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Temporal trends in ^{137}Cs concentrations in the bark, sapwood, heartwood, and whole wood of four tree species in Japanese forests from 2011 to 2016

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ABSTRACT

To understand the changes in radiocesium (^{137}Cs) concentrations in stem woods after the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident, we investigated ^{137}Cs concentrations in the bark, sapwood, heartwood, and whole wood of four major tree species at multiple sites with different levels of radiocesium deposition from the FDNPP accident since 2011 (since 2012 at some sites): Japanese cedar at four sites, hinoki cypress and Japanese konara oak at two sites, and Japanese red pine at one site. Our previous report on ^{137}Cs concentrations in bark and whole wood samples collected from 2011 to 2015 suggested that temporal variations were different among sites even within the same species. In the present study, we provided data on bark and whole wood samples in 2016 and separately measured ^{137}Cs concentrations in sapwood and heartwood samples from 2011 to 2016; we further discussed temporal trends in ^{137}Cs concentrations in each part of tree stems, particularly those in ^{137}Cs distributions between sapwood and heartwood, in relation to their species and site dependencies. Temporal trends in bark and whole wood samples collected from 2011 to 2016 were consistent with those reported in samples collected from 2011 to 2015. Temporal variations in ^{137}Cs concentrations in barks showed either a decreasing trend or no clear trend, implying that ^{137}Cs deposition in barks is inhomogeneous and that decontamination is relatively slow in some cases. Temporal trends in ^{137}Cs concentrations in sapwood, heartwood, and whole wood were different among species and also among sites within the same species. Relatively common trends within the same species, which were increasing, were observed in cedar heartwood, and in oak sapwood and whole wood. On the other hand, the ratio of ^{137}Cs concentration in heartwood to that in sapwood (fresh weight basis) was commonly increased to more than 2 in cedar, although distinct temporal trends were not found in the other species, for which the ratio was around 1 in cypress and pine and below 0.5 in oak, suggesting that ^{137}Cs transfer from sapwood to heartwood shows species dependency. Consequently, the species dependency of ^{137}Cs transfer within the tree appears easily, while that from the environment to the trees can be masked by various factors. Thus, prediction of ^{137}Cs concentrations in stem wood should be carried out carefully as it still requires investigations at multiple sites with a larger sample size and an understanding of the species-specific ^{137}Cs transfer mechanism.

1. Introduction

Recent studies have confirmed the fact that radiocesium (^{137}Cs and ^{134}Cs) derived from the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident has transferred to the stem wood of trees (e.g., Kuroda et al., 2013; Mahara et al., 2014; Ohashi et al., 2014; Masumori et al., 2015). By 6 months after the accident at the

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latest, radiocesium transferred not only to the sapwood (outer, living part of stem wood) but also to the heartwood (inner, dead part of stem wood) of Japanese cedar (*Cryptomeria japonica*), oak (*Quercus serrata*), and pine (*Pinus densiflora*) (Kuroda et al., 2013). Furthermore, radiocesium was detected in the inner heartwood near the pith (Mahara et al., 2014; Ohashi et al., 2014; Ogawa et al., 2016). Despite such rapid and inward transfer of radiocesium in these woods, reports of periodic monitoring are scarce.

In forest ecosystems, the inventory of radiocesium in stem wood is quite small, usually less than 2% (Tikhomirov and Shcheglov, 1994; Mamikhin et al., 1997; Komatsu et al., 2016; Imamura et al., 2017). However, the national interim permissible levels for radiocesium concentration in wood for some uses in Japan are currently low; e.g., 40 Bq kg^{-1} (dry weight basis) for firewood for cooking and 50 Bq kg^{-1} for logs for mushroom cultivation (Forestry Agency, 2011, 2012). This implies that even low-level contamination can limit the use of wood for decades or more. Thus, understanding the temporal changes in radiocesium concentrations in stem woods is critically important for forest planning and management in Fukushima and neighboring prefectures.

The distribution pattern of radiocesium concentration within stem wood and its temporal change could be different among species. In the case of the Chernobyl Nuclear Power Plant (CNPP) accident, the stem woods of Scots pine (*P. sylvestris*) and silver birch (*Betula pendula*) had different radial ^{137}Cs distribution patterns 10 years after the accident (Soukhova et al., 2003). For Japanese tree species, although Cs distribution data are limited, the distribution pattern of K concentration in stem wood is known to vary and is categorized into the following three types; type 1 has higher concentrations in sapwood than those in heartwood, type 2 has higher concentrations in heartwood than those in sapwood, and type 3 shows a peak in concentration at the sapwood–heartwood boundary (Okada et al., 1993a, 1993b). Recent studies conducted several years after the FDNPP accident (Mahara et al., 2014; Ohashi et al., 2014; Ogawa et al., 2016) suggest that radiocesium distribution patterns in cedars (*Cryptomeria japonica*) would be type 2 and those in oaks (*Q. serrata*) and pines (*P. densiflora*) would be type 1. However, 6 months after the accident, the radiocesium concentration in cedars was higher in sapwood than that in heartwood (Kuroda et al., 2013), and this distribution changed during the first 3 years at least (Ogawa et al., 2016). These facts emphasize the necessity for continuous monitoring of radiocesium distribution in the stem woods of each major species.

