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# Downward migration of $^{137}$Cs promotes self-cleaning of forest ecosystem by reducing root uptake of Japanese cedar in Fukushima

Junko Takahashi [a,*], Satoshi Iguchi [b], Takuya Sasaki [c], Yuichi Onda [a]

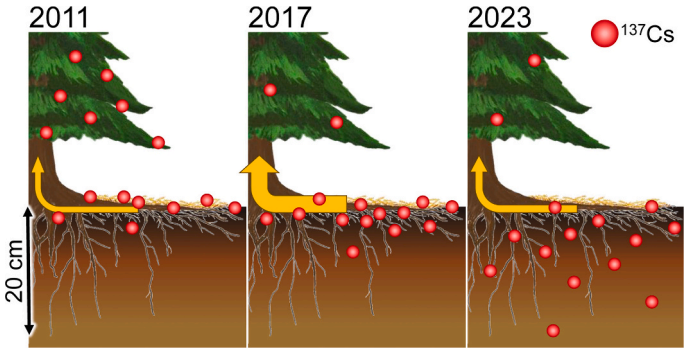
[a] Center for Research in Radiation, Isotopes, and Earth System Science, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8572, Japan
[b] Master's Program in Geosciences, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8572, Japan
[c] College of Geoscience, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8572, Japan

## HIGHLIGHTS

- $^{137}$Cs uptake amount by root determines long-term contamination in forest ecosystem.
- There are few time series data of $^{137}$Cs in tree roots after the nuclear accident.
- We monitored the depth distribution of $^{137}$Cs in cedar tree roots from 2011 to 2023.
- The inventory of $^{137}$Cs in roots decreased after 2020 due to downward migration.
- Downward migration of $^{137}$Cs can be regarded as a "self-cleaning" of the forest.

## GRAPHICAL ABSTRACT

**Temporal change in $^{137}$Cs absorption by cedar tree roots.**

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## ABSTRACT

Approximately 70 % of the area highly $^{137}$Cs-contaminated by the Fukushima Daiichi Nuclear Power Plant accident is forested. Decontamination works in most of these forests have not progressed, and the forestry industry remains stagnant. Although the long-term dynamics of $^{137}$Cs in the forest ecosystem will be controlled by the amount of $^{137}$Cs absorbed by roots in the future, temporal changes in $^{137}$Cs of tree roots have rarely been reported. In the present study, we monitored the depth distribution of $^{137}$Cs in the soil and absorptive very fine (VF) roots of 0.5 mm or less in a Japanese cedar forest from 2011 to 2023. As a result, the $^{137}$Cs inventory in the mineral soil increased over time due to the migration from the forest canopy and litter layers, whereas that in the VF roots tended to decrease since 2020, although there was a large variation. Temporal decrease in the exchangeable $^{137}$Cs fraction with fixation and temporal increase in VF root biomass with their growth were not clearly observed, the $^{137}$Cs concentration in the VF roots at 0–2 cm decreased with the decrease in $^{137}$Cs concentration in the litter layers. Although the $^{137}$Cs concentration in the VF roots below 2 cm tended to increase with increasing $^{137}$Cs concentration in the soil at the same depth, the downward migration of $^{137}$Cs within the soil can reduce the amount of $^{137}$Cs absorbed by roots because the VF root biomass decreases exponentially with depth. In other words, $^{137}$Cs can be removed from the long-term active cycles of forest ecosystems as they migrate deeper into the soil. This natural migration process can be regarded as a "self-cleaning" of the forest ecosystem,

* Corresponding author.
*E-mail addresses:* takahashi.junko.ka@u.tsukuba.ac.jp (J. Takahashi), onda@geoenv.tsukuba.ac.jp (Y. Onda).

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the green and sustainable remediation using such self-cleaning should be actively adopted for the future forest management.

## 1. Introduction

Numerous studies after the Chernobyl Nuclear Power Plant (ChNPP) accident have revealed that radiocesium ($^{137}$Cs) deposited on forests was intercepted by the canopy, then migrated to the litter layer and eventually to the soil layer, where some of it has been absorbed by roots and circulated through the forest ecosystem for a long time (International Atomic Energy Agency, 2002). Meanwhile, a large proportion of the $^{137}$Cs deposited on forests after the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident in 2011 has already migrated to the forest floor (litter and soil layers) (93–99 % as of 2015; Imamura et al., 2017a, 89–97 % as of 2018; Onda et al., 2020). Although the migration of $^{137}$Cs from the litter layer to the mineral soil surface was significantly faster in Fukushima than in Chernobyl (Imamura et al., 2020), >80 % of the deposited $^{137}$Cs has been distributed in the 0–5 cm of soil surface (Manaka et al., 2022; Takahashi et al., 2019; Yoschenko et al., 2022b). The annual amount of $^{137}$Cs runoff from forest ecosystems through streams and rivers due to leaching and erosion is only 0.02–0.3 % of the total deposition amount (Iwagami et al., 2017). It is an important fact that the extremely high Cs-adsorption ability of soils (Sawhney, 1972) has prevented the spread of $^{137}$Cs contamination to the groundwater and the food chain.

However, the persistence of $^{137}$Cs in the soil surface may prolong forest contamination and become a potential long-term source of $^{137}$Cs to downstream areas. In fact, the air dose rates in urban areas are declining more rapidly than those in forests, which were similar to the physical attenuation rate of radionuclides (Saito et al., 2019). Of the 489,950 ha forest area in 35 municipalities in Fukushima Prefecture that were decontaminated to reduce air dose rates, only 10,374 ha (approximately 2 %) were decontaminated by clear-cutting and/or removal of litter layers (Kawasaki, 2021). Evrard et al. (2019) mentioned that further forest decontamination is impractical because the total estimated cost of decontaminating all forested areas exposed to radiation dose rates >1 mSv yr$^{-1}$ would exceed 16 trillion JPY (approximately 120 billion USD). The forestry industry in the difficult-to-return and former-evacuation zones remains stagnant. The resumption of proper forest management is among the biggest challenges that must be overcome.

The recently developed concept of “green and sustainable remediation” considers the socio-economic aspects and secondary environmental impacts of remediation from pollutants (Hou and Al-Tabbaa, 2014; Yasutaka et al., 2016). One approach to sustainable remediation is applying nature-based solutions (Keesstra et al., 2018), wherein restoration and rehabilitation strategies are based on natural processes and cycles rather than artificial maintenance, which requires huge amounts of energy and money. Since the ChNPP accident, the reduction of $^{137}$Cs concentrations by natural decay, dilution with precipitation and water addition from inflowing rivers, vertical turbulent diffusion and others have been evaluated as “self-cleaning” processes in rivers and lakes (Santschi et al., 1990) and the sea (Styro et al., 2001). The effects of these “self-cleaning” processes in the terrestrial environment are less well known than in the aquatic environment (Onda et al., 2020). Although some studies on $^{137}$Cs absorption control by potassium fertilization have been conducted in plantation forests (Kobayashi et al., 2019; Komatsu et al., 2017), the direct removal of radioactive materials is still the mainstream of forest decontamination after the FDNPP accident. Little attention has been paid to the effectiveness of self-cleaning methods based on natural processes and cycles in forests.

