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Spatial variation and radiocesium flux of litterfall in hardwood-pine mixed forest and cedar plantations based on long-term monitoring data

Keigo Hisadome[1,2] · Yuichi Onda[3] · Nicolas Loffredo[3] · Ayumi Kawamori[2] · Hiroaki Kato[3]

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Abstract
Long-term monitoring revealed that radiocesium (^{137}Cs) deposition that fall from the canopy with litterfall was characterized by forest types, seasonal change, and canopy closure (CC). The sum of autumn and spring ^{137}Cs deposition values divided by the sum of summer and winter tended to have higher values in the cedar forest. The angle of view α when the correlation between the CC size and litterfall amount is strongest, was 5° or less in the young cedar forest, but varied in the hardwood–pine mixed forest. This indicates the spatial distribution of ^{137}Cs deposition has heterogeneity and homogeneity in each forest.

Keywords Radiocesium concentration · Radiocesium deposition · Litterfall amount · Canopy closure · Seasonal change

Introduction

Radioactive materials were released into the atmosphere after the TEPCO Fukushima Daiichi Nuclear Power Plant (FDNPP) accident that occurred on March 11, 2011. More than 70% of the land area where radiocesium (^{137}Cs) was deposited was forested [1, 2]. More than 60% of the ^{137}Cs deposited in the forest was first intercepted by the canopy [3], although much of this fallout was washed away by rain (throughfall) passing through the canopy [4, 5]. Moreover, tree parts (e.g., leaves and twigs) were knocked off by rain and air movement during rainfall events, and then transferred to the forest floor [6–11], causing secondary contamination of the forest floor. After reaching the forest floor, more than 80% of the ^{137}Cs was immediately deposited within 2 cm of the ground surface [3], and then gradually migrated into the soil with the decomposition of leaves and branches (i.e., litter) that had fallen to the forest floor [12].

Much of the deposited ^{137}Cs has remained within the forest, where it is taken up by plant roots [13], transferred to leaves, and then deposited again onto the forest floor. Therefore, ^{137}Cs has repeatedly circulated through the forest system, increasing deposition on the forest floor (i.e., secondary contamination of the forest floor) [3, 14, 15]. By contrast, relatively small amounts of ^{137}Cs have been transferred out of the forest, carried with litter in stream water [16] or with sedimentary organic matter and soil in surface water flowing over the forest floor due to rainfall [17].

Our previous studies revealed that litterfall amounts deposited from the canopy and ^{137}Cs deposition not only differed depending on initial deposition onto each experimental plot and among tree species (i.e., evergreen vs. deciduous), but also with the maturity of the cedar forest [18]. This is due to differences in the amounts of ^{137}Cs initially intercepted by the canopies of young and mature cedar forests. Such differences in ^{137}Cs deposition in the canopy lead to differences in the distribution of ^{137}Cs deposited in forest soil [19]. Therefore, the density of the canopy and its capacity to block radionuclides from reaching the forest floor are strongly correlated [20].

Throughfall and stemflow were the primary routes of ^{137}Cs transfer from the canopy to the forest floor in the first 200 days after the initial fallout occurred [14]; since then, litterfall has played an important role [21, 22]. The spatial

✉ Keigo Hisadome
kgo.hisadome@ajiko.co.jp

1 Environmental Regeneration Section, Environment & Energy Technical Department, Asia Air Survey Co., Ltd, Minamisoma, Fukushima 975-0039, Japan

2 Graduate School of Life and Environmental Sciences, University of Tsukuba, Tsukuba, Ibaraki 305-8572, Japan

3 Center for Research in Isotopes and Environmental Dynamics, University of Tsukuba, Tsukuba, Ibaraki 305-8572, Japan

distribution of ^{137}Cs in soil [23] and the air dose rate at the forest floor [24] reflect the distribution of ^{137}Cs in the canopy. Therefore, studies of litterfall are essential for determining the spatial and temporal variability in the transfer of ^{137}Cs from the canopy to the forest floor. It is necessary to calculate the dose exposure level and clarify the spatial distribution characteristics of litterfall to evaluate secondary environmental contamination.

A number of studies have attempted to determine litterfall amounts and ^{137}Cs deposition levels in Fukushima and the surrounding forests [25–27]. However, such studies included only short-term surveys (~ 1 year), and therefore did not detect variations in litterfall and ^{137}Cs concentration caused by seasonal changes or weather conditions. Studies have also been conducted in the area affected by Chernobyl fallout (e.g., Bunzl et al. [13]; Schimmack et al. [7]); these were also based on data collected over a short period of 1–2 years. In the area affected by the FDNPP accident, Japanese cedar accounts for 24% of the forest area, while secondary forest (mainly pine and oak) accounts for a further 58% [28]. The litterfall from cedar trees varies among seasons [29] and years [30]. Recently, Yoshihara et al. [31], Kato et al. [32] and Nishikiori et al. [33] acquired data on the amount of litterfall and ^{137}Cs concentration over 5–6 years, but did not consider the spatial variability of litterfall or ^{137}Cs transfer in the forest. To characterize the effects of secondary contamination of the forest floor, studies of various forest species are necessary, but very few studies (e.g., Takada et al. [23]; Naoe et al. [34]) have been conducted on litterfall in secondary or hardwood forests. To understand the transfer of ^{137}Cs to the forest floor in different forest types in Fukushima, long-term datasets collected over many years, including data on spatial variability, will be required.

To study the transfer of ^{137}Cs in Fukushima forests, we collected data from three forest types (hardwood–pine mixed forest, mature cedar forest, and young cedar forest) in Kawamata Town, Yamakiya District, where long-term forest research was conducted immediately after the FDNPP accident. In the Yamakiya model forest, we collected and evaluated litterfall amounts. We studied the relationships among the angle of view (α) defining canopy closure (CC: meaning the percentage of canopy closing the sky), litterfall amount, and the amount of ^{137}Cs deposited (meaning radiocesium flux of litterfall), as well as the relationship between litterfall amount and ^{137}Cs concentration.