The difference of radiocesium behavior among sites is also a matter of great interest. The ^{137}Cs transfer factor from soil to stem wood has been reported to differ among soil types, moisture regimes, and distance from the CNPP (Shcheglov et al., 2001). Furthermore, ^{137}Cs distribution patterns in stem wood may also differ with soil type (Soukhova et al., 2003). However, following the FDNPP accident, most studies have been conducted at a single site, with a single species, or for a single year. For a better understanding of radiocesium contamination in stem wood, continuous and comparable monitoring of multiple species at multiple sites is required. Our institute, Forestry and Forest Products Research Institute (FFPRI), has consistently investigated the ^{137}Cs distribution among tree needle/leaf, branch, bark, stem wood, organic layer, and mineral soil layer in nine forests after the FDNPP accident: Japanese cedar (*Cryptomeria japonica* D.Don) at four sites, hinoki cypress (*Chamaecyparis obtusa* Endl.) and Japanese konara oak (*Quercus serrata* Murray) at two sites and Japanese red pine (*Pinus densiflora* Sieb. & Zucc.) at one site. Regarding the bark and stem wood, we have reported data on cedar, oak, and pine samples collected in 2011 (Kuroda et al., 2013) and also those on the four species collected every year from 2011 to 2015 (Imamura et al., 2017). In the latter report, we suggested that temporal trends in ^{137}Cs concentrations in barks and stem woods were different among sites even within the same species.

In the present study, reporting new data on ^{137}Cs concentrations in bark and whole wood samples collected in 2016 and separately measuring ^{137}Cs concentrations in sapwood and heartwood samples collected every year from 2011 to 2016 (from 2012 at some sites), we discussed temporal trends in ^{137}Cs concentrations in each part of tree stems, particularly those in ^{137}Cs distributions between sapwood and heartwood, in relation to their species and site dependencies.

2. Material and methods

The study sites were located in Kawauchi Village (sites 1 and 2), Otama Village (site 3), and Tadami Town (site 4) of Fukushima Prefecture and in Mt. Tsukuba (site 5) of Ibaraki Prefecture (Fig. 1). Sites 1–5 correspond to sites KU1, KU2, OT, TD, and TB in the study by Imamura et al. (2017), respectively. These sites received differing levels of radiocesium deposition from the FDNPP accident (Fig. 1, Table 1). Mean temperatures in the areas are 10.0°C–13.8 °C, and annual precipitation is 1213–1465 mm in all areas except Tadami, where the annual precipitation is 2368 mm because of heavy snow during winter (data are mean values from 1981 to 2010; Japan Meteorological Agency, 2016). The soils at the sites are classified as Brown forest or Black soils.

Four major tree species, cedar (*Cryptomeria japonica*), cypress

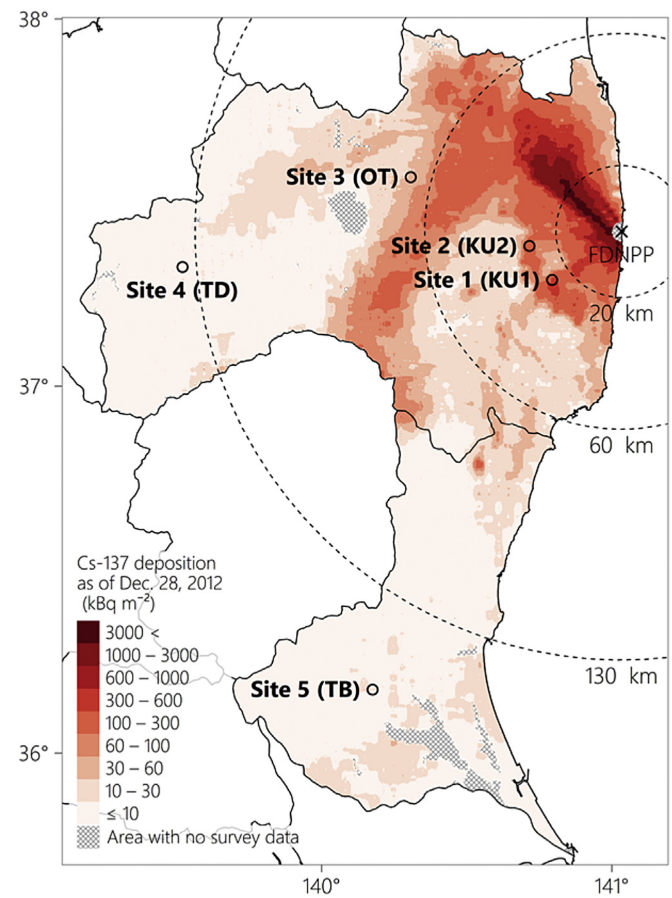

Fig. 1. Locations of study sites (*open circle*) and ^{137}Cs deposition (decay-corrected to December 28, 2012; MEXT, 2013) in Fukushima and Ibaraki Prefectures, Japan. *Broken-line circles* show distances from the Fukushima Dai-ichi Nuclear Power Plant (FDNPP, *marked with a cross*). Site names in parentheses correspond to those in the study of Imamura et al. (2017).

Table 1
Description of sample trees and study sites.

Site [a]	Species		Leaf habit	Age (y) [b]	DBH (cm) [c]	Height (m) [c]	^{137}Cs deposition (kBq m^{-2}) [d]
Site 1 (KU1)	Cedar	*Cryptomeria japonica*	Evergreen	43	25.5 ± 7.1	17.5 ± 2.8	390
	Cypress	*Chamaecyparis obtusa*	Evergreen	26	17.8 ± 2.4	10.8 ± 0.7	370
	Oak	*Quercus serrata*	Deciduous	26	12.5 ± 2.5	11.0 ± 1.2	370
Site 2 (KU2)	Cedar	*Cryptomeria japonica*	Evergreen	57	38.3 ± 8.1	24.8 ± 2.3	140
Site 3 (OT)	Cedar	*Cryptomeria japonica*	Evergreen	42	27.0 ± 5.5	21.2 ± 1.6	36
	Oak	*Quercus serrata*	Deciduous	43	22.1 ± 5.8	15.0 ± 1.6	38
	Pine	*Pinus densiflora*	Evergreen	43	24.5 ± 5.8	15.4 ± 1.0	38
Site 4 (TD)	Cedar	*Cryptomeria japonica*	Evergreen	38	25.1 ± 5.1	18.6 ± 3.4	<10
Site 5 (TB)	Cypress	*Chamaecyparis obtusa*	Evergreen	43	22.8 ± 5.2	16.6 ± 1.3	11

DBH: diameter at breast height (1.3 m).
[a] Site names in parentheses correspond to those in the study of Imamura et al. (2017).
[b] Tree age of sample trees as of 2011.
[c] Mean and standard deviation of sample trees.
[d] Data of the sixth airborne monitoring survey and airborne monitoring survey outside 80 km from the FDNPP, decay-corrected to December 28, 2012 (MEXT, 2013).