A deep understanding of the functions and processes of nature is required to successfully implement nature-based solutions (Keesstra et al., 2018). At the late stage, >10 years since the FDNPP accident, the effects of $^{137}$Cs that directly deposited on the canopy and tree bodies are negligible, and absorption of $^{137}$Cs by roots is the only source of uptake into the tree (Ohashi et al., 2022). In other words, the amount of $^{137}$Cs uptake by roots will control the long-term dynamics in the forest ecosystem in the future, therefore the importance of estimating the magnitude of root uptake should be more emphasized (Yoschenko et al., 2022a). However, studies on $^{137}$Cs in roots are limited due to the difficulty of collecting, pre-treating and measuring them. Especially, as the $^{137}$Cs concentration decreases exponentially through deeper forest soil layers, plant roots must be collected quantitatively from each soil depth in increments of several centimeters. After the ChNPP accident, Fesenko et al. (2001) pointed out that it is important to consider the depth distribution of tree root biomass to estimate the $^{137}$Cs concentration in trees; however, the depth distribution of $^{137}$Cs concentration in tree roots was not reported. Although the $^{137}$Cs inventory of tree roots in an entire forest by excavating tree roots (Coppin et al., 2016) and the depth distribution of $^{137}$Cs in tree roots at 5-cm intervals (Sakashita et al., 2020) have been reported after the FDNPP accident, none of these studies have investigated temporal changes in $^{137}$Cs concentration in tree roots.

This is the first study to clarify temporal changes in the depth distribution of $^{137}$Cs in cedar tree roots from 2011 to 2023. We collected very fine (VF) tree roots (≤0.5 mm) of Japanese cedars using soil samples from the previously reported monitoring site in the former evacuation zone of Fukushima (Takahashi et al., 2019). Recently, a functional classification of fine roots smaller than 2 mm into absorptive and transport fine roots has been proposed (McCormack et al., 2015). Absorptive fine roots possess nutrient and water absorption abilities that are highly sensitive to various soil conditions and are current-year roots that are less affected by translocation and accumulation of $^{137}$Cs within a tree. The absorptive fine root of Japanese cedar has been reported to be approximately 0.5 mm or less (Wada et al., 2019). Moreover, we clarified the relationship between the $^{137}$Cs concentrations in litter and soil layers and VF roots at various depths to determine the factors influencing $^{137}$Cs absorption by tree roots. Furthermore, we discussed the potential self-cleaning effects of forest ecosystems as a sustainable remediation strategy based on natural processes and cycles.

## 2. Materials and methods

### 2.1. Site description

The study site is a young cedar forest (37°35′08″N, 140°41′26″E) located in Yamakiya District, Kawamata Town, Fukushima Prefecture, approximately 35 km northwest of the FDNPP (Fig. 1a). The estimated deposition density of $^{137}$Cs by airborne monitoring is 440 kBq m$^{-2}$ in this forest (Kato et al., 2019a). Comprehensive monitoring of the depth distribution of $^{137}$Cs in soils (Takahashi et al., 2019), soil infiltration water (Takahashi et al., 2022), throughfall, litterfall, stemflow, and leaves (Kato et al., 2019b) has been conducted at this site.

As of 2011, the forest was a 15-year-old plantation with a stand density of 2600 trees ha$^{-1}$, and the trees were planted approximately 2 m apart. The soil classification is non-allophanic (aluandic) andosol derived from volcanic ash. However, andic properties (IUSS Working Group WRB, 2022) are not observed on the surface soil where $^{137}$Cs accumulated although it has a high organic matter content, low bulk density, and high saturated hydraulic conductivity. The litter layer was approximately 5–6 cm thick without an Oa layer. The forest has not been managed since the FDNPP accident and is dark with little understory vegetation.

### 2.2. Soil sampling

Two sampling plots were established on a relatively flat surface in the forest (Fig. 1a, b). Litter and soil samples were collected using a scraper plate (Campbell et al., 1988) with a 30 cm × 15 cm frame. Since 2016, the second plot has been moved approximately 9 m away from the first plot, and annual sampling has continued in summer. Soil samples were collected up to a depth of 5 cm at 5-mm intervals, 5–10 cm at 1-cm intervals, and 10–20 cm at 5-cm intervals. Although sampling was performed only once during each survey, the sampling area (450 $cm^2$) corresponds to 23 liner cores with a diameter of 5 cm, which is sufficiently large to ensure representativeness (Khomutinin et al., 2004) and is able to estimate the mean $^{137}$Cs inventory with an uncertainty of 16 % in the cedar stand (Kato et al., 2022). This corresponds to the preferred sampling area with a diameter > 10 cm for estimating fine root biomass, as reviewed by Addo-Danso et al. (2016). However, because root analysis was not originally planned, the distance from the tree trunk to the sampling point varied, with the closest distance to the trunk (approximately 35 cm) recorded in 2020, followed by that in 2023, 2012, 2017, and 2015 (Table 1). The $^{137}$Cs depth distribution data from 2011 to 2023, the details of soil properties and sampling methods have been previously reported (Takahashi et al., 2024, 2019).

### 2.3. Pretreatment of root samples

Root samples (Fig. 1c) were collected by further separating only the roots with tweezers from the fresh organic matter, plant roots, and gravels which were stored after the collected soil samples were passed through a 2 mm sieve. Samples only containing skin and lacking an obvious internal root tissue were not included. The roots measured were absorptive VF roots of 0.5 mm or less (McCormack et al., 2015; Wada et al., 2019) of the current year's growth of five years, 2012, 2015, 2017, 2020 and 2023. There was little understory vegetation at the study site, so all root samples could be considered cedar roots.

The weight of VF roots collected from the soil layers at 5-mm or 1-cm intervals was too small to be measured for $^{137}$Cs; therefore, four or two layers of roots were mixed at 2-cm intervals. The mixed root samples were placed in a beaker filled with distilled water and washed repeatedly with an ultrasonic homogenizer (Sonifier 250; Branson Ultrasonics, Danbury, CT, USA) until no soil particles settled at the bottom of the beaker. The $^{137}$Cs may have leached from the root cells during ultrasonic washing. For example, shaking experiments have shown that approximately 30 % of the $^{137}$Cs in cedar litters is dissolved in 1 day (Sakai et al., 2015). However, the $^{137}$Cs concentration in the distilled water used for washing was not measured in this study because the $^{137}$Cs concentrations were extremely low and it was difficult to determine whether $^{137}$Cs leached from the roots or from soil particles attached to the root surface. The washed roots were dried in an oven at 70 °C for at least 24 h, then powdered using a bead-type grinder.

The root samples collected using a scraper in 2011 were not stored; however, the fresh organic matter samples, including the roots that were separated from the soil samples collected for the soil profile survey (Takahashi et al., 2019), were stored. Therefore, VF root samples from the A1 horizon (0–14 cm depth) in 2011 were collected and pretreated in the same procedures.

### 2.4. Measurement of $^{137}$Cs concentration

The $^{137}$Cs concentrations in the samples were determined by measuring gamma rays at 662 keV using a high-purity n-type germanium coaxial gamma-ray detector (EGC25-195-R; Canberra-Eurisys, UK) coupled to an amplifier (PSC822; Canberra, UK) and a multichannel

