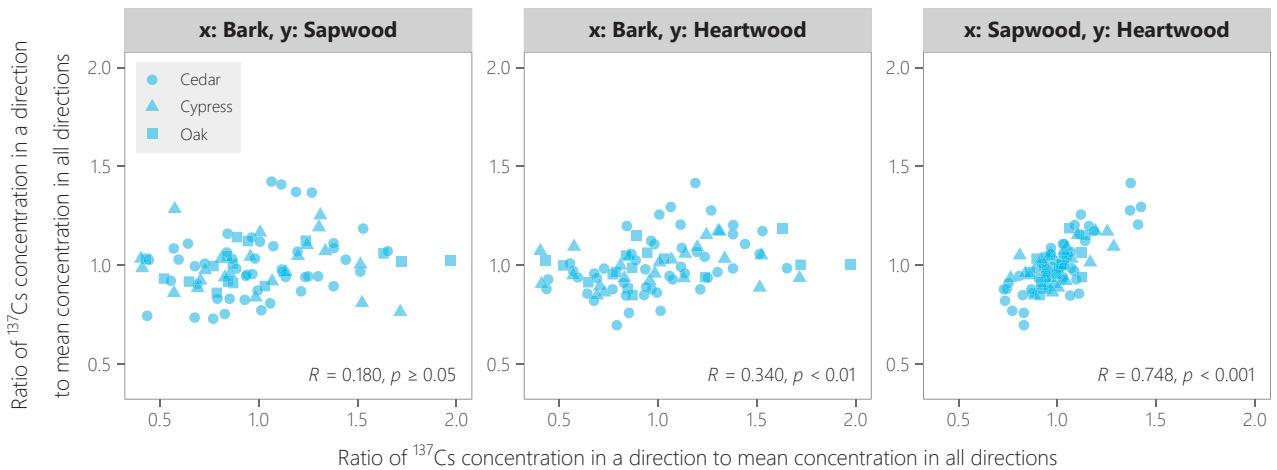
directions in all stem parts (bark, sapwood, and heartwood) (Fig. 3). This fact indicates that sampling from more than one direction of a tree stem should be done by collecting samples from parts apart from each other to obtain representative data of an individual tree, especially for the bark, which showed a relatively large RSD of <sup>137</sup>Cs concentration in the circumferential direction.

Correlations in the circumferential distribution patterns of the <sup>137</sup>Cs concentration between the stem parts (bark, sapwood, and heartwood) were checked using the Pearson product-moment correlation coefficient (Fig. 4). There was no correlation between the distribution patterns in the bark and those in the sapwood ( $R = 0.180, p \geq 0.05$ ). Even though it has been shown experimentally that Cs can be absorbed from bark surface and transferred to sapwood (Wang et al. 2016, 2018), we assumed that <sup>137</sup>Cs absorption through bark had not occurred dominantly since 2012 because dissolved <sup>137</sup>Cs on tree surfaces, which is thought to be absorbed by trees, decreased drastically during 2011 (Nishikiori et al. 2015). The lack of a correlation between the bark and the sapwood appears to support this assumption. However, there was a weak



**Fig. 3. The difference in  $^{137}\text{Cs}$  ratio\* between two directions within a tree stem in 2015.**

\* Ratio of  $^{137}\text{Cs}$  concentration in a direction to mean concentration in all directions (the same variable with the y-axis of Fig. 2). The trees of which stems were divided into eight directions were analyzed ( $n = 10$ ). Each point shows a mean difference in the  $^{137}\text{Cs}$  ratio between two directions (eight combinations for adjacent position and four combinations for opposite position) in a tree.  $p$ -values indicate the significance level of the left-tailed, paired t-test.