(*Chamaecyparis obtusa*), oak (*Q. serrata*), and pine (*P. densiflora*), were selected for study. Cedar and cypress are the most important timber species in Japan, although the population of cypress is small in Fukushima as it is the northern limit of its natural distribution. Oak is a dominant species in secondary forests and is in high demand as logs for shiitake mushroom cultivation. Pine is not only an important timber species in plantations but also a dominant species in secondary forests, providing valuable matsutake mushrooms with ectomycorrhizal fungi. The leaf habit of cedar, cypress, and pine is evergreen and that of oak is deciduous.

Nine permanent study plots (0.06–0.24 ha plot^{-1} in area) were established as follows: three plots each at sites 1 (cedar, cypress, and oak) and 3 (cedar, oak, and pine) and one plot each at sites 2 (cedar), 4 (cedar), and 5 (cypress). Mature trees aged around 40 years were primarily targeted on the assumption that they would reach cutting age during the next few decades. Among the nine plots, the aboveground biomass of trees with a diameter at breast height of >10 cm as of 2015 is largest in the site-5 cypress plot (34 kg m^{-2}), followed by sites-2 and -3 cedar plots (22–29 kg m^{-2}), sites-1 and -4 cedar and site-1 cypress plots (16 kg m^{-2}), site-3 oak and pine plots (12–13 kg m^{-2}), and site-1 oak plot (9 kg m^{-2}); the relative biomass increment rate in woods is high in site-1 cypress and site-4 cedar plots (8%–10% yr^{-1}), intermediate in sites-1 to -3 cedar and site-5 cypress plots (2%–5% yr^{-1}), and low in sites-1 and -3 oak and site-3 pine plots (0%–1% yr^{-1}) (Imamura et al., 2017).

Samples were collected every summer (July–September) from 2011 to 2016. As exceptions, the first sampling of the cedars at site 2 was conducted in November 2011, the first and second sampling of the cypresses at site 5 were carried out in February 2012 and January 2013, respectively, and the first sampling of the cypresses and oaks at site 1 was done in summer of 2012. Three trees of differing diameters (small, medium, and large) were felled at each site every year (Imamura et al., 2017). A summary of the sampled tree species, ages, diameters at breast height, and heights at each plot is shown in Table 1. Bark, sapwood, and heartwood samples were collected from the stems around breast height as described by Kuroda et al. (2013). After grinding the samples with a cutting mill (6-mm sieve), the 2011 samples were oven-dried at 105 °C for 48 h and those collected after 2012 were dried at 75 °C for 48 h. The sample weight dried at 75 °C was converted to that at 105 °C by multiplying the ratio derived from the drying experiment: 0.98 for the bark samples and 0.99 for the wood samples (Table S1 in supplementary data). Finally, the bark samples were packed into a 100-mL polystyrene container or a 0.7-L Marinelli pouch, and the wood samples were packed into a 2.0-L Marinelli pouch.

The radioactivity of ^{137}Cs in the samples was determined using gamma-ray spectrometry, and the concentration was calculated on a dry weight (105 °C) basis. The gamma-ray peak of ^{137}Cs (662 keV) was measured using a high-purity Ge detector (GC2020-7500SL-2002CSL, Canberra, Meriden, USA) at the Japan Frozen Foods Inspection Corporation for the 2011 samples and a high-purity Ge detector (GEM20-70, GEM40P4-76, GEM-FX7025P4-ST, or GWL-120-15-LB-AWT, ORTEC, Oak Ridge, USA) at FFPRI for the samples collected from 2012 to 2016. Data regarding the ^{137}Cs concentrations in the barks and whole woods from 2011 to 2015 can be found in the study of Imamura et al. (2017), and those in the cedar heartwoods in 2011 can be found in the study of Kuroda et al. (2013). Measurement accuracy was checked by using the reference standard material IAEA-152 at the Japan Frozen Foods Inspection Corporation and the soil sample 01 of the IAEA-CU-2006-03 world-wide proficiency test on the determination of gamma-emitting radionuclides (IAEA/AL/171) at FFPRI. The ^{137}Cs concentration in the whole wood (C_{WW}) was calculated as the mean of the ^{137}Cs concentrations in the sapwood (C_{SW}) and heartwood (C_{HW}) weighted by the biomass of the sapwood (M_{SW}) and heartwood (M_{HW}) for each individual as follows: $C_{WW} = (C_{SW} \times M_{SW} + C_{HW} \times M_{HW})/(M_{SW} + M_{HW})$. Linear regression analysis was performed to detect any overall trends in the ^{137}Cs concentration in each stem component of each species at each site during the study period.

3. Results

The ^{137}Cs concentrations (decay-corrected to sampling date; the same hereafter) in the bark of cedars at one of four sites (site 1), cypresses at both two sites (sites 1 and 5), oaks at one of two sites (site 3), and pines at site 3 showed decreasing trends during 2011–2016 (2012–2016 for the cypresses), whereas the others did not show any clear trends (Figs. 2 and 3; see Table S2 in supplementary data for the numerical data). These trends in the barks are similar to the trends observed during 2011–2015 (or 2012–2015) reported by Imamura et al. (2017). The mean ^{137}Cs concentrations at the plots where a decreasing trend was seen were two to four times lower in 2016 than those in the first sampling year (2011 or 2012).