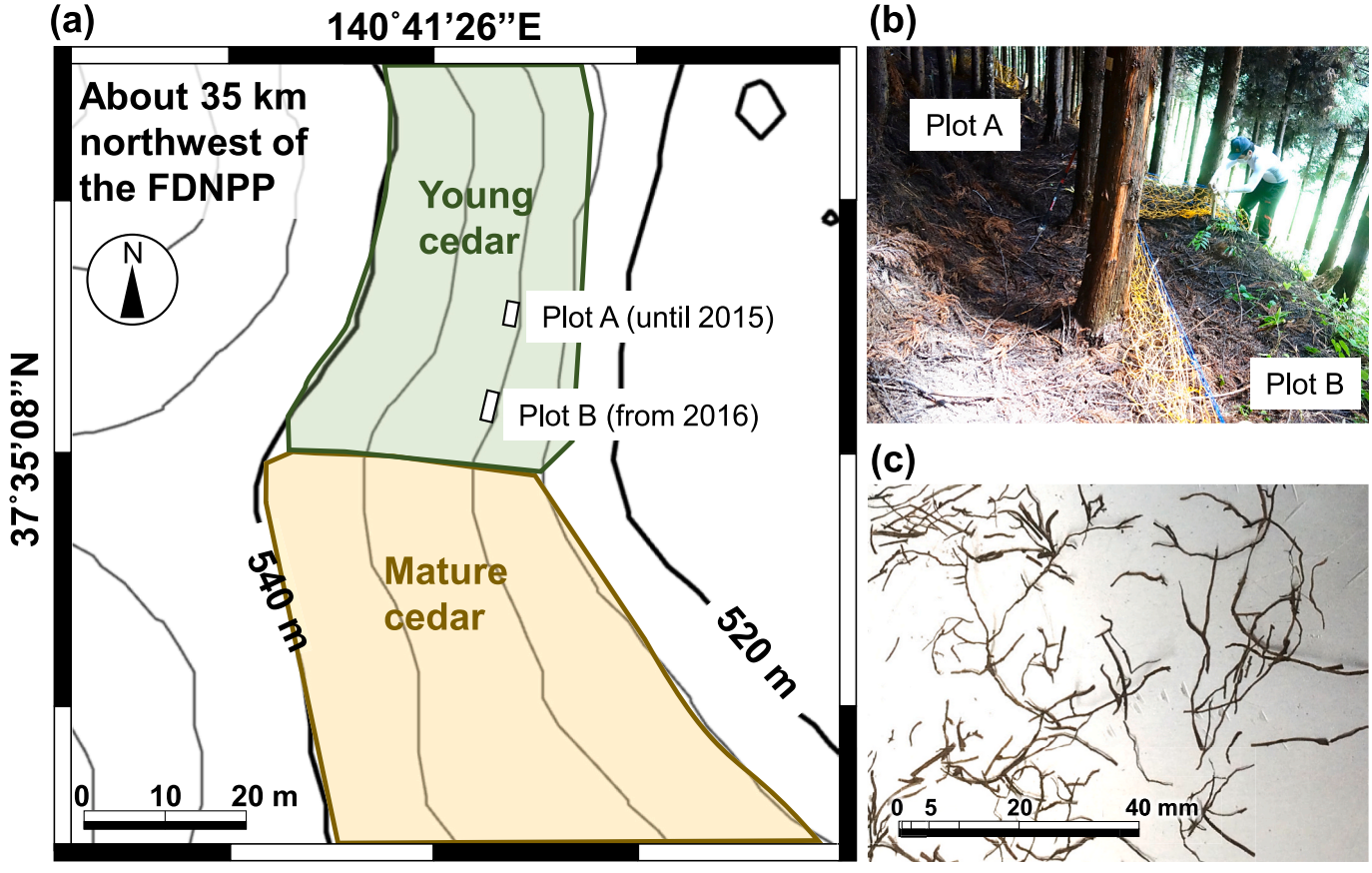


**Fig. 1.** Location of the study site (a), sampling plots (b), and the collected VF roots (c).

**Table 1**
Depth distribution of the VF root biomass of cedar trees (g $m^{-2}$) for each survey.

| Sampling date | | Aug. 28, 2012 | July 26, 2015 | July 2, 2017 | July 2, 2020 | Aug. 9, 2023 | Mean g $m^{-2}$ | SD g $m^{-2}$ | CV % | Distribution ratio % |
|---|---|---|---|---|---|---|---|---|---|---|
| Year since the accident | | 1.5 | 4.4 | 6.3 | 9.3 | 12.4 | | | | |
| Distance from tree trunk (cm) | | 85 | 105 | 90 | 35 | 45 | | | | |
| Mean basal area ($cm^2$)[a] | | 260 | 300 | 330 | 380 | 430 | | | | |
| Depth (cm) | 0–2 | 5.4 | 4.8 | 4.3 | 6.2 | 6.1 | 5.4 | 0.8 | 15.6 | 28.4 |
| | 2–4 | 6.0 | 1.3 | 4.9 | 5.2 | 5.4 | 4.6 | 1.9 | 40.7 | 22.2 |
| | 4–6 | 0.7 | 1.1 | 4.1 | 5.8 | 2.4 | 2.8 | 2.1 | 75.9 | 11.1 |
| | 6–8 | 0.7 | 1.1 | 2.3 | 6.3 | 2.0 | 2.5 | 2.2 | 91.0 | 9.5 |
| | 8–10 | 0.2 | 0.9 | 1.9 | 4.8 | 2.2 | 2.0 | 1.7 | 86.4 | 7.7 |
| | 10–15 | 0.3 | 0.7 | 3.3 | 7.5 | 2.0 | 2.8 | 2.9 | 105.0 | 9.6 |
| | 15–20 | 0.4 | 0.7 | 4.2 | 7.4 | 3.2 | 3.2 | 2.8 | 89.6 | 11.5 |
| Total (0–4 cm) | | 11.4 | 6.1 | 9.2 | 11.4 | 11.6 | 11.6 | 2.3 | 23.6 | 50.6 |
| Total (0–10 cm) | | 13.0 | 9.3 | 17.4 | 28.4 | 18.0 | 18.0 | 7.2 | 41.6 | 78.9 |
| Total (0–20 cm) | | 13.8 | 10.7 | 24.9 | 43.3 | 23.2 | 23.2 | 12.8 | 55.1 | 100.0 |

[a] Estimated by measuring the diameter at breast height (DBH) of 18 trees around the sampling plots.

analyzer (DSA1000; Canberra, UK). Background levels were corrected by measuring blank samples for 48 h every 1–2 months. The measurement was stopped generally when the counting error was <10 %, however, <20 % was applied if it took >7 days. All radioactivity concentrations were decay-corrected to the sampling date. For soil samples collected at 5-mm or 1-cm intervals (0–10 cm), the $^{137}$Cs concentrations were measured separately and calculated at 2-cm intervals by the weighted average over the sample weight.

### 2.5. Measurement of $^{133}$Cs concentration

The $^{133}$Cs concentrations of the root, litter, and soil samples were determined by allowing the samples to totally decompose. The root and litter samples (0.1 g) were placed in clean beakers and decomposed on a hot plate by concentrated nitric acid and hydrogen peroxide. For soil samples at depths of 0–10 cm, mixed soil samples with two or four soil layers were prepared based on sampling weight. Soil samples (0.5 g) were weighed into Teflon beakers on a hot plate; perchloric acid and concentrated nitric acid were added to decompose the organic matter, and hydrofluoric acid was added to facilitate total decomposition. The samples were dried and dissolved in 2 % nitric acid, which was diluted accordingly. The $^{133}$Cs concentration was determined using an inductively coupled plasma–mass spectrometry (Agilent 7700; Agilent Technologies, Santa Clara, CA, USA). The decomposition was performed twice, and each sample was measured once; the average value was obtained. In addition, Al concentration in root samples was measured using an atomic absorption spectrometry (ZA3300; Hitachi High-Tech Corporation, Tokyo, Japan) to evaluate the contamination of soil particles.

### 2.6. Extraction of exchangeable Cs in soil samples

The exchangeable Cs fraction in 0–2 cm and 10–15 cm soil was extracted using the same mixed soil samples as in Section 2.5. According to Bunzl et al. (1997), 1 h shaking was conducted using a 1 M ammonium acetate solution with a soil:solution ratio of 1:10 at room temperature. After centrifugation, the suspension was filtered by 0.20 μm membrane filter. Concentrations of $^{137}$Cs and $^{133}$Cs were determined by the same methods described in Sections 2.4 and 2.5.