Materials and methods

Forest sites

In this study, we selected Kawamata Town (Yamakiya District, Fukushima Prefecture, Japan) as the study area. The study site is located upstream of the Kuchibuto River within the Abukuma River water system. We selected three forest stands as experimental plots (Fig. 1), in which several previous studies have been conducted, to investigate the transfer of ^{137}Cs in the forest environment [3, 10, 11, 14, 19, 32], soil and litter layers [35, 36], and the air dose rate [24]. Following the FDNPP accident in 2011, the three experimental plots in the Yamakiya District received ^{137}Cs deposition amounts of 300–600 kBq/m^2 according to the Third Airborne Monitoring Survey [37, 38] (as of July 2, 2011).

To study the transfer and accumulation of radioactive materials in different forest types in Yamakiya District, a mature cedar (*Cryptomeria japonica*) forest (about 50 years old), and young cedar forest (about 23 years old) were selected as representative coniferous forests, and a mixed forest consisting of Japanese oak (*Quercus aliena*) and Japanese red pine (*Pinus densiflora*) was selected as a representative hardwood–pine mixed forest. The leaves of cedar and red pine in mixed forests are evergreen and fall all year round, while almost all the leaves of Japanese oak in mixed forests fall in autumn.

Litterfall from the forest canopies was sampled periodically to investigate the distribution of radioactive materials in the forest, and changes in the accumulation of radioactive materials over time. To collect litterfall from the canopy, three litterfall collectors were installed in each stand, denoted hereafter as TRAP1, TRAP2, and TRAP3 of the mature cedar (MC), young cedar (YC), and hardwood–pine (HP) forests. The litterfall collector was a structure comprising a fine mesh attached to a table, with a height of 1 m and an area of 1 × 1 m, constructed from a PVC pipe frame (see photo in Fig. 1).

Figure 1 illustrates the arrangement of the tree trunks, canopy, and litterfall collectors in each stand.

Sample collection and measurement methods

Four years of monitoring was conducted from July 3, 2011, to June 22, 2015. Initially, we collected samples twice monthly, but gradually reduced the sampling frequency, collecting samples in 2015 whenever the litterfall collectors contained sufficient amounts of sample. The litterfall trapped in each litterfall collector was sealed in a polyethylene bag and transported to the laboratory. In the laboratory, samples were dried in an oven at 110 °C for 24 h, weighed, and pulverized in a U-8 container made of polystyrene with a volume of 100 mL. The resulting samples were used for measurement of the ^{137}Cs concentration. In addition, samples from the hardwood–pine mixed forest were separated into hardwood and pine samples through September 2012. To analyze the spatial distribution of Cs concentration and litterfall amount as well as the change with time, we used