The ^{137}Cs concentrations in the sapwood of the cedars showed different trends among the sites, an increasing trend at site 2, a decreasing trend at site 4, and no distinct trends at sites 1 and 3 (Fig. 2). The cypresses showed an increasing trend at site 1 and no distinct trend at site 5, and the oaks showed increasing trends at both sites 1 and 3 (Fig. 3). The pines (investigated at a single site 3) did not show any remarkable temporal variations in the sapwood ^{137}Cs concentrations (Fig. 3). Compared with the first sampling year, the mean ^{137}Cs concentrations at the plots where an increasing trend was seen (the cedars at site 2, cypresses at site 1, and oaks at sites 1 and 3) were two to four times higher in 2016, and

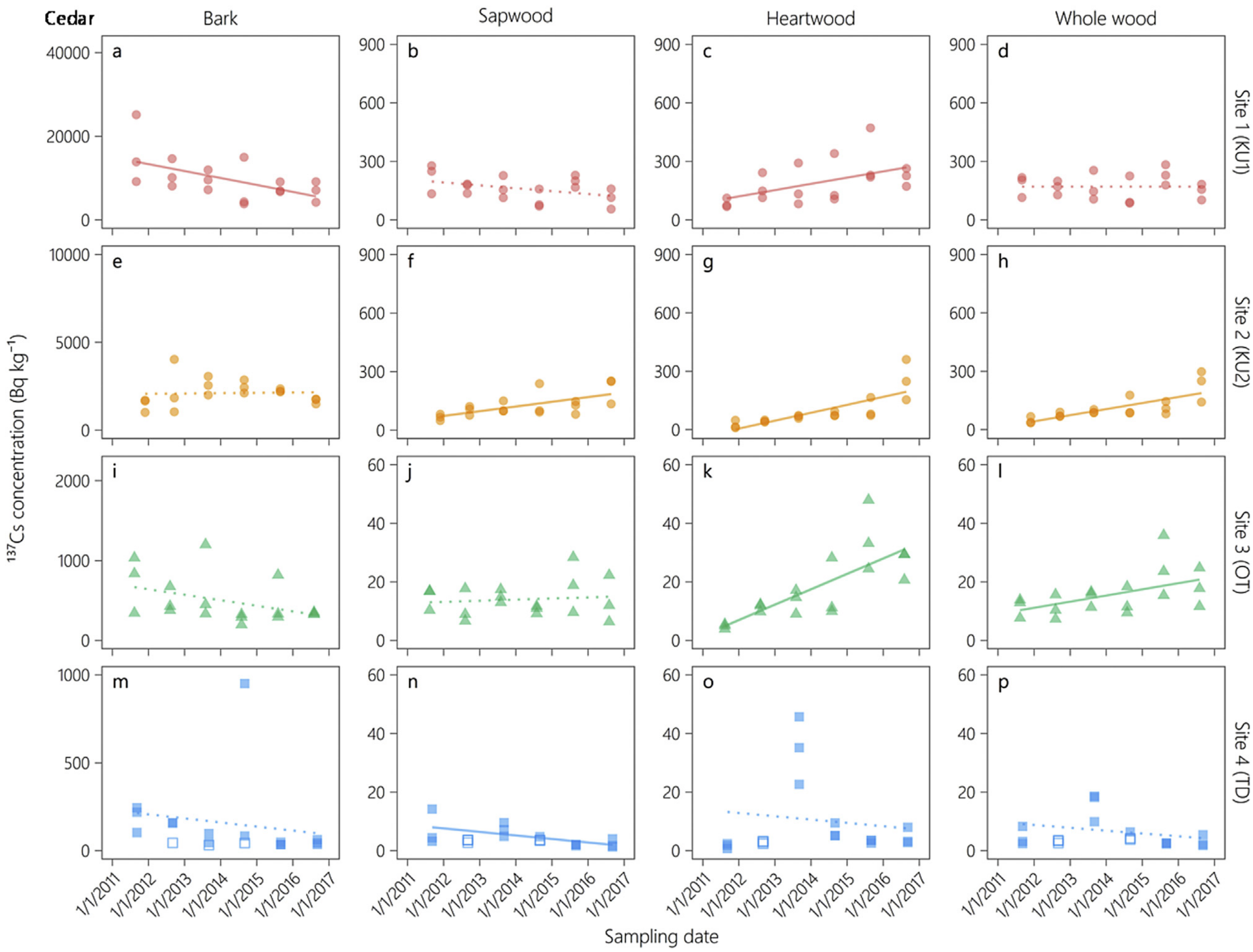

Fig. 2. Temporal variations in ^{137}Cs concentrations (dry weight basis; decay-corrected to sampling date) in the bark, sapwood, heartwood, and whole wood of Japanese cedar (*Cryptomeria japonica*) sampled at sites 1 (a–d; *circle*), 2 (e–h; *circle*), 3 (i–l; *triangle*), and 4 (m–p; *square*). *Open squares* in m–p indicate that ^{137}Cs was not detected (ND) in the samples and show the detection limit values. *Solid lines* indicate that linear regressions were significant ($p < 0.05$), and *broken lines* indicate that they were not significant. The ND data points were excluded from the regressions. Data on ^{137}Cs concentrations in the barks and whole woods from 2011 to 2015 were provided in the study of Imamura et al. (2017) and those for the heartwoods in 2011 were provided in the study of Kuroda et al. (2013).

those at the plots where a decreasing trend was seen (the cedars at site 4) were three times lower. Comparison of the ^{137}Cs concentrations in the 2016 sapwoods between species within the same site (sites 1 and 3) showed that the oaks had the highest values, followed by the cypresses, cedars, and pines; the values in the pines were five to nine times lower than those in the cedars and oaks at site 3.