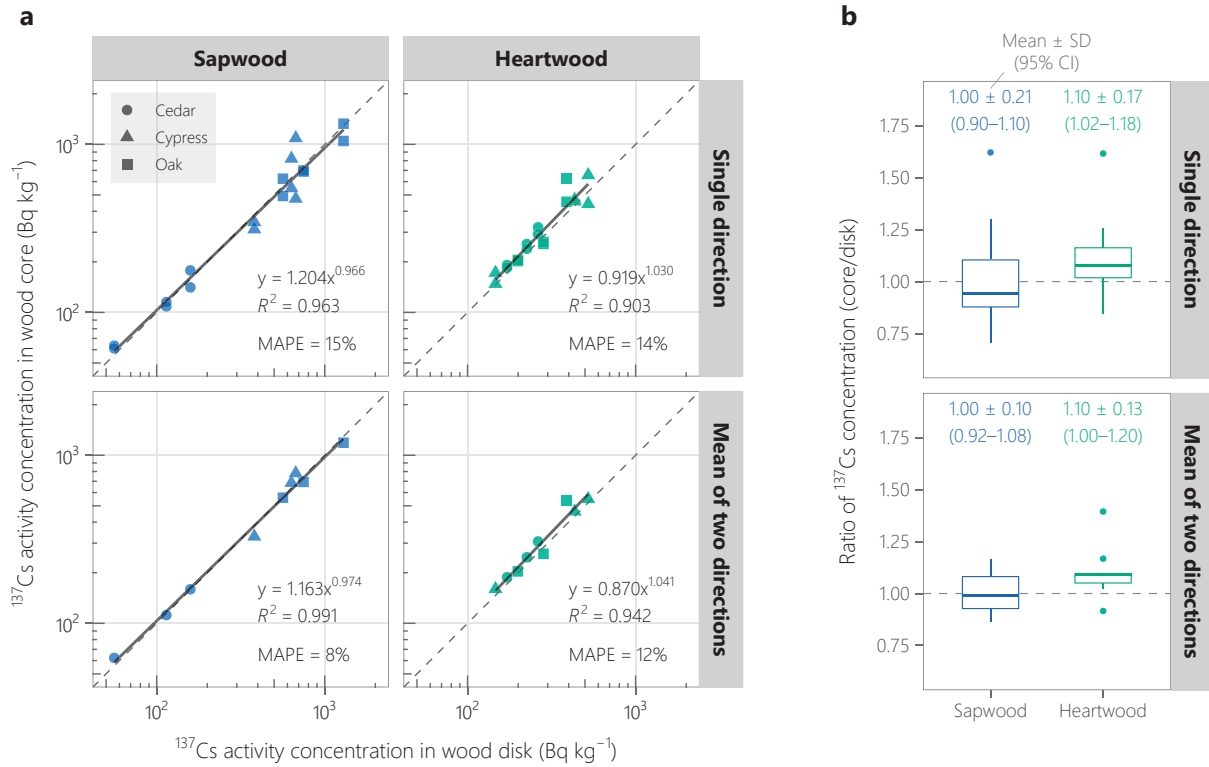


**Fig. 4. Correlation of the circumferential distribution patterns of the  $^{137}\text{Cs}$  concentration between the stem parts in 2015.**

correlation between the distribution patterns in the bark and those in the heartwood ( $R = 0.340, p < 0.01$ ), implying that  $^{137}\text{Cs}$  might have been transferred intensively from the bark surface to the stem wood in 2011 and remained somewhat in the heartwood. Such a “memory effect” in the heartwood was also suggested in our previous study comparing the  $^{137}\text{Cs}$  and  $^{133}\text{Cs}$  distributions in tree stems collected in 2014 (Ohashi et al. 2020). However, the correlation between the bark and the heartwood is thought to become weaker with time as the formation of new heartwood and the diffusion of  $^{137}\text{Cs}$  in the heartwood proceeds. Lastly, there was a strong correlation between the distribution patterns in the sapwood and those in the heartwood ( $R = 0.748, p < 0.001$ ). This strong correlation indicates that  $^{137}\text{Cs}$  transfer in stem wood is likely to be more dominant in the radial direction than in the circumferential (tangential) direction, probably owing to the existence of ray (Kuroda et al. 2020).