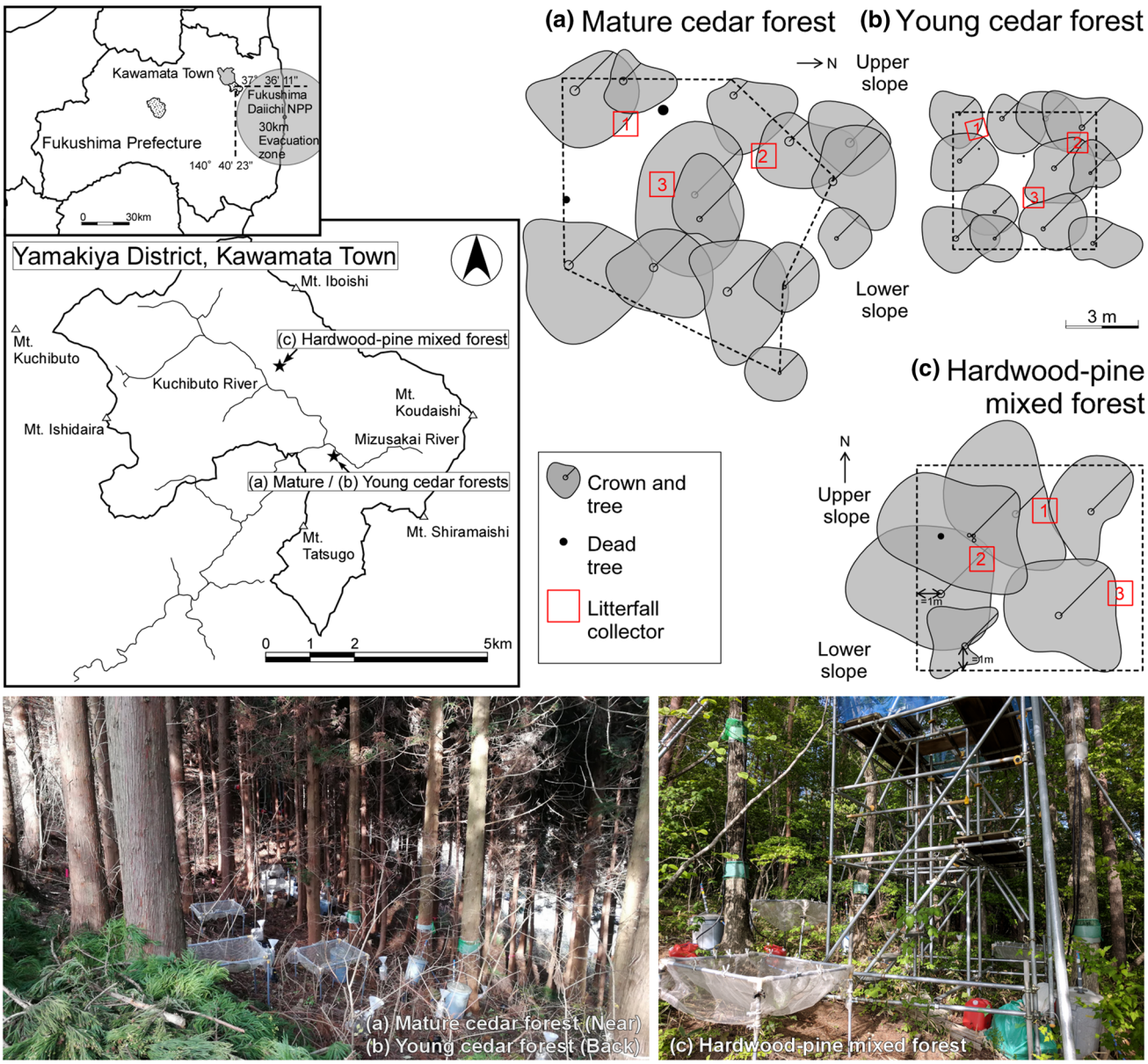

Fig. 1 Left: Location of the study site in Kawamata Town (Yamakiya District, Fukushima Prefecture, Japan). Sampling was conducted at three locations including a hardwood–pine mixed forest, mature cedar forest, and young cedar forest. Right: Canopy distribution and locations of litterfall collectors in the **a** mature cedar, **b** young cedar, and **c** hardwood–pine mixed forest experimental plots. Photographs show litterfall collectors installed in the cedar and hardwood–pine mixed forests.

^{137}Cs with a long half-life of about 30 years, instead of ^{134}Cs with a short half-life of about 2 years.

The ^{137}Cs concentrations in the prepared samples were measured using a high-purity n-type germanium coaxial gamma-ray detector (EGC25-195-R; Canberra-Eurisys, Meriden, CT, USA) with an amplifier (PSC822; Canberra-Eurisys) and a multichannel analyzer (DSA1000; Canberra, Paris, France). During the preparation and measurement of samples, care was taken to prevent cross-contamination. The ^{137}Cs concentration and ^{137}Cs deposition were calculated through analysis of measured spectral data. Unless otherwise specified, the ^{137}Cs concentration was corrected to the value of each sampling date. ^{137}Cs deposition was calculated as the product of litterfall amount and ^{137}Cs concentration per collection period and per collector.

For the sampling period without ^{137}Cs concentration data (from April to June 2014 in the mature cedar forest and young cedar forest, and from April to August 2014 in the hardwood-pine mixed forest), we estimated the ^{137}Cs concentration in litterfall material based on the logarithmic regression analysis presented in Kato et al. [32]

Calculation of CC from hemispherical photographs

Four hemispherical photographs were taken at each of the nine sites where litterfall collectors were located (Fig. 2). A fisheye lens (FC-E9; Nikon, Tokyo, Japan) was attached to a compact digital camera (COOLPIX E 5700; Nikon) and the shutter speed was fixed at 1/250 s. The ISO sensitivity was 100, the aperture value was fixed at F5, and photographs were taken without a flash or zoom. All photographs were acquired on June 22, 2015.

The CC data were analyzed using the commercially available CanopOn 2 software (ver. 2.03c; Takenaka [39]) to obtain hemispherical photographic images, which were converted into black and white binary images (see upper right panel, Fig. 2); we calculated the average porosity and tree CC at seven levels of α (1°, 3°, 5°, 8°, 14°, 22°, and 30°: see left panels, Fig. 2) from the converted images. After calculating the values for each photograph, the average values were calculated for each sampling site and used as the representative values.

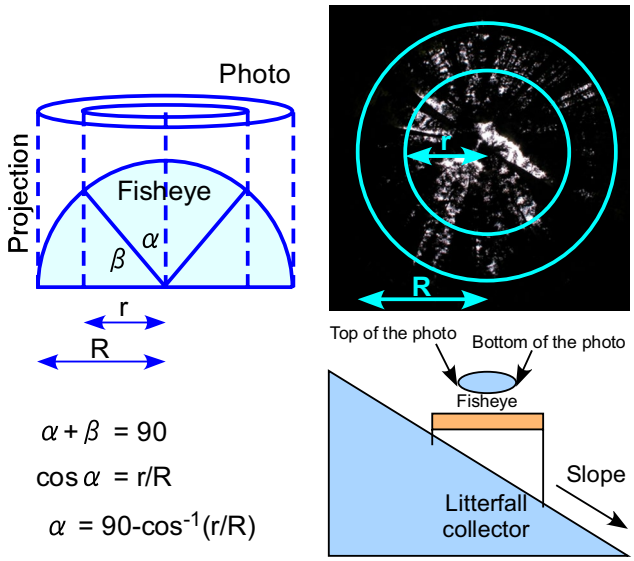

Fig. 2 Lower right: Four photographs taken at the site of each litterfall collector. The direction of the slope is from top to bottom in all images. The exposure of the camera was set according to the method of Chen et al. [44]. Left: Relationship between the angle of view (α) for calculating canopy closure (CC) and the radius r in the projected hemispherical photograph. CC differed depending on α (radius r). Upper right: An example hemispherical photograph taken at the location of a litterfall collector (YC-TRAP 3) in the young cedar forest. The radius r reached its maximum value R when $\alpha = 90°$

Results

Litterfall amount by season

The litterfall amounts recovered from each collector were summed for spring, summer, autumn, and winter (Table 1) and plotted as a time series to show temporal trends (Fig. 3).

In the mature cedar forest, the litterfall amount was higher in autumn and spring and lower in summer and winter, with this pattern seen every year from 2011 to 2013. The maximum litterfall occurred in autumn 2013, with MC-TRAP3 reaching a value of 993.58 g/m^2 and gradually decreasing thereafter. The litterfall amount collected in MC-TRAP2 was generally greater than in the other collectors.