In the heartwood, the ^{137}Cs concentrations in the cedars at three of four sites (sites 1, 2, and 3), cypresses at one of two sites (site 1), and oaks at one of two sites (site 3) showed increasing trends and were 2–11 times higher in 2016 than those in the first sampling year (Figs. 2 and 3). Regarding the ^{137}Cs concentrations of the others, the cedars at site 4 (even excluding the outliers in 2013), cypresses at site 5, oaks at site 1, and pines at site 3 did not show a clear temporal trend. Although the ^{137}Cs concentrations in the heartwood of the cedars at site 4 in 2013 were 5–23 times higher than those in other years, those in the bark and sapwood were at the same level as those in the other years (Fig. 2m–o).

The ^{137}Cs concentrations in the whole woods showed increasing trends in the cedars at two of four sites (sites 2 and 3), cypresses at one of two sites (site 1), and oaks at both sites (sites 1 and 3), and were two to five times higher in 2016 than those in the first sampling year (Figs. 2 and 3). On the other hand, the ^{137}Cs concentrations showed a decreasing trend in the pines at site 3 and were two times lower in 2016 than those in 2011 (Fig. 3). The other samples did not show a distinct temporal trend. These trends in the whole woods are consistent with the trends observed during 2011–2015 (or 2012–2015) reported by Imamura et al. (2017).

Even after excluding the radioactive decay effect of ^{137}Cs (−2.3% yr^{-1}) by decay-correcting the ^{137}Cs concentrations to the same date (March 11, 2011; Figs. S1 and S2 in supplementary data), the temporal trends were the same as described above (Figs. 2 and 3); the only exception was those in the whole wood of pines at site 3, which showed a decreasing trend when decay-corrected to each sampling date (Fig. 3) but no distinct trend when the radioactive decay effect was excluded (Fig. S2).

The ratio of the ^{137}Cs concentration (dry weight basis) in the heartwood to that in the sapwood (hereafter, HW/SW concentration ratio) showed increasing trends in the cedars at three of four sites (sites 1, 2, and 3) but at all four sites if the outliers in 2013 at site 4 are excluded; no distinct trends were observed in the other species (Fig. 4). The mean ratios in 2011 were 0.2–0.4 for all species, and those in 2016 were 1.2–2.3 in cedar and 0.3–0.6 in the other three species. When the HW/SW concentration ratios were derived on a fresh weight basis, the ratios were higher than those derived on a dry weight basis in the three species except the oak (Fig. 5). In