### 2.7. Statistical analysis

Correlations between $^{137}$Cs and $^{133}$Cs concentrations in the root, litter, and soil samples at each depth were evaluated using Pearson's correlation coefficient, and significant differences were determined. Temporal changes in the $^{137}$Cs inventory ratios of the root/litter and soil were determined by performing linear regression using the least-squares method. All statistical analyses were performed using IBM SPSS Statistics 27.

## 3. Results and discussion

### 3.1. Variation of VF root biomass and its factors

Table 1 shows the depth distribution of the VF root biomass, distance from the nearest tree trunk and mean basal area of cedar trees. Japanese cedar is a deep-rooted species (Karizumi, 2010) and is often planted on lower slopes and valleys where the soil is moist and fertile (Mashita, 1960). However, the VF root biomass decreased with depth, accounting for approximately 28 % at 2-cm depth to 51 % at 4-cm depth in the five-year average of observations. The total biomass of VF roots in 0–20 cm ranged from 10.7 g $m^{-2}$ to 43.3 g $m^{-2}$ with very large variations (the coefficient of variation (CV:) 55 %).

One factor contributing to this variation is seasonal variation. The highest root biomass can be observed from July to August because fine roots actively die and abscise (Konôpka et al., 2006). However, all of our surveys were conducted in July and August. To discuss the other two factors, distance from the nearest cedar trunk and tree growth, a comparison was made with a dataset by Karizumi (2015). Fig. 2a shows the relationship between the rate of decrease in root biomass and the distance from the nearest tree trunk. The distance from the trunk in this study represents the approximate distance from the center of the sampling point to the trunk, and the rates of decrease were calculated by the VF root biomass in 2020 closest to the trunk (35 cm) as 1. Although these data from Karizumi (2015) were for fine (F) roots <2 mm, they obtained similar results for larger roots with an exponential decrease according to distance from the trunk. Fig. 2b shows the relationship between VF and F root biomass in surface soil (0–15 cm) and basal area at breast height, indicating the changes in the root biomass with tree growth. According to Karizumi (2015), the density of F roots in the surface soil layer reaches a maximum when the cedar trees are relatively young with a basal area of 100–200 $cm^2$, and then gradually decreases and remains almost flat because the root density in the surface layer is high enough in dense forest to limit growth due to mutual interference and competition. On the other hand, fine roots in deeper soil layers or larger roots tend to increase the biomass with growth. Therefore, they also reported that the variation of root biomass is smallest in the surface soil layer and larger in the deeper soil layers. In our study, the CV in the 0–2 cm soil layer was relatively small (16 %), while those in the deeper layers were large (Table 1). Although there may be differences in changes in root biomass within the top and bottom in the surface 15 cm of soil as they grow, no clear temporal trend was observed in our study because of the dense tree density and large basal areas (Fig. 2b). Although root system development may contribute to changes in the $^{137}$Cs inventory with time, it was considered that the factor of the large variation in root biomass was mainly the distance of roots from tree trunks in the present study.

The root biomass decreases with distance from the trunk, but the greater the distance from the trunk, the larger the area (Fig. 3). In our

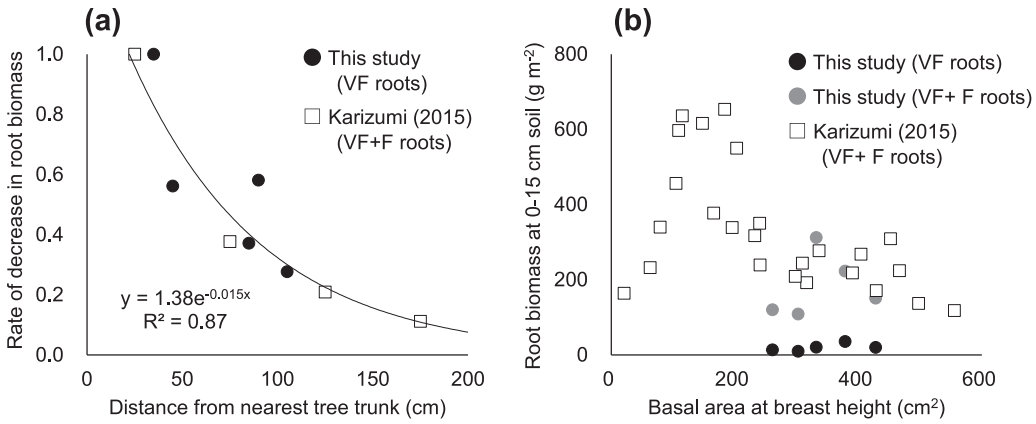


**Fig. 2.** Relationship between the root biomass and the distance from the nearest tree trunk (a) or basal area at breast height (b) in Japanese cedar forests. (a) The rates of decrease were calculated by the VF root biomass at 35 cm from the nearest tree trunk as 1 for this study, and calculated by the fine (F) root biomass <2 mm at 25 cm from the nearest tree trunk in a six year-plantation forest with a stand density of 2500 trees $ha^{-1}$ as 1 for Karizumi (2015). The approximate equation was estimated by all data of our study and Karizumi (2015). (b) Root biomass at 0–15 cm in our study was calculated to be consistent with the depth in Karizumi (2015; 147 cedar trees at 25 sites).

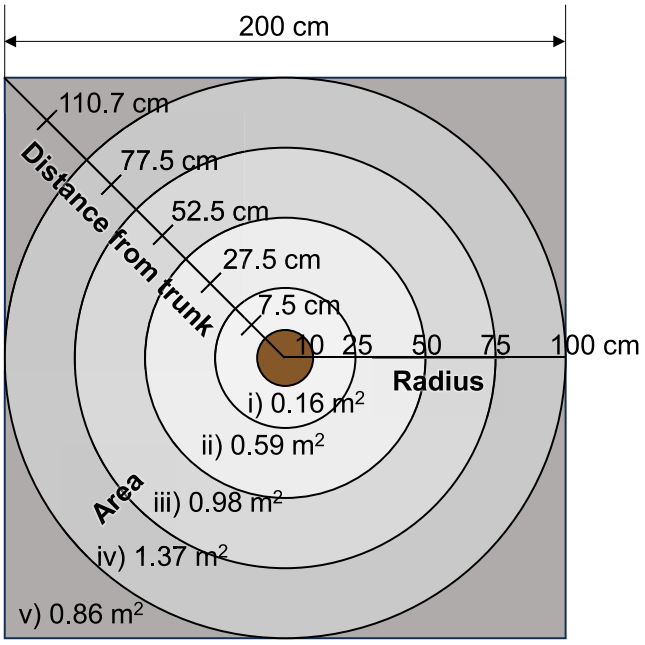


**Fig. 3.** Area corresponding to distance from trunk. Each cedar tree was assumed to be planted 2 m apart with a DBH of 20 cm. Based on the exponential equation between the root biomass and the distance from the nearest tree trunk (Fig. 2a), VF root biomass in each area was calculated to be 1.7 g/0.16 $m^2$, 6.5 g/0.59 $m^2$, 16.0 g/0.98 $m^2$, 18.3 g/1.37 $m^2$ and 15.6 g/0.86 $m^2$ from i–v, respectively. Thus, the average of VF biomass in the study site was estimated to be 14.5 g $m^{-2}$.

study site, cedar trees are planted at 2 m intervals, we attempted to calculate the average VF root biomass per forest unit by assuming that one cedar tree with a DBH of 20 cm is planted at 2 m × 2 m. The distance from the trunk was divided into five steps (Fig. 3) and calculated using the exponential equation (Fig. 2a) between the root biomass and the distance from the nearest tree trunk, the average of VF biomass in the study site was estimated to be 14.5 g $m^{-2}$. This value is smaller than the VF root biomass of cedar trees (31–159 g $m^{-2}$) in the Chubu and Kansai districts in the central part of Japan reported by Wada et al. (2019). In addition, the accuracy of this estimation is not high because the data for the reference value at a decrease rate of 1 in Fig. 2a is only one in 2020. Further investigations are needed to improve the estimation of the detailed depth and horizontal distribution of root biomass.