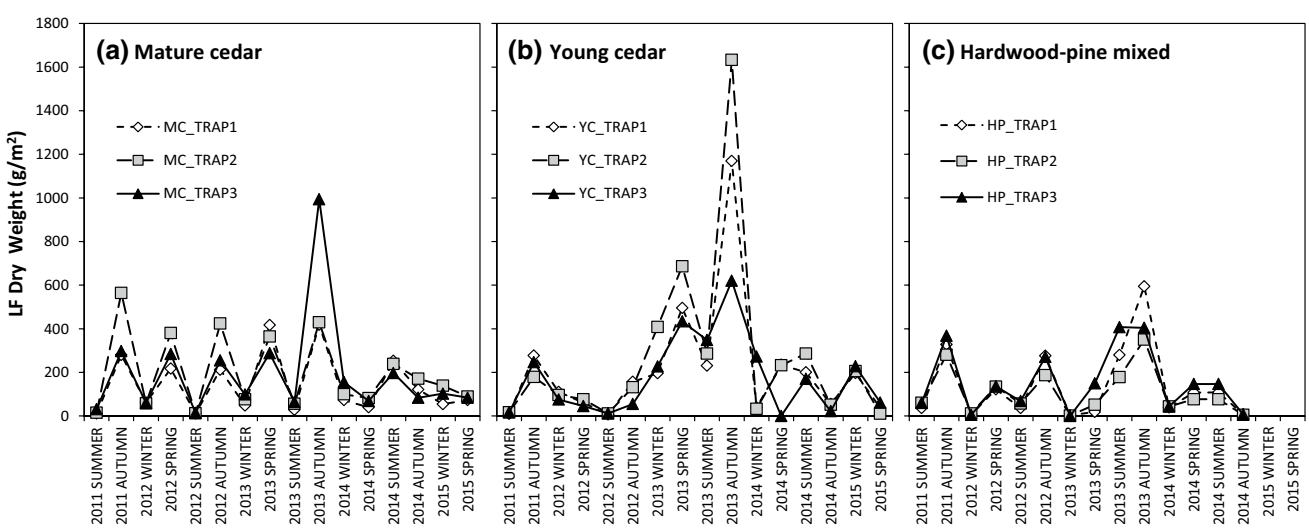

Fig. 3 Seasonally aggregated litterfall (LF) amounts in the three collectors (TRAP1–3) in the **a** mature cedar, **b** young cedar, and **c** hardwood–pine mixed forest plots. In the hardwood–pine mixed forest, hardwood and pine were measured separately through September 2012, but are displayed as the total amount per collector

Table 1 The sampling period in each experimental plot was divided into four seasons (spring, summer, autumn, and winter)

Season	Sampling date	
	Cedar	Hardwood-pine mixed
2011 SUMMER	2011/7/10–9/23	2011/7/10–9/24
2011 AUTUMN	2011/9/23–12/28	2011/9/24–12/28
2012 WINTER	2011/12/28–2012/3/8	2011/12/28–2012/3/9
2012 SPRING	2012/3/8–6/22	2012/3/9–6/22
2012 SUMMER	2012/6/22–9/21	2012/6/22–9/21
2012 AUTUMN	2012/9/21–12/29	2012/9/21–12/29
2013 WINTER	2012/12/29-2013/3/1	2012/12/29-2013/3/1
2013 SPRING	2013/3/1–7/14	2013/3/1–6/24
2013 SUMMER	2013/7/14–9/18	2013/6/24–9/18
2013 AUTUMN	2013/9/18–12/16	2013/9/18–12/16
2014 WINTER	2013/12/16–2014/4/4	2013/12/16–2014/4/4
2014 SPRING	2014/4/4–6/17	2014/4/4–8/25
2014 SUMMER	2014/6/17–10/20	2014/8/25–10/20
2014 AUTUMN	2014/10/20–2015/1/5	2014/10/20–2015/1/5
2015 WINTER	2015/1/5–3/16	
2015 SPRING	2015/3/16–6/22	

The frequency and timing of sample collection differed by year because the length of each season varied slightly among study years

In the young cedar forest, the peak litterfall amounts occurred in autumn 2011, spring 2013, and autumn 2013. In autumn 2013, litterfall was particularly high, with 619.98–1,633.17 g/m^2 obtained from the collectors. These amounts exceeded the maximum amount collected in the mature cedar forest at the same time. The standard deviation of the average litterfall amount in collectors in autumn 2013 was 414.12 g/m^2 in the young cedar forest, which was greater than the amounts collected during other periods (2.41–111.88 g/m^2). Generally, more litterfall was collected in YC-TRAP2 than in the other collectors in the young cedar forest.

The maximum litterfall amount in the hardwood–pine mixed forest also occurred in autumn 2013 (594.47 g/m^2 in HP-TRAP1), but its standard deviation was 104.69 g/m^2, which was lower than those of the cedar forests. Generally, more litterfall was collected from HP-TRAP3 than from the other collectors in this forest. The amount of fallen leaves was generally greater under hardwoods than under red pine, but otherwise, the temporal trends in amounts of fallen leaves were similar among collectors.

^{137}Cs concentration in litterfall over time

Temporal trends were similar among the three collectors in both the mature and young cedar forests. In 2011 and 2012, the ^{137}Cs concentration exhibited a local maximum in summer followed by a sharp decline, with a tendency for the absolute values to decrease slightly over time (Fig. 4). After 2013, the ^{137}Cs concentration remained almost unchanged throughout the year, and the variation in concentration among collectors tended to be small. The highest ^{137}Cs concentration in all stands was 574 kBq/kg, which was recorded in MC-TRAP2 in the mature cedar forest from August 19 to September 23, 2011. In the mature cedar forest, the ^{137}Cs concentration in all three collectors tended to be slightly higher from July to October of each year, whereas in the young cedar forest, the timing of peak concentration varied among the three collectors.

In the hardwood–pine mixed forest, there were small but non-significant differences in the ^{137}Cs concentrations among collectors. In HP-TRAP1 and HP-TRAP3, a moderate decline in concentration occurred over time. The peak ^{137}Cs concentration occurred in summer 2011 in HP-TRAP2, and the maximum concentrations were relatively low in 2013 and 2014. The ^{137}Cs concentration in pine litter was high from July 31 to December 5, 2011 (74–130 kBq/kg), with a subsequent sharp decline in all three collectors. For hardwood trees, the peak ^{137}Cs concentration of 273 kBq/kg in HP-TRAP2 was recorded from July 31 to September 19, 2011.