### 3.2 $^{137}\text{Cs}$ concentration observational error via partial sampling

The  $^{137}\text{Cs}$  concentration in the sapwood determined via partial sampling (core sampling) had a one-to-one correspondence with that determined via bulk sampling (disk sampling) in 2016, regardless of the number of cores collected from a tree (Fig. 5a). Because the ratio of the  $^{137}\text{Cs}$  concentration in the core to the disk was 1.00 on average (Fig. 5b), we determined that there was no systematic error in the  $^{137}\text{Cs}$  concentration in the sapwood core. However, the ratio in the heartwood was 1.10 on average and its 95% confidence interval ranged to values greater than 1.00 (Fig. 5b), showing that there was a positive systematic error (overestimation) in the  $^{137}\text{Cs}$  concentration in the heartwood core. This result is contrary to our assumption that there can be a negative systematic error (underestimation) in the  $^{137}\text{Cs}$  concentration in the inner part (heartwood) of a wood core because the core contains too much wood near the pith, in which the  $^{137}\text{Cs}$

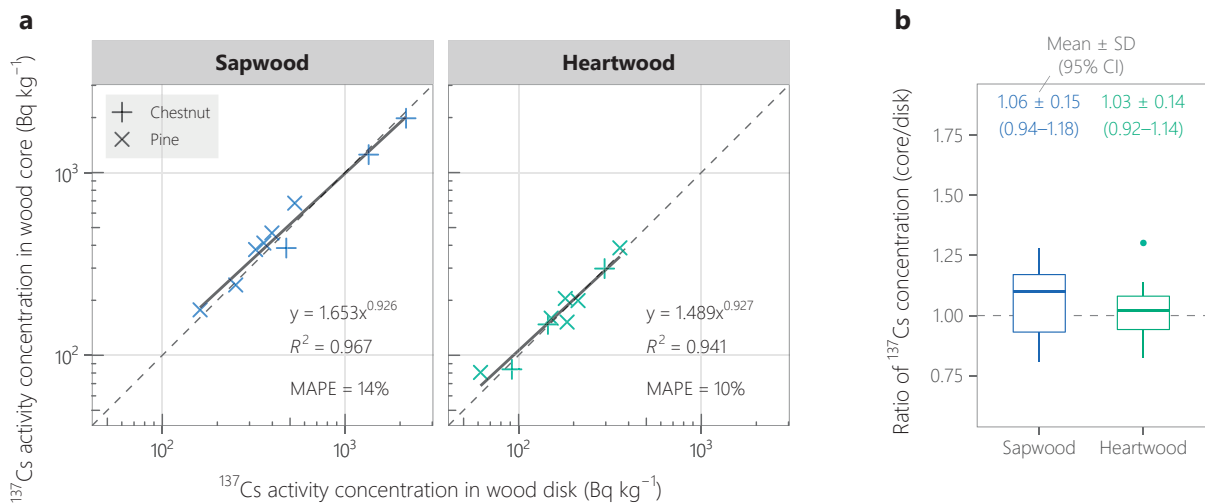


**Fig. 5. Comparison of the <sup>137</sup>Cs concentrations in the wood disk and core samples (single direction or the mean of two directions) in 2016.**

The disk and core samples were chipped using a cutting mill with 6-mm and 2-mm sieves, respectively. *MAPE* mean absolute percentage error, *SD* standard deviation, *CI* confidence interval

concentration is relatively low in many cases (e.g., Mahara et al. 2014, Ohashi et al. 2014, 2020). However, there are some cases in which the <sup>137</sup>Cs concentration in wood near the pith is higher than that on the outer side (Ogawa et al. 2016, Ohashi et al. 2020). Therefore, we might have collected samples from such trees in 2016 by chance because there was no systematic

error in the <sup>137</sup>Cs concentration in the heartwood cores in the period of 2017–2019 (Fig. 6). Unfortunately, the species dependency of the systematic error is difficult to discuss in this study because of the small sample size and the uncertainty in estimating the <sup>137</sup>Cs concentration in heartwood from the partial sampling remains. Nevertheless, because the radial distribution



**Fig. 6. Comparison of the <sup>137</sup>Cs concentrations in the wood disk and core samples (mean of two directions) in the period of 2017–2019**

The disk samples were chipped using a cutting mill with a 6-mm sieve, and the core samples were roughly chipped using pruning shears. *MAPE* mean absolute percentage error, *SD* standard deviation, *CI* confidence interval

of the  $^{137}\text{Cs}$  concentration in heartwood is presumed to become homogeneous with time following an accident (Ogawa et al. 2016), we expect that the uncertainty is smaller or negligible in recent samples (e.g., samples collected 10 years after the accident).