### 3.2. Depth distribution of $^{137}Cs$ and $^{133}Cs$ concentrations in roots and soils

Fig. 4 and Fig. 5 show the depth distribution of $^{137}Cs$ and $^{133}Cs$ concentrations in the litter/soil (a), VF roots (b) and VF root/soil ratio of $^{137}Cs$ concentration (c). We previously reported that the young cedar forest in the present study have a high stand density, and the initial canopy interception and subsequent secondary deposition of $^{137}Cs$ were larger compared with the less dense mature cedar forest, resulting in a clear increase in $^{137}Cs$ inventory in the forest floor (litter and soil) and a slower decrease in $^{137}Cs$ concentration in the litter layer over time during the 6-year observation period (Takahashi et al., 2019). The $^{137}Cs$ concentration in the litter layer still decreases exponentially to 0.84 Bq $g^{-1}$ by 2023 (Fig. 4a). This is almost the same level as that in mature cedar forests (0.38 Bq $g^{-1}$); hence it is assumed that the effect of initial canopy interception is now almost over. Reflecting this decrease, the $^{137}Cs$ concentration in the soil layers increased with time. Especially after 2020, the $^{137}Cs$ concentration below 2 cm clearly increased, while that in 10–20 cm showed no clear trend (Takahashi et al., 2024).

The $^{137}Cs$ concentration in the VF roots was higher in the surface layer and decreased with depth, similar to that in the soil (Fig. 4b). However, the $^{137}Cs$ concentration in the VF roots below 15 cm exceeded that in the soil layers. Sakashita et al. (2020) reported that the similar result was observed below 5 cm as of 2013. Regarding temporal changes, the $^{137}Cs$ concentration in roots in the 0–2 cm layer decreased with time, unlike the soil.

The $^{133}Cs$ concentration was lower in the litter layer (approximately 1/25–1/10 of the soil) and slightly lower in the humus-rich surface layer (Fig. 5a). This trend was reported by Nguyen et al. (2022), indicating that the dominant source of $^{133}Cs$ is primary minerals in the soil. According to the geochemical map of Japan (Geological Survey of Japan, 2005), the Cs concentration in the study region is 2.4–3.4 ppm, which is roughly similar to the $^{133}Cs$ concentration in the lower soil layers (below 4 cm).

The $^{133}Cs$ concentration in the roots was low in the surface 0–4 cm of soil (Fig. 5b). The percentage of exchangeable $^{133}Cs$ fraction to total $^{133}Cs$ in soils ranged from 0.52 % to 0.61 % in 0–2 cm, but from 0.92 % to 0.96 % in 10–15 cm. This was also similar to Nguyen et al. (2022) who

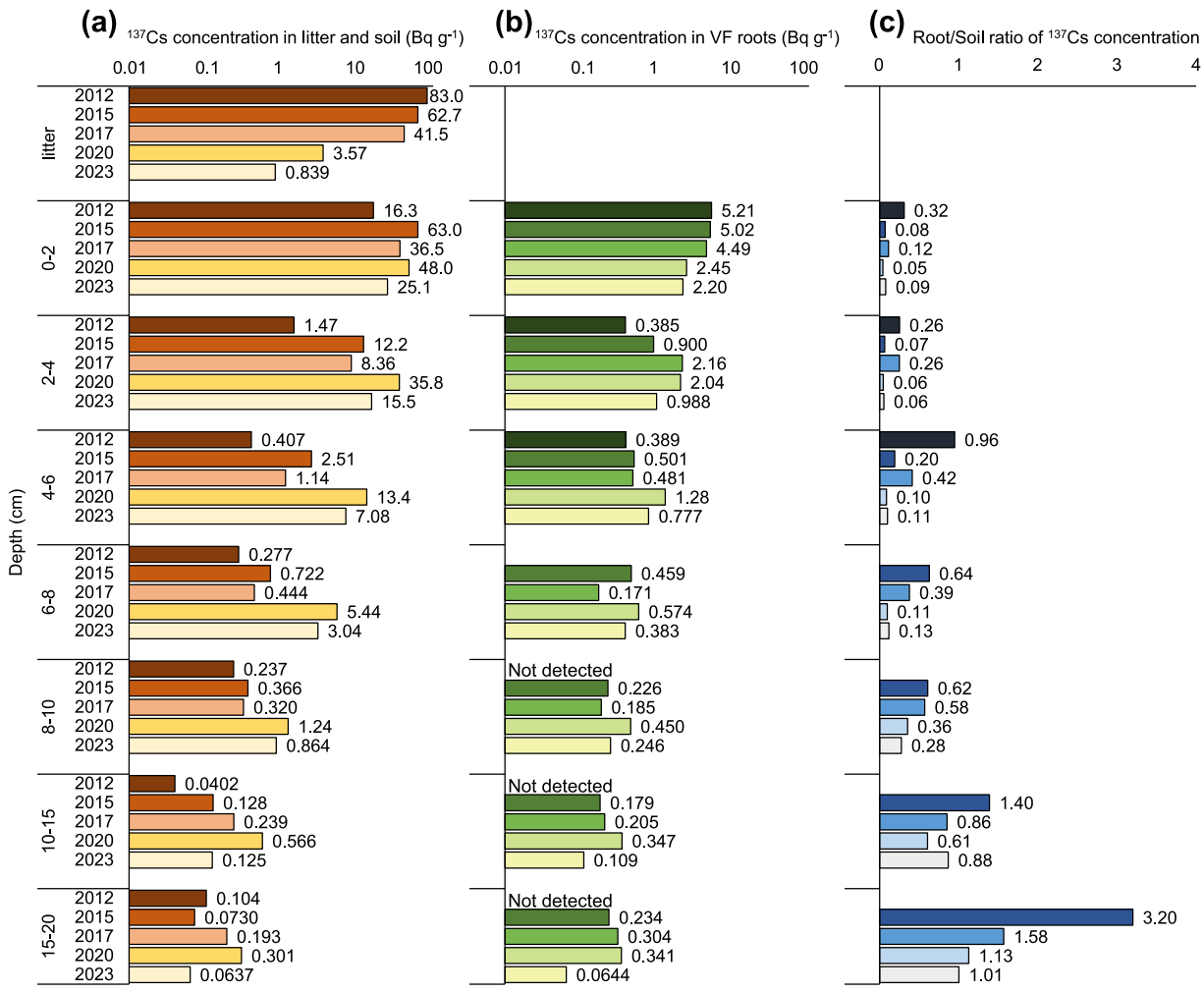


**Fig. 4.** Depth distribution of $^{137}$Cs in litter and mineral soil layers (a), very fine (VF) roots (b) and VF root/soil ratio of $^{137}$Cs concentration (c). The concentrations in the soil at 0–10 cm depth were calculated as weighted averages measured at 5- or 10-mm intervals. The blanks indicate that they were below the detection limit for $^{137}$Cs concentrations.

reported 0.72 % in 0–5 cm and 1.0 % in 10–15 cm, considering that the surface soil layers were significantly affected by the aging process, which is caused by the dry-wetting cycle. In addition to the slightly lower $^{133}$Cs concentration in the surface soil and litter layers, this lower fraction of exchangeable $^{133}$Cs may cause the lower $^{133}$Cs concentration of VF roots in the surface 0–4 cm of soil. On the other hand, relatively high values of $^{133}$Cs concentration of VF roots were found in 4–8 cm layers in 2012 and in 10–20 cm layers in 2015. In order to evaluate the contamination of soil particles in the root samples, the Al concentration in the decomposed solution of roots was measured. As a result, the Al concentration ranged from 6.6 mg $g^{-1}$ to 10.9 mg $g^{-1}$ with CV 19 %, which tended to be higher than the values (2.8–6.4 mg $g^{-1}$) reported in Miwa et al. (1998, 1994). However, there was no relationship between $^{133}$Cs and Al concentrations in roots, the reason for the higher $^{133}$Cs concentrations in these samples was not clarified.