^{137}Cs deposition by season

^{137}Cs deposition was summarized by season and litterfall amount, and changes over time were recorded (Fig. 5). Changes in ^{137}Cs deposition tended to reflect those in litterfall amount. The greatest ^{137}Cs deposition in the mature and young cedar forests occurred in autumn 2013. The maximum deposition was 31.7 kBq/m^2 in the mature cedar forest (MC-TRAP3) and 58.6 kBq/m^2 in the young cedar forest (YC-TRAP2), whereas in the hardwood–pine mixed forest, ^{137}Cs deposition did not vary. The standard deviation of the average ^{137}Cs deposition in autumn 2013 was 8.7 kBq/m^2 in the mature cedar forest and 14.2 kBq/m^2 in the young cedar forest, representing much greater variation than in other periods (0.1–4.3).

Relationship between *α* and CC

The CC for each value of α was calculated for each collector in each forest, and the relationship between α and CC was investigated (Fig. 6). In the mature cedar forest, CC followed similar trends among the three collectors, tending to increase as α decreased. This led to all collectors being covered by forest canopy.

In the young cedar forest, the sites of YC-TRAP1 and YC-TRAP2 showed a tendency toward increased CC as α decreased, as observed in the mature cedar forest, whereas YC-TRAP3 showed a tendency toward decreased CC as α

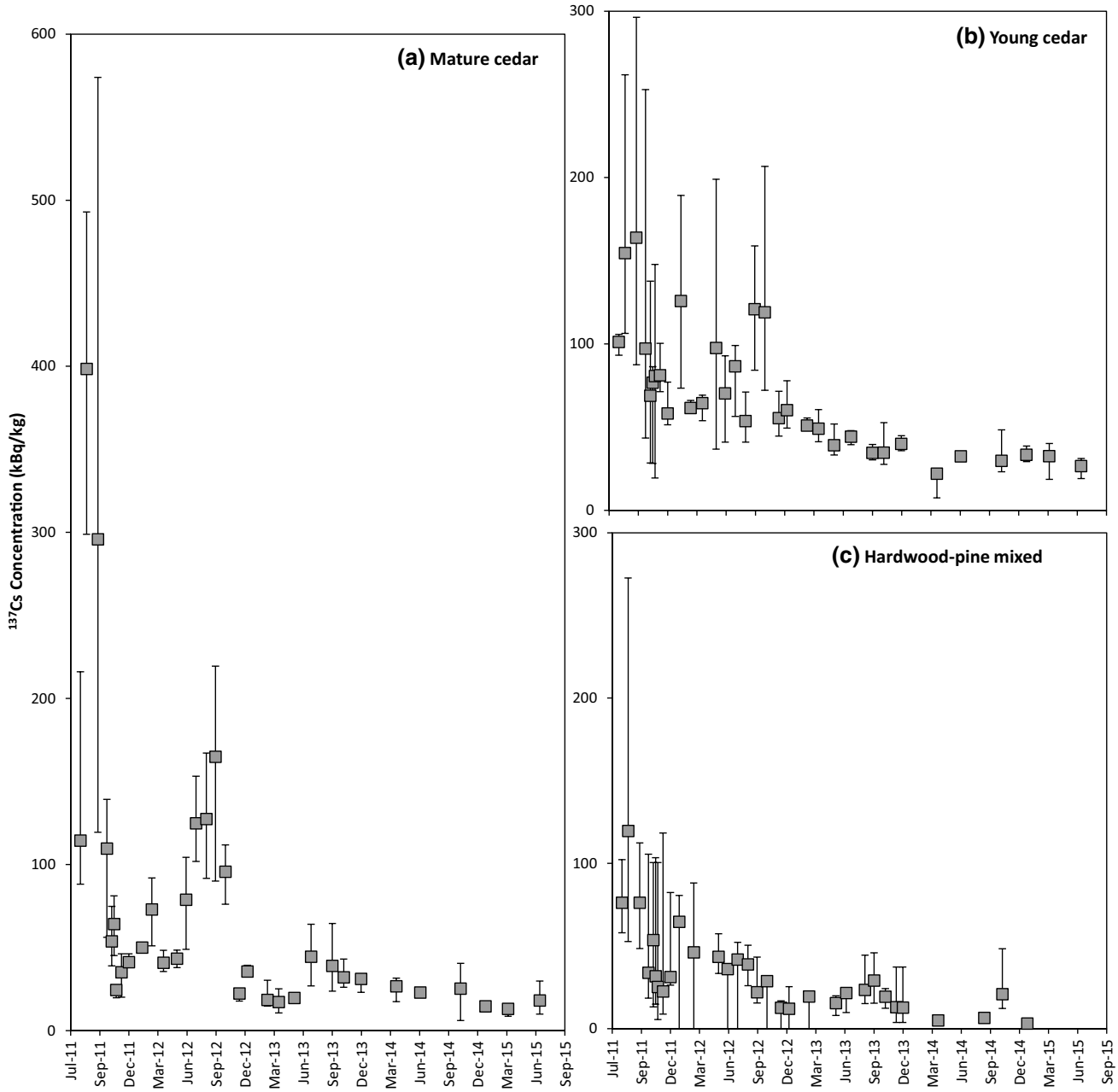

Fig. 4 Average radiocesium (^{137}Cs) concentration by sample collection time in the three litterfall collectors in the **a** mature cedar, **b** young cedar, and **c** hardwood–pine mixed forest types. Error bars indicate the maximum and minimum values. In the hardwood–pine mixed forest, the hardwood and pine were measured separately through September 2012, but are displayed as the total concentration for each collector; the average value for each experimental plot is shown

decreased. The canopy immediately above the collector was only open over YC-TRAP3.