The MAPEs of the  $^{137}\text{Cs}$  concentration in the sapwood and heartwood determined from a single core were 14%–15%, and the observational errors were estimated to be 16%–18%. These errors likely arose from the variability of the  $^{137}\text{Cs}$  concentration in the circumferential direction because they are similar to the mean RSD of the  $^{137}\text{Cs}$  concentration among the directions (12%–14%, as described in Section 3.1). When averaging the  $^{137}\text{Cs}$  concentrations in two cores (directions), the observational errors decreased to 8%–14%. This is consistent with the theoretical error in the mean value for two directions, approximately 12%, which can be calculated using the law of error propagation and by assuming an observational error of 17% (16%–18%, as mentioned above) (i.e.,  $\sqrt{17^2 + 17^2} \div 2$ ). Therefore, the reduction in the  $^{137}\text{Cs}$  concentration observational error when increasing the number of cores was demonstrated well in this study. In the case of increasing the number of cores to 4, 8, and 16, the observational error is expected to be reduced to 9%, 6%, and 4%, respectively (Table 1).

There was no systematic error in the sapwood and heartwood  $^{137}\text{Cs}$  concentrations determined from the two cores that were roughly chipped using pruning shears (Fig. 6). The observational errors were estimated to be 12%–17%, slightly

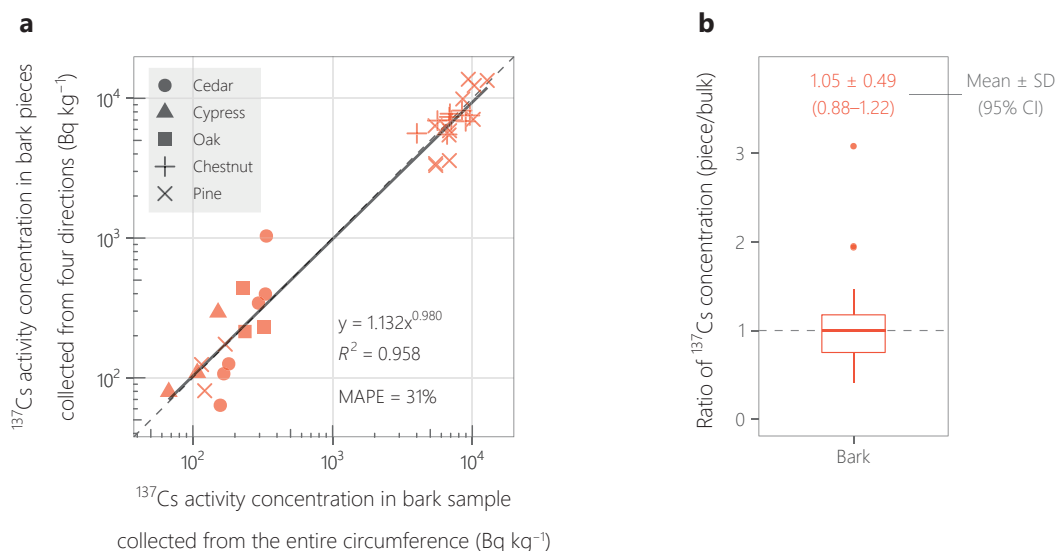
larger than those in the concentrations determined from two normally chipped cores (8%–14%) but smaller than those in the concentrations determined from a single normally chipped core (16%–18%). Therefore, this relatively simple method using two roughly chipped cores appears to be reasonable for radioactivity measurement and smaller observational errors than using a single normally chipped core, as well as taking less time for sample processing than chipping cores with a cutting mill.