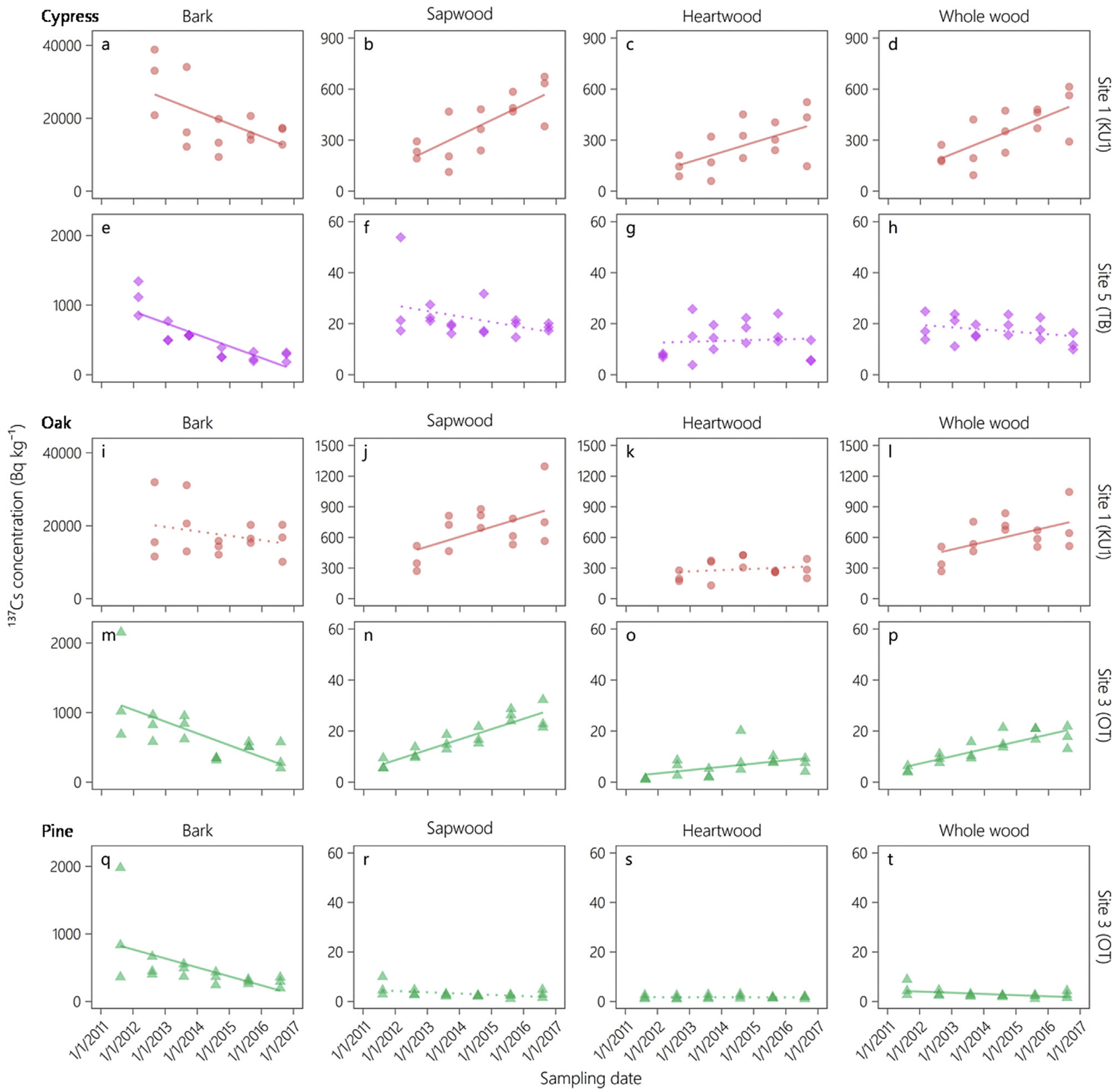

Fig. 3. Temporal variations in ^{137}Cs concentrations (dry weight basis; decay-corrected to sampling date) in the bark, sapwood, heartwood, and whole wood of Japanese cypress (*Chamaecyparis obtusa*; a–h), oak (*Quercus serrata*; i–p) and pine (*Pinus densiflora*; q–t) sampled at sites 1 (*circle*), 3 (*triangle*), or 5 (*diamond*). *Solid lines* indicate that linear regressions were significant ($p < 0.05$), and *broken lines* indicate that they were not significant. Data on ^{137}Cs concentrations in the barks and whole woods from 2011 to 2015 were provided in the study of Imamura et al. (2017).

the oak, the ratios based on the dry and fresh weights were almost the same due to similar moisture content in the sapwood and heartwood. In the cypresses and pines, on a fresh weight basis, most of the ratios in the last 3 years exceeded 1, meaning a higher ^{137}Cs concentration in the heartwood than that in the sapwood.

4. Discussion

The decreasing trends in ^{137}Cs concentrations in the bark at five of nine study plots indicate that ^{137}Cs has been decontaminated due to rainwater leaching and debarking. Those leaching and turnover rates in the bark, however, would be slower than those in the needles and branches (Imamura et al., 2017). The indistinct trends at the other four plots are partly due to large variations in ^{137}Cs concentration among individuals. Although some individuals showed much higher ^{137}Cs concentrations in the bark than those in the others, e.g., a cedar at site 4 in 2014 (Fig. 2m) and an oak and a pine at site 3 in 2011 (Fig. 3m, q), the concentrations in the sapwood and heartwood of these individuals were not high and at the same levels as others. These facts imply that the bark of some individuals has experienced inhomogeneous ^{137}Cs contamination that may be immobile, making the decreasing trend indistinct. The site and species dependencies of temporal trends in the bark ^{137}Cs concentrations are difficult to discuss, because the trends were not consistent between the sites and the species. However, species dependency is expected to become important in the future because

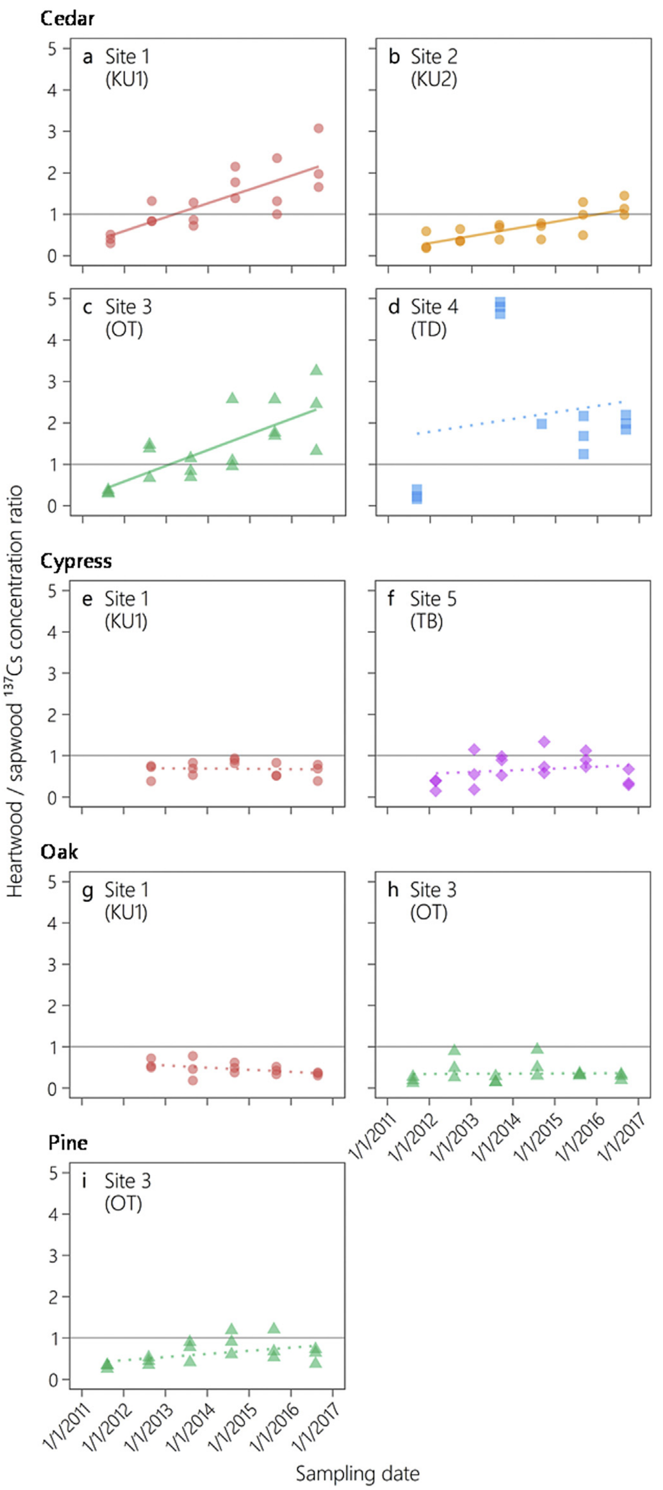

Fig. 4. Temporal variations in heartwood/sapwood ^{137}Cs concentration ratio (dry weight basis) in Japanese cedar (*Cryptomeria japonica*; a–d), cypress (*Chamaecyparis obtusa*; e, f), oak (*Quercus serrata*; g, h), and pine (*Pinus densiflora*; i) sampled at sites 1 (*circle*), 2 (*circle*), 3 (*triangle*), 4 (*square*), or 5 (*diamond*). *Solid lines* indicate that linear regressions were significant ($p < 0.05$), and *broken lines* indicate that they were not significant. Data on ^{137}Cs concentrations in the cedar heartwoods of 2011 were provided in the study of Kuroda et al. (2013).