In the hardwood–pine mixed forest, the degree of CC was relatively homogeneous regardless of α at the sites of HP-TRAP1 and HP-TRAP2. For HP-TRAP3, CC increased as α decreased, and we confirmed that the collector was immediately covered by canopy. In all forests, CC at the

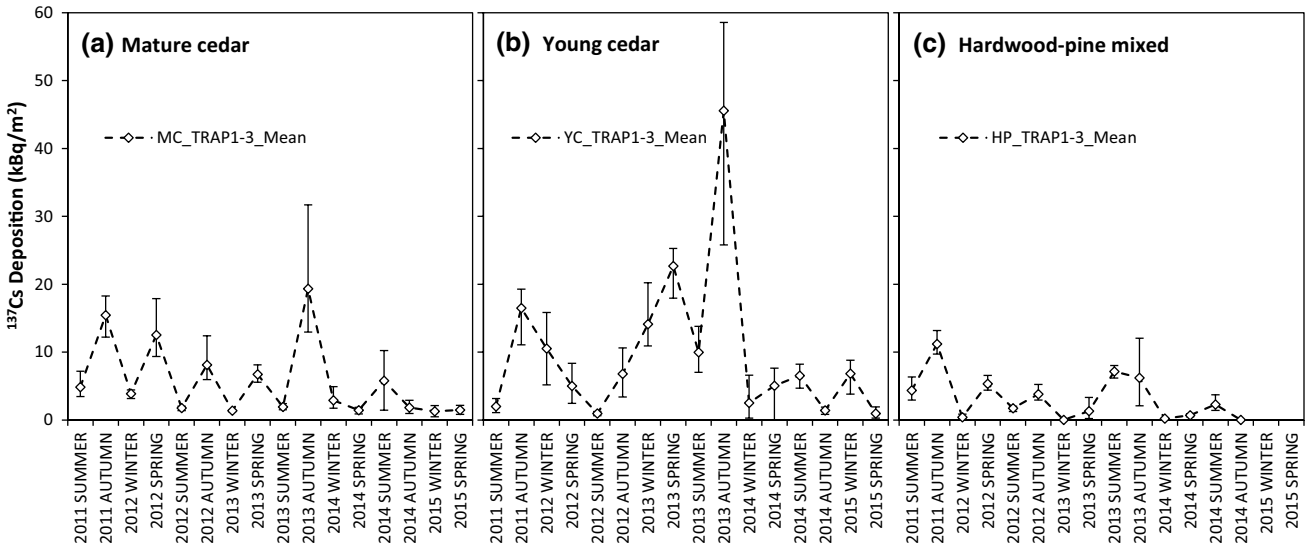

Fig. 5 Seasonally aggregated ^{137}Cs deposition in litterfall samples showing the average value of the three collectors in the **a** mature cedar, **b** young cedar, and **c** hardwood–pine mixed forest types. Error bars indicate the maximum and minimum values. In the hardwood–pine mixed forest, hardwood and pine were measured separately through September 2012, but are displayed as the total concentration for each collector; the average value for each experimental plot is shown

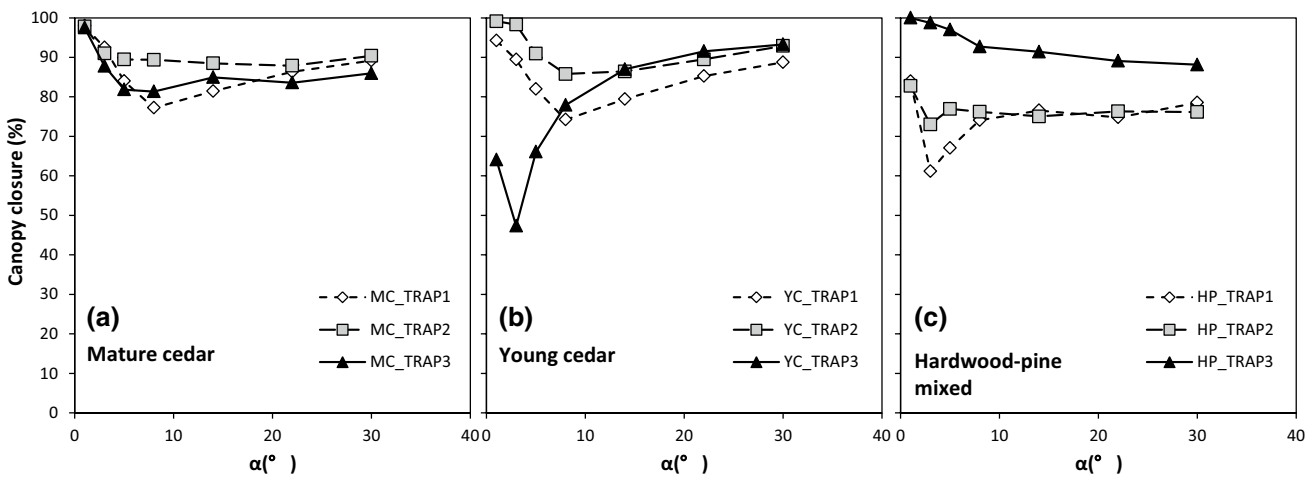

Fig. 6 Angle of view (α) and CC for each collector in the **a** mature cedar, **b** young cedar, and **c** hardwood–pine mixed forest experimental plots. The hemispherical photographs were taken on June 22, 2015

sites of the collectors became uniform (i.e., homogenous) as α increased.