The  $^{137}\text{Cs}$  concentration in the bark determined from the partial sampling (piece sampling) also had a one-to-one correspondence with that determined from the bulk sampling (sampling from the entire circumference), and no systematic error was observed in the period of 2017–2020 (Fig. 7); however, the observational error was estimated to be 38%, showing that the random error is relatively large. This error is considerably larger than the theoretical error in the mean value

**Table 1. Estimates of the observational error in the  $^{137}\text{Cs}$  concentration determined via partial sampling of bark and wood from a tree stem.**

Part	Number of partial samples per tree				
	1	2	4	8	16
Bark	76%	54%	38%	27%	19%
Wood	17%	12%	9%	6%	4%

The estimates were calculated using the law of error propagation and assuming that observational error via sampling bark pieces (3 cm × 3 cm) from four directions is 38% and that via sampling a wood core (diameter 12 mm) from a single direction is 17%.



**Fig. 7. Comparison of the  $^{137}\text{Cs}$  concentrations in the bark samples collected from the entire circumference and from four directions of the tree stems in the period of 2017–2020.**

The samples collected from the entire circumference were chipped using a cutting mill with a 6-mm sieve, and the samples collected from four directions were cut into approximately 5-mm pieces in the axial and tangential directions using pruning shears.

MAPE mean absolute percentage error, SD standard deviation, CI confidence interval



**Table 2. Sample size (number of sample trees) required to achieve the same relative standard error of the <sup>137</sup>Cs concentration in an investigation in which the observational error in the <sup>137</sup>Cs concentration is 5% and the sample size is 3.**

RSD among trees <sup>1)</sup> (%)	RSE <sup>2)</sup> (%)	Observational error in <sup>137</sup> Cs concentration				
		10%	15%	20%	30%	40%
0	3	12	27	48	108	192
5	4	7	15	25	55	98
10	6	5	8	12	24	41
20	12	4	4	6	9	14
30	18	3	4	4	6	8
40	23	3	3	4	5	6
50	29	3	3	3	4	5

1) Relative standard deviation of the <sup>137</sup>Cs concentration among trees within a forest stand.

2) Relative standard error of the mean value of the <sup>137</sup>Cs activity concentration within a forest stand when the observational error in the <sup>137</sup>Cs concentration is 5% and the sample size is 3.

for four directions, 17%, calculated by assuming that the RSD of the <sup>137</sup>Cs concentration in the bark among the directions was 34%, from the results in Section 3.1 (i.e.,  $\sqrt{34^2 \times 4} \div 4$ ). This gap is most likely due to the small piece sizes (3 cm × 3 cm). Therefore, to reduce the <sup>137</sup>Cs concentration observational error in bark determined via partial sampling, it is necessary to increase the size of the bark pieces collected. If it is difficult to increase the size, increasing the number of bark pieces will also reduce the observational error. For example, the observational error is estimated to be reduced to 27% when bark pieces are collected from eight directions (Table 1).

### 3.3 Sampling strategy

The partial sampling of bark and wood from a standing tree was demonstrated to increase the observational error (which is considered random and unsystematic) of the <sup>137</sup>Cs concentration compared with bulk sampling from a felled tree. Whether this increase in the observational error is acceptable or not depends on the objective of a study. For example, in a case where the objective is to estimate the <sup>137</sup>Cs concentration in the bark or wood of a single tree, the concentration should be determined from bulk sampling with a felled tree, or if it is difficult to fell the tree, determined via partial sampling with a large number of bark pieces or wood cores from the tree (Table 1).

In contrast, in a case where the objective is to estimate the mean value of the <sup>137</sup>Cs concentration in a forest stand, the estimation error (i.e., the standard error) increased by partial sampling from the trees can be canceled or even become smaller by increasing the number of sample trees (the sample size). The sample size for partial sampling required to achieve the same relative standard error as bulk sampling depends on the sample size, the observational error of the <sup>137</sup>Cs concentration in the bulk sampling, and the RSD of the <sup>137</sup>Cs concentration among the trees in the forest stand. Here, based on the monitoring survey conducted by FFPRI from 2011 to 2016 (Kuroda et al. 2013, Imamura et al. 2017b, Ohashi