Although the depth distributions between $^{137}$Cs and $^{133}$Cs concentrations were different, the trend that the concentration ratio of root/soil was higher downward was similar (Figs. 4c and 5c). Sakashita et al. (2020) pointed out that the higher $^{137}$Cs root/soil concentration ratio in deeper soil layers may represent the translocation of $^{137}$Cs from the tree bodies on above-ground or roots in the shallow soil layer to roots in the deeper soil layers occurred and may contribute to promote the downward migration of $^{137}$Cs in soils and changing the long-term depth distribution of $^{137}$Cs. Our result that the $^{133}$Cs concentration is higher in the deeper soil layers indicates that the downward translocation of $^{137}$Cs within the roots cannot be a passive transport following a concentration gradient because the $^{133}$Cs concentrations are $10^8$–$10^9$ times higher than $^{137}$Cs at molecular concentrations. Several studies have shown that $K^+$ ions absorbed by the roots are transported through the xylem to the leaves, of which 40–50 % are translocated back to the roots (Jeschke et al., 1985, 1992), and the $K^+$ transporter transports $K^+$ ions and other ions in this order: K > Cs > Rb > Na > $NH_4$ (Gupta et al., 2018). Thus, Cs ions could be translocated to roots in deeper soil layers by active transport. On the other hand, other factors may also explain the higher $^{137}$Cs root/soil concentration ratio in deeper soil layers. We found that the higher exchangeable fraction in deeper soil layers not only for $^{133}$Cs but also for $^{137}$Cs (5.3 %–6.8 % in 0–2 cm and 10.7–12.6 % in 10–15 cm), indicating the higher bioavailability of both $^{137}$Cs and $^{133}$Cs in the deeper layers.

### 3.3. Factors determining Cs concentrations of VF roots

In order to clarify the factors determining Cs concentrations in roots, some correlation analyses were performed. As a result, the $^{137}$Cs concentrations in the VF roots at 0–2 cm for five years were positively correlated with those in litter layers (Fig. 6a), but not correlated with those in the 0–2 cm of soil layers (Fig. 6b). The percentage of exchangeable $^{137}$Cs fraction in the 0–2 cm of soil layers was 5.7 %, 6.7 %, 6.8 %, 5.3 %, and 5.8 % in order from 2012, showing no clear temporal change and a very similar relationship with Fig. 6b.

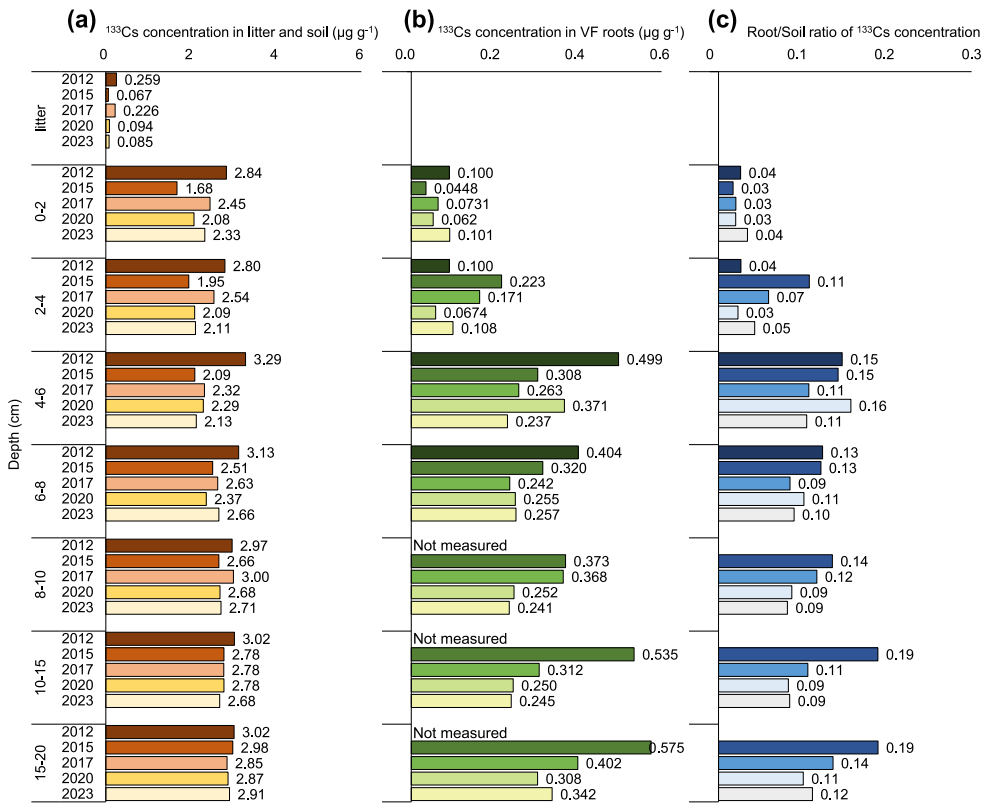


**Fig. 5.** Depth distribution of $^{133}$Cs in litter and mineral soil layers (a), very fine (VF) roots (b) and VF root/soil ratio of $^{133}$Cs concentration (c). The concentrations in the soil at 0–10 cm depth were measured using mixed soil samples with two or four soil layers based on sampling weight. The blanks indicate that they were not measured for $^{133}$Cs concentrations because $^{137}$Cs concentrations were below the detection limit.