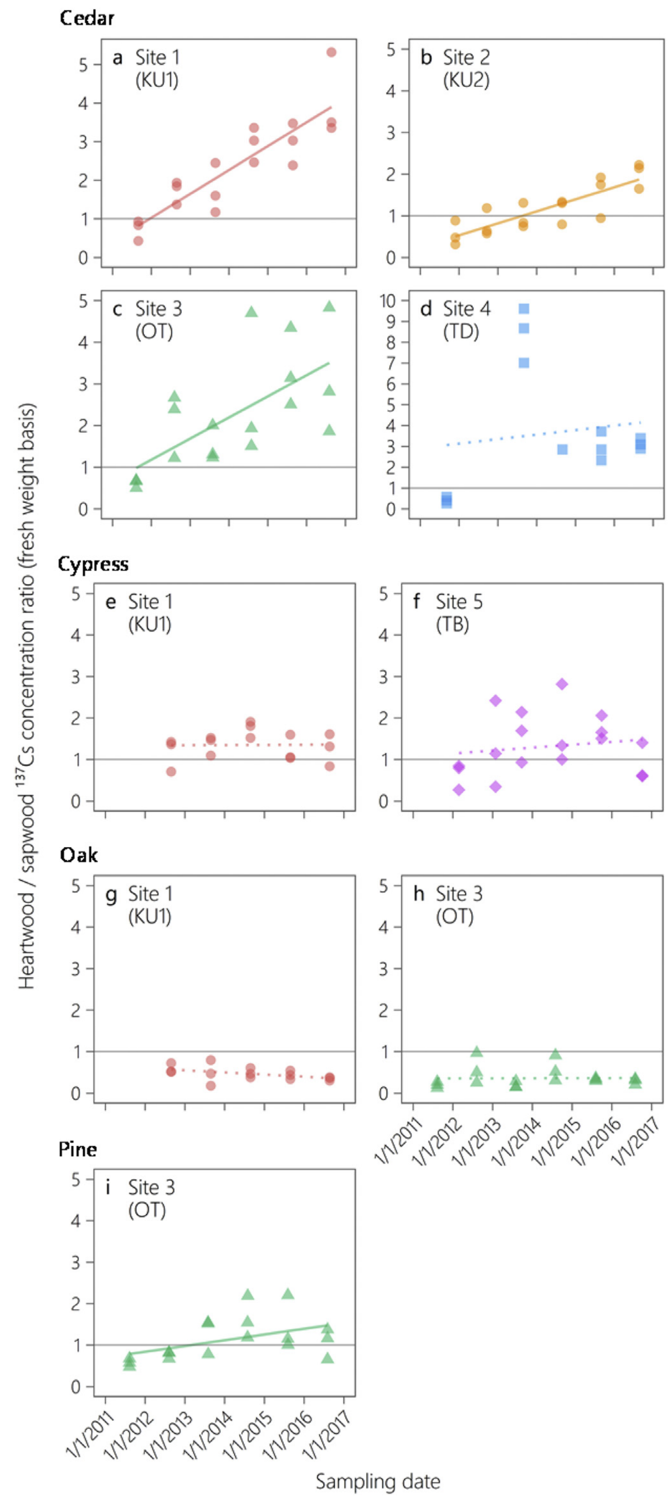

Fig. 5. Temporal variations in the heartwood/sapwood ^{137}Cs concentration ratio (fresh weight basis) in Japanese cedar (*Cryptomeria japonica*; a–d), cypress (*Chamaecyparis obtusa*; e, f), oak (*Quercus serrata*; g, h), and pine (*Pinus densiflora*; i) sampled at sites 1 (*circle*), 2 (*circle*), 3 (*triangle*), 4 (*square*), or 5 (*diamond*). *Solid lines* indicate that linear regressions were significant ($p < 0.05$), and *broken lines* indicate that they were not significant. Data on ^{137}Cs concentrations in the cedar heartwoods of 2011 were provided in the study of Kuroda et al. (2013).

the turnover time of bark may differ among species. As such information is scarce, continuous monitoring together with biological research focusing on bark formation and desquamation is necessary for long-term prospects of ^{137}Cs bark contamination.

Increasing trends in the sapwoods and whole woods suggest that the amount of ^{137}Cs uptake by roots exceeded that of discharge via defoliation and death of branches and roots and also imply the possibility of continuous direct uptake from the bark. The occurrence of ^{137}Cs direct uptake from the bark has been suggested for cherry trees (Tagami et al., 2012) and demonstrated experimentally in cedars (Wang et al., 2016); however, it is unclear whether it occurs over a number of years. Although it is impossible to determine the contributions from root uptake and bark absorption in the present study, we speculate that the contribution of the latter may be small because increasing trends in the sapwood and whole wood were found in the plots where the bark did not show a decreasing trend (Figs. 2e–h and 3i–l). In the trees that did not show clear trends in their ^{137}Cs whole wood concentrations decay-corrected to the same date (cedars at sites 1 and 4, cypresses at site 5, and pines at site 3; see Figs. S1 and S2), the absorption and discharge of ^{137}Cs would be in equilibrium, suggesting that ^{137}Cs dynamics in some locations reached a "steady state" phase in the 5 years after the FDNPP accident, similar to that after the CNPP accident (Calmon et al., 2009). The decreasing trend in sapwood, found only in the cedars at site 4, would be due to ^{137}Cs transfer from sapwood to heartwood but not due to ^{137}Cs discharge from the wood, because there was no clear decreasing trend in the whole wood and the HW/SW concentration ratios increased from 2011 to 2016.