et al. 2017), we assumed a sample size of the bulk sampling of three, an observational error of the bulk sampling of 5% (the error caused by the counting error of the radioactivity measurement), and a RSD among the trees of roughly 30%–40% (Fig. S1). The sample size required for an observational error of 10%–20% (assuming the error caused by the partial sampling of the sapwood and heartwood from two directions) in this case was estimated to be 3–4 (Table 2). This is nearly the same sample size with bulk sampling, meaning that the partial sampling of stem wood is acceptable for estimating the mean value of the <sup>137</sup>Cs concentration in a forest stand without increasing the sample size compared with bulk sampling in this case. For another example, the sample size required for an observational error of 40% (assuming the error caused by the partial sampling of the bark) was estimated to be 6–8 (Table 2). Therefore, even though the observational error caused by partial sampling is relatively large, the mean value of the <sup>137</sup>Cs concentration in a forest can be estimated with the same accuracy as bulk sampling by increasing the sample size in a realistic range.

However, there are also limitations in partial sampling as well as bulk sampling. Frequent sampling of bark and wood from the same tree reduces transport pathways of water, photosynthate, and minerals, and might alter the <sup>137</sup>Cs concentration in the remaining stem parts, or results in tree death in the worst case. It is uncertain how many samples we can collect without affecting the tree significantly. Therefore, to sustain a monitoring program for decades by partial sampling, it would be better to change sample trees every year within a forest stand and assign enough time interval to trees for bark regeneration after sampling.

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#### Supplementary data

Supplementary data can be found at

<https://www.ffpri.affrc.go.jp/pubs/bulletin/461/461toc-en.html>

Fig. S1. Relative standard deviation (RSD) of the <sup>137</sup>Cs activity concentration in bark, sapwood, and heartwood among trees within a forest stand observed in the monitoring data from 2011 to 2016. *Circles* indicate the mean RSD values in each sampling year, and *error bars* indicate their 95% confidence intervals (*n* of the trees in the forest stand = 3; *n* of the forest stands = 7–9). The monitoring data were provided in the studies of Kuroda et al. (2013), Imamura et al. (2017b), and Ohashi et al. (2017). The species contained in the data are cedar (*Cryptomeria japonica*), cypress (*Chamaecyparis obtusa*), pine (*Pinus densiflora*), and oak (*Quercus serrata*).

# 福島原発事故で汚染された樹幹からの部分的なサンプリングによって生じる 樹皮および材中の<sup>137</sup>Cs濃度の観測誤差

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## 要旨

福島第一原子力発電所事故によって汚染した樹皮や材の放射性セシウム (<sup>137</sup>Cs) 濃度を長期的にモニタリングするためには、試料は樹木を伐倒せずに立木から部分的に採取することが望ましい。しかし、樹皮や材中の<sup>137</sup>Csの分布は均一ではないと想定されるため、部分的なサンプリングによる観測誤差を明らかにし、その妥当性を確認する必要がある。本研究では、樹皮および材の<sup>137</sup>Cs濃度の樹幹円周方向の分布を明らかにするとともに、部分的なサンプリングによる樹皮および材の<sup>137</sup>Cs濃度の観測誤差を明らかにすることを目的とした。2015年に伐採した樹幹を8方位(一部4方位)に分割して求めた<sup>137</sup>Cs濃度の円周方向分布には、樹皮と材でそれぞれ平均34%と13%の相対標準偏差があったが、特定の方位への偏りや樹種依存性は見られなかった。2016年以降に樹幹から部分的にサンプリングした試料(樹皮は3cm×3cmのピース×4方向、材は直径12mmのコア×1-2方向)から求めた<sup>137</sup>Cs濃度には、樹皮と材でそれぞれ約38%と約8-18%の観測誤差があると推計され、いずれも偶然誤差だと考えられた。例えばこの部分的なサンプリングによって<sup>137</sup>Cs濃度の林分平均値を伐倒サンプリング(観測個体数3)と同程度の精度で推定するためには、樹皮の場合は観測個体数を6-8に増やす必要があることがわかった。

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