Discussion

Litterfall amount by season

The maximum litterfall amounts were recorded in the mature cedar forest (MC-TRAP1 and MC-TRAP3), young cedar forest (YC-TRAP1 and YC-TRAP2, YC-TRAP3), and hardwood–pine mixed forest (HP-TRAP1 and HP-TRAP2) in autumn 2013 (September 18 to December 16, 2013) (Fig. 3). The causal influence of weather on this pattern was examined based on meteorological observations from Fukushima City provided by the Japan Meteorological Agency [40]. A total of 118 mm of rain fell over 24 h from October 15 to 16, 2013, representing an extreme level of precipitation; 40–50 mm over 24 h is considered a large amount for this time of year and the average monthly precipitation is 257 mm. The precipitation was caused by a large and strong typhoon, Typhoon 26, which approached Fukushima

on the morning of October 16, recording maximum winds of 12.4 m/s and maximum instantaneous winds of 24.8 m/s on that day. Both this continuous precipitation and maximum wind speed were notably high during this fall 2013 sampling period. On the other hand, another 167 mm of continuous rainfall was recorded from September 20 to 21 2011, but little litterfall was found in the collectors over the three subsequent days. These results suggest that the large amount of litterfall observed in autumn 2013 was caused by both the heavy rainfall and strong winds associated with Typhoon No. 26, which approached the area on October 15 to 16. Because such rainfall and strong winds events associated with Typhoon occur more frequently in autumn than in summer, the litterfall amount tended to be higher in autumn than in summer. In December 2013, 22 cm of snowfall was recorded in Fukushima City. This was comparable to the monthly snowfall totals of 14–22 cm for December 2010, 2011, and 2012. Those years lacked noticeable snowfall, and litterfall was therefore unlikely to have been affected by snowfall [40].

In the mature cedar forest, the litterfall amount was high in autumn and spring, with little collected in summer and winter; however, this seasonal pattern was less obvious in the young cedar forest. The reason for this pattern was that the litterfall amounts collected during spring 2012 (March 8 to June 22, 2012) and autumn 2012 (September 21 to December 29, 2012) were markedly higher than during, versus immediately before and after, the sampling periods in the mature cedar forest. Meanwhile, in the young cedar forest, such increases were not observed, with similar amounts collected up to the middle of summer and winter.

^{137}Cs deposition by season

^{137}Cs deposition was generally higher in autumn and spring and lower in summer and winter (Fig. 5). In autumn 2013, ^{137}Cs deposition increased markedly in both cedar forests. The seasonal change in ^{137}Cs deposition was roughly in agreement with the seasonal change in litterfall amount; therefore, the trend in ^{137}Cs deposition strongly reflected that of litterfall.

^{137}Cs deposition gradually declined over the study period, whereas the litterfall amount did not show a decreasing trend. To better understand the changes in litterfall amount by season, and the accompanying changes in ^{137}Cs deposition, we summed the autumn and spring values and divided that value by the sum of summer and winter values to obtain a seasonal index (Table 2).

In the cedar forests, seasonal index values were larger than 1 (1.72–8.55) for the 3 years examined, except from summer 2014 to spring 2015. Autumn and spring tended to have higher index values than summer and winter. Even in the hardwood–pine mixed forest, autumn and spring values tended to be higher, although the values were slightly below 1 in the third year. The degree of variation was generally greater for litterfall amount than ^{137}Cs deposition, with considerable variations seen in many cases.

Relationship between litterfall amount, ^{137}Cs concentration, and ^{137}Cs deposition

The total litterfall amount collected during the survey period, and the average ^{137}Cs concentration of litterfall, were recorded for each collector and each forest stand. In collectors with large litterfall amounts, there was a decreasing trend in ^{137}Cs concentration over time in all three forest stands (Fig. 7a). The ratio of the maximum to the minimum value for each collector was 1.38 for litterfall amount and 1.12 for ^{137}Cs concentration in the mature cedar forest, with corresponding values of 1.53 and 1.22 in the young cedar forest.

The total amount of ^{137}Cs deposition divided by the total litterfall amount in each collector throughout the study period was used to determine the ^{137}Cs concentration, with these two parameters being positively correlated in both the mature and young cedar forests (Fig. 7b). The ratio of the maximum to the minimum value for each collector was 1.38 for litterfall amount and 1.23 for ^{137}Cs deposition in the mature cedar forest, with corresponding values of 1.53 and 1.34 for the young cedar forest. As with the ^{137}Cs concentration, the variation in ^{137}Cs deposition did not increase as sharply as the variation in litterfall amount. Moreover, the variation in ^{137}Cs deposition in the mature cedar forest

Table 2 Seasonal index representing the sum of autumn and spring ^{137}Cs deposition values divided by the sum of summer and winter values

Seasonal index	LF dry weight (g/m^2)			^{137}Cs deposition (kBq/m^2)		
The value of sum of autumn and spring divided by sum of summer and winter	Mature cedar	Young cedar	Hardwood-pine mixed	Mature cedar	Young cedar	Hardwood-pine mixed
2011AUT-2012SPR/2011SMR-2012WTR	8.55	2.73	7.75	3.21	1.72	3.46
2012AUT-2013SPR/2012SMR-2013WTR	7.53	2.27	5.75	4.74	1.96	2.93
2013AUT-2014SPR/2013SMR-2014WTR	4.26	3.22	1.70	4.29	4.05	0.94
2014AUT-2015SPR/2014SMR-2015WTR	0.63	0.18	0.06	0.47	0.18	0.01