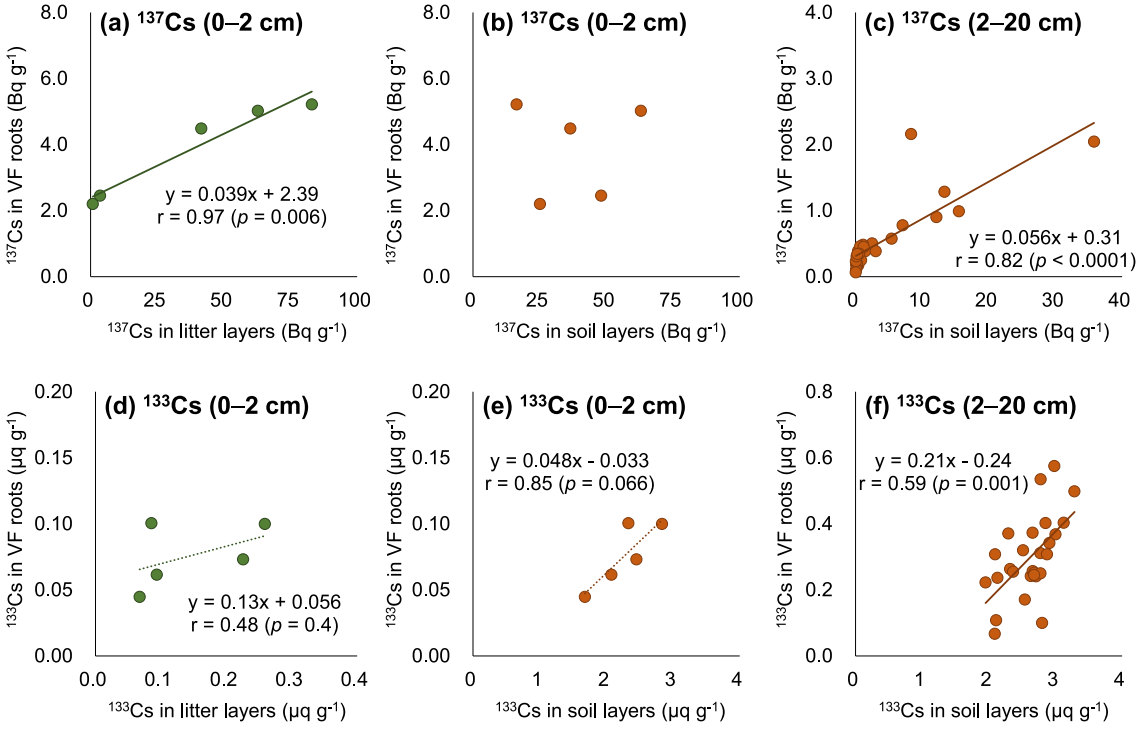


**Fig. 6.** Correlations of $^{137}$Cs (top) or $^{133}$Cs (bottom) concentrations between the VF roots at 0–2 cm and litter layers (a, d), the VF roots and soil layers at 0–2 cm (b, e) and the VF roots and soil layers at 2–20 cm (c, f) for five years data. The solid lines indicate a significant correlation ($p < 0.05$), and the dashed lines indicate that the correlation is $p > 0.05$.

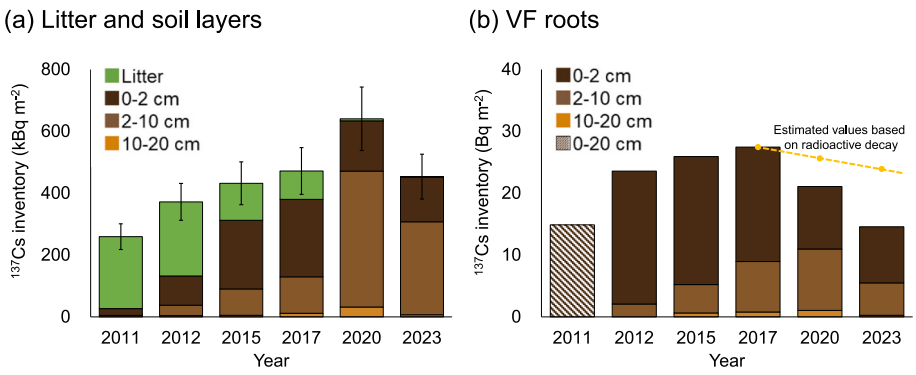


**Fig. 7.** Temporal changes in the $^{137}$Cs inventory in litter and soil layers (a), and in VF roots (b). The data for roots in 2011 is the value obtained using different sampling methods and not available by depth. (a) Error bar represents the estimated error of 16 % based on the sampling area of 450 cm$^2$ (Kato et al., 2022). (b) Yellow line represents the decrease in the $^{137}$Cs inventory estimated by only radioactive decay based on the observed value in 2017, which was the peak of the $^{137}$Cs inventory of VF roots.

In contrast, a higher correlation was observed between the $^{133}$Cs concentrations in the VF roots and the soil layer at 0–2 cm (Fig. 6e) than those between the roots and the litter layer (Fig. 6d), probably because the $^{133}$Cs concentration in the litter layer is approximately one-tenth that in the soil, unlike that for $^{137}$Cs. In fact, the exchangeable fraction of $^{133}$Cs in the litter layer is often below the detection limit (Manaka et al., 2019).

In the soil layer below 2 cm, positive correlations were found between the VF roots and the soil layers for both $^{137}$Cs and $^{133}$Cs concentrations (Fig. 6c, f). Root extension into the litter layer has been shown to be effective not only in Mor-type forests with thick Oa litter layers and nutrient-poor E soil horizon but also in all forest types if the litter layer persists throughout the year and there is sufficient moisture (Sayer et al., 2006). However, little root extension into the litter layer was observed in our study site, which has no Oa layer. In our previous study, we installed zero-tension lysimeters in an adjacent mature cedar forest (Fig. 1a) and found that the $^{137}$Cs inventory in the litter layer decreased by 7.3 kBq m$^{-2}$ within one year (August 2017–July 2018), of which approximately 0.6 kBq m$^{-2}$ was supplied to the soil as rainfall infiltration water (Takahashi et al., 2022). These values were much higher than the $^{137}$Cs inventory in the VF roots, suggesting that the $^{137}$Cs supplied from the litter layer was important at a depth of 0–2 cm because of its high bioavailability. If the $^{137}$Cs concentration in the litter layer decreases further in the future, it can be assumed that the $^{137}$Cs concentration in roots in the 0–2 cm-deep soil layer will be determined by the $^{137}$Cs concentration in soil as well as $^{133}$Cs.

Several simulation models have been developed to predict $^{137}$Cs concentrations in forest components and to estimate the amount of $^{137}$Cs absorbed by roots using the soil, litter and wood concentrations, or the amount of soil water absorbed by the roots. According to a report comparing the long-term simulations of these models (Hashimoto et al., 2021), the uncertainty of root uptake amount of $^{137}$Cs and the exclusion of effects of the root system development cause the discrepancy among the models. Moreover, some models consider $^{137}$Cs uptake from both litter and soil layers, whereas others only consider $^{137}$Cs uptake from the soil layer. Roots absorb $^{137}$Cs, which is then transported through the trunk to the branches and leaves. Thus, the $^{137}$Cs concentration in the roots does not directly reflect the amount of $^{137}$Cs taken up by the roots. However, the lack of time-series data on $^{137}$Cs concentrations in roots and the finding of whether roots absorb $^{137}$Cs from both the litter and soil layers or only from the soil layer cause the errors of these models, and our findings are expected to contribute to the improvement of the model.

### 3.4. Temporal change in the $^{137}$Cs inventory in VF roots

The calculation of the $^{137}$Cs inventory is strongly influenced by the root biomass. In this study, the root biomass varied greatly due to the difference in distance from the trunk at each sampling. Therefore, the $^{137}$Cs inventory was calculated using the estimated average of VF biomass (14.5 g m$^{-2}$) and the distribution ratio of each layer (Table 1). Moreover, we obtained the value for July 2011 using root samples collected from the A1 horizon (0–14 cm) by multiplying by the 0–20 cm biomass. This value was considered comparable to that of the other four surveys because the $^{137}$Cs inventory of the roots in the 15–20 cm-deep soil layer was small (0–3.6 %).

The $^{137}$Cs inventories in the forest floor (litter and 0–20 cm-deep soil layers) in 2011, 2012, 2015, 2017, 2020 and 2023 were 259, 372, 432, 472, 641 and 454 kBq m$^{-2}$, respectively, and increased with time until 2020 due to secondary deposition through litterfall and throughfall from the canopy (Fig. 7a). The initial deposition density estimated by airborne monitoring was 440 kBq m$^{-2}$ at this study site (Kato et al., 2019a), so 641 kBq m$^{-2}$ in 2020 may have been increased by stemflow because the sampling point was closer to the tree trunk (Imamura et al., 2017b). In addition, deposition of $^{137}$Cs in the forest is highly spatially variable, resulting in that an estimated error of $^{137}$Cs inventory based on the sampling area was 16 % for the 450 cm$^2$ in this study (Kato et al., 2022). The $^{137}$Cs inventory in the litter layers decreased significantly with time, and $^{137}$Cs in the surface soil layers migrated into deeper soil layers. However, the percentages of $^{137}$Cs in the 10–20 cm-deep soil layer were 0 %, 1.1 %, 1.2 %, 2.5 %, 5.0 % and 1.5 % of the $^{137}$Cs inventory in the forest floor from 2011 to 2023, respectively; thus, 95 % of $^{137}$Cs is still present within 10 cm of the surface layer.