Several studies (e.g., Katayama et al., 1986; Chigira et al., 1988; Kohno et al., 1988; Kudo et al., 1993; Kagawa et al., 2002) have reported that cedar shows higher K and/or Cs concentrations in its heartwood than those in its sapwood (categorized as type 1 by Okada et al., 1993a). Our results are consistent with such reports, and furthermore, the increasing trends in the HW/SW concentration ratios suggest that ^{137}Cs transfer from sapwood to heartwood has not yet come to equilibrium in cedars. Interestingly, stable isotope analysis of the same wood samples (cedars collected in 2014) showed that HW/SW concentration ratios (dry weight basis) for ^{39}K, ^{85}Rb, and ^{133}Cs were 2.3–6.1, 2.0–5.4, and 1.0–2.7, respectively (Nagakura et al., 2017). This fact implies that Cs transfer from the sapwood to heartwood is less active than that of K and Rb. Although the HW/SW concentration ratio of ^{137}Cs in the cedar showed a significant increasing trend in the 5 years after the accident, the ratio in 2016 is similar to that of ^{133}Cs. Thus, we infer that ^{137}Cs distribution between the sapwood and heartwood of cedar at our study sites almost reached an equilibrium as of 2016.

The reason why the ^{137}Cs concentrations in the site-4 cedar heartwoods were exceptionally high in 2013 is unclear. As the concentrations in the bark were at the same level as those in the other years, this may not be caused by differences in initial deposition but could be caused by an internal (physiological) factor.

In the other three species, the low HW/SW concentration ratios (dry weight basis), almost below 1, suggest that ^{137}Cs transfer from sapwood to heartwood is passive. Although ^{137}Cs concentration increased in the heartwood of the cypresses at site 1 and oaks at site 3, it is reasonable to consider this as a consequence of increasing ^{137}Cs concentrations in the sapwood, because the HW/SW concentration ratios remained constant around 0.5. Such low HW/SW concentration ratios in the oaks were consistent with a previous study that reported oaks with a higher K concentration in the sapwood than that in the heartwood (categorized as type 2; Okada et al., 1993b). With regard to the pine, Okada et al. (1993a) categorized it as type 3, with a concentration peak at the sapwood–heartwood boundary. As we separated the wood into two components (sapwood and heartwood), it is impossible to judge whether there was a peak at the boundary or not; however, similar ^{137}Cs concentrations in the sapwood and heartwood after 2013 (Fig. 4i) imply that the distribution is possibly type 3. On the other hand, the radial distribution of K concentration in the cypress has been categorized as type 1, the same as that in the cedar (Okada et al., 1993a), and, in fact, a higher ^{137}Cs concentration has been found in heartwood than that in sapwood in a study on global fallout contamination (Kohno et al., 1988). Nevertheless, in the present study, the HW/SW concentration ratios (dry weight basis) in the cypresses were around 1 throughout the study period and did not appear to increase thereafter. Further studies are needed to determine whether the HW/SW concentration ratios in the cypresses increase in the future.

The transfer mechanism of alkali metals from sapwood to heartwood is not completely understood for each species (Sokołowska, 2013; Spicer, 2014). In the cedar, a tracer experiment indicated that Rb is selectively transported to the heartwood via ray parenchyma (Okada et al., 2011). In addition, downward diffusion from upper to lower heartwood was suggested after an investigation of vertical ^{137}Cs distribution in cedars from 2011 to 2013 (Ogawa et al., 2016). In the other three species, although the low HW/SW concentration ratios on a dry weight basis suggest that the transfer from the sapwood to heartwood is passive, the ratios on a fresh weight basis were around 2 in some of the cypresses and pines. As a ratio far exceeding 1 cannot be explained by diffusion alone, it is reasonable to consider that radial transport via ray parenchyma more or less contributes to ^{137}Cs transfer from the sapwood to heartwood in cypress and pine. For Scots pine (*P. sylvestris*) and silver birch (*B. pendula*), Soukhova et al. (2003) explained the differences in ^{137}Cs radial distributions in the stem woods between species by the difference in ray composition, in particular the presence or absence of ray tracheids. In the present study, as only the pines have ray tracheids, the difference in ray composition cannot satisfactorily explain the different ^{137}Cs radial distributions among the species. Consequently, understanding the contribution of active transport via ray parenchyma is important when modeling ^{137}Cs transfer in the stem woods and to the prospects of each species for wood use.

5. Conclusions

Temporal trends in ^{137}Cs concentrations in bark and whole wood samples collected from 2011 to 2016 were consistent with trends in ^{137}Cs concentrations in those samples collected from 2011 to 2015 (Imamura et al., 2017) and did not show clear site or species dependencies. On the other hand, ^{137}Cs distributions within stem wood (between sapwood and heartwood) differed among species as follows: cedars had a higher ^{137}Cs concentration (fresh weight basis) in their heartwood, cypresses and pines had similar concentrations in their sapwood and heartwood, and oaks showed a lower concentration in their heartwood. As of 2016, the ratio of the ^{137}Cs concentration in heartwood to that in sapwood was in equilibrium in cypress, oak, and pine; however, the ratio in cedar was still changing. Thus, prediction of ^{137}Cs concentrations in stem wood should be carried out carefully as it still requires longer-term investigations at multiple sites with a larger sample size and an understanding of the species-specific ^{137}Cs transfer mechanism.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.jenvrad.2017.09.008.

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