The $^{137}$Cs inventories in the VF roots were 14.9, 23.6, 25.9, 27.4, 21.1 and 14.6 Bq m$^{-2}$ from 2011 to 2023, respectively; and increased from 2011 to 2017 but decreased after 2020 (Fig. 7b). The $^{137}$Cs inventory in the VF roots in the 0–2 cm-deep soil layer reached 91 % in 2012; however, it tended to decrease with time to approximately 48 % in 2020 and 62 % in 2023. As mentioned above, the $^{137}$Cs concentration in the VF roots in 0–2 cm decreased with time (Fig. 4b) as the $^{137}$Cs concentration in the litter layer decreased (Fig. 4a), resulting in the positive correlation between the VF roots and litter $^{137}$Cs concentrations (Fig. 6a). Since the VF root biomass decreased exponentially with depth, the decrease in $^{137}$Cs concentration of the roots in the 0–2 cm layer was very effective in reducing the $^{137}$Cs inventory.

Fesenko et al. (2001) showed that the better estimation of the $^{137}$Cs concentration in trees can be obtained by multiplying the depth distribution of $^{137}$Cs in soils by the depth distribution of its bioavailability and tree root biomass in each soil layer. Moreover, a study that estimated the rooting depth of $^{137}$Cs by plant uptake by comparing the $^{137}$Cs/$^{133}$Cs ratio of the exchangeable fraction in soil and aboveground plants showed that deep-rooted species, such as bamboo grasses, have a lower accumulation of $^{137}$Cs than superficial-rooted species such as ferns (Nguyen et al., 2022). Although these reports have already highlighted

the importance of the root system distribution, this is the first study to demonstrate that changes in the depth distribution of $^{137}$Cs in the soil associated with downward migration reduce the amount of $^{137}$Cs absorbed by trees.

Other factors that will reduce the $^{137}$Cs concentration and inventory in VF roots after 2020 include the radioactive decay of $^{137}$Cs and fixation of $^{137}$Cs by soils. In the 12 years since the accident, even $^{137}$Cs has been reduced by about 24 % due to radioactive decay. However, the estimated value of the $^{137}$Cs inventories of VF roots in 2020 and 2023 if those were reduced only by radioactive decay based on the year 2017 were larger than the observed values in 2020 and 2023, indicating that only radioactive decay cannot explain this decrease (Fig. 7b). It is well known that the fixation of Cs ions by soils takes time and is called the aging effect (e.g., Roig et al., 2007). In fact, it has been confirmed that the exchangeable fraction of $^{137}$Cs decreases after 2–4 years following the Fukushima accident (Manaka et al., 2019). However, as mentioned in Section 3.3, there is no clear temporal change in the percentage of exchangeable fraction of $^{137}$Cs in the 0–2 cm of soil layers in this study, it was not possible to evaluate this aging effect.

Several model simulations predict that the $^{137}$Cs concentrations in wood would increase after a nuclear accident, then gradually decrease (Hashimoto et al., 2021). This trend is similar to that of the $^{137}$Cs inventory in the roots examined in this study (Fig. 7b). They discussed that the initial increase in $^{137}$Cs concentration in wood is due to an increase in the $^{137}$Cs inventory of soil layers, and the subsequent decrease is due to radioactive decay, dilution by increased amounts of wood biomass, and immobilization of $^{137}$Cs in the soil (Hashimoto et al., 2021). However, the relationship of depth distribution between root biomass and $^{137}$Cs was not discussed, indicating that our findings could improve the model predictions.

### 3.5. Suggestion of self-cleaning effect in forest ecosystems

Our results revealed that natural processes, such as the downward migration of $^{137}$Cs, have the effect of self-cleaning, which reduces root uptake or $^{137}$Cs circulation in the forest ecosystem. It has also been reported that the air dose rate in the forest is largely contributed by $^{137}$Cs contained in the surface soil within 5 cm, and the progression of the downward migration of $^{137}$Cs in the soil has a significant effect on reducing air dose rate (Malins et al., 2021). Hence, it can be concluded that the downward migration of $^{137}$Cs is effective in reducing both root uptake and the air dose rate, indicating that green and sustainable remediation using such self-cleaning effects should be actively adopted for the future forest managements instead of physical decontamination, such as the removal of litter layers. Especially, the faster reduction of $^{137}$Cs concentration in the litter layer in Fukushima forests than in Chernobyl (Imamura et al., 2020) suggests a higher self-cleaning effect in Fukushima.

In addition, thinning is expected to increase canopy openness, throughfall, and soil infiltration water in the forest and promote the decomposition of litter and soil organic matter that retains $^{137}$Cs due to increased solar radiation. These may actively promote the downward migration of $^{137}$Cs in the soil. Conversely, it has long been recognized that unmanaged plantation forests without proper thinning increase the risk of soil erosion owing to little understory vegetation and reduced water permeability (Miura et al., 2002; Nanko et al., 2008). Therefore, it can increase $^{137}$Cs runoff to downstream areas. The resumption of proper forest management (thinning) could be the most effective sustainable remediation method based on the self-cleaning of forest ecosystems.

There is often a desire for more complete remediation of radioactive contamination than is necessary. It was pointed out that if the science underlying ecological remediation are not understood by the general public, implementation will fail for lack of public support (Cairns, 2000). Our results showed that little downward migration occurred below 20 cm, indicating that the risk of reaching groundwater is very low. On the other hand, it was reported that most roots in apple orchards are distributed below 10 cm of the soil, and conversely, nitrogen fertilization may be effective in reducing radiation risk by delaying the downward migration of $^{137}$Cs through increased fixation to surface soils (Matsuoka et al., 2020). A deep understanding and dissemination of these natural processes tailored to each ecosystem are important for promoting green and sustainable remediation.

## 4. Conclusions

In the present study, we monitored the depth distribution of $^{137}$Cs concentration and inventory in the soil and VF roots from 2011 to 2023 at a young cedar forest site. The $^{137}$Cs concentration in the litter layer continued to decrease exponentially as of 2023, resulting in that the $^{137}$Cs concentration in soil layers increased with time. Especially after 2020, the $^{137}$Cs concentration below 2 cm clearly increased because of the downward migration of $^{137}$Cs to deeper soil layers. The $^{137}$Cs concentrations in the VF roots at 0–2 cm correlated positively with $^{137}$Cs concentrations in the litter layer and decreased with time. Although the $^{137}$Cs concentrations below 2 cm positively correlated with $^{137}$Cs concentrations in the soil and tended to increase with time, the $^{137}$Cs inventory in VF roots, i.e. the amount of $^{137}$Cs absorbed by VF roots, would be reduced with temporal downward migration of $^{137}$Cs because the VF root biomass decreases exponentially in deeper layers. This natural downward migration process can be regarded as a “self-cleaning” of the forest ecosystem, resulting in the reduction of root uptake of $^{137}$Cs and air dose rate. This study provided information on the detailed distribution of the absorptive root system and its $^{137}$Cs concentration, which was tended to be lacking. Further investigations such as the root system development, spatial variability of $^{137}$Cs and depth distribution of $^{137}$Cs exchangeable fraction are necessary to improve the long-term prediction of root uptake of $^{137}$Cs.

## CRediT authorship contribution statement

**Junko Takahashi:** Writing – original draft, Visualization, Validation, Supervision, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization. **Satoshi Iguchi:** Investigation, Formal analysis. **Takuya Sasaki:** Methodology, Investigation, Formal analysis. **Yuichi Onda:** Supervision, Project administration, Methodology, Funding acquisition.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

A data set of the depth distribution of $^{137}$Cs concentration in soils from 2011 to 2023 is available at doi:10.34355/CRiES.U.Tsukuba.00206. All other data supporting the findings of this study are available in this article.

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