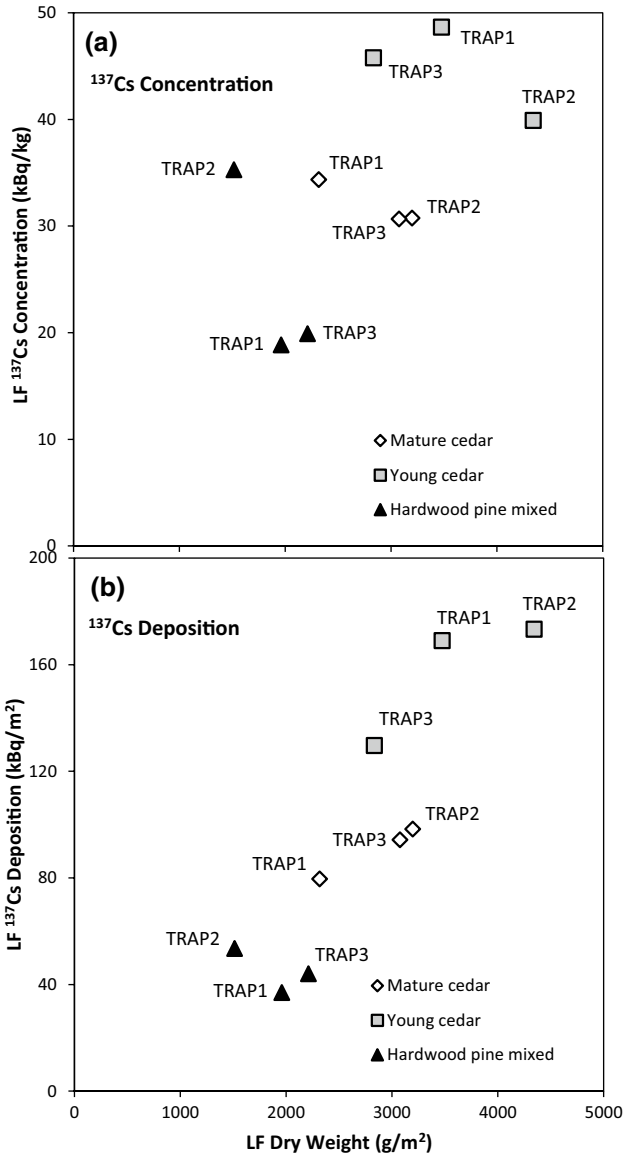

Fig. 7 **a** Total litterfall amount collected in each experimental plot from July 3, 2011, to June 22, 2015, and the average ^{137}Cs concentration for each litterfall collector. The average ^{137}Cs concentration was obtained by dividing total ^{137}Cs deposition over the entire study period by total litterfall over the same period. **b** Total litterfall collected in each experimental plot from July 3, 2011, to June 22, 2015, and total ^{137}Cs deposition for each litterfall collector

was smaller than that in the young cedar forest. In the hardwood–pine mixed forest, the ratio of the maximum to the minimum value for each collector was 1.46 for litterfall amount and 1.45 for ^{137}Cs deposition, showing almost identical levels of variation.

Smaller variations were recorded in ^{137}Cs concentration and ^{137}Cs deposition than in litterfall amount in the two cedar forests. The degree of variation in ^{137}Cs deposition was 10% less than that of litterfall amount in the mature cedar forest, with corresponding values of 13% for the young cedar forest and 1% for the hardwood–pine mixed forest (Fig. 7b). These differences were smaller than the reported throughfall from the canopy to the forest floor in the same experimental plots, which accounted for 34%, 34%, and 16% of ^{137}Cs deposition in the three forest types, respectively [10]. The deposited ^{137}Cs that accumulated over a long period of time was homogeneous, with small variations according to the exact location in the forest and rainfall amount;. In contrast, variations due to litterfall were relatively large and heterogeneous. This discrepancy may be caused by differences in the conditions after ^{137}Cs deposition had ceased compared to when the deposition was ongoing.

Variation by forest location was smaller for ^{137}Cs deposition than for litterfall amount, likely due to the inverse correlation between ^{137}Cs concentration and litterfall amount being generally weak in collectors with high ^{137}Cs deposition amounts; meanwhile, ^{137}Cs concentrations were high in collectors with low ^{137}Cs deposition amounts. In the cedar forests, the ^{137}Cs concentration tended to be lower in collectors with larger amounts of litterfall (Fig. 7a). Overall, larger litterfall amounts were observed in collectors with more branches and leaves; such collectors also had lower ^{137}Cs concentrations.

In addition to the litterfall amount, spatial variations in ^{137}Cs concentration and ^{137}Cs deposition were greater in the young cedar forest than in the mature cedar forest. This was probably due to the greater height of trees and higher position of branches in the mature cedar forest, along with the more homogenous canopy (Fig. 7a, b).

In the hardwood–pine mixed forest, litterfall amount and ^{137}Cs deposition were not positively correlated, as observed in the cedar forests (Fig. 7b). This lack of correlation was because the ^{137}Cs concentration was excessively high in HP-TRAP2, which had the smallest litterfall amount among the three collectors (Fig. 7a). The ^{137}Cs concentration was higher in HP-TRAP2 than in the other two collectors due to the collection of branches from a tree with high ^{137}Cs concentrations. To identify trees with high ^{137}Cs concentrations, it is necessary to first determine the species at the sampling sites of all three collectors, and then measure the weight and ^{137}Cs concentration associated with each tree species and site. Determination of litterfall amounts and variations in ^{137}Cs concentrations at this scale in hardwood–pine mixed forests composed of various tree species should be conducted in a future study.

Factors controlling differences in litterfall amount and ^{137}Cs concentration among litterfall collectors and seasons

Overall, a weak inverse correlation was observed between litterfall amount and ^{137}Cs concentration (Fig. 7a). In addition to the fluctuations in litterfall amount (Fig. 3) and ^{137}Cs

concentration (Fig. 4) over time, variations were also noted among collectors. To examine the factors controlling these variations in greater detail, we analyzed the magnitude of the litterfall amount and obtained collector-specific values, and then determined the degree of dispersion among these values for all collectors. To normalize the degree of variation, coefficients of variation (CVs) for litterfall amount and ^{137}Cs concentration at each sampling time were calculated by subtracting the minimum value from the maximum value for each litterfall collector and dividing the result by the average value.

In both the mature and young cedar forests, the CVs of both litterfall amount and ^{137}Cs concentration were often less than 1, indicating no large variations (Fig. 8a–c). However, the CVs of ^{137}Cs concentration were 1–1.5 in summer 2011, summer 2013, summer 2014, and spring 2015 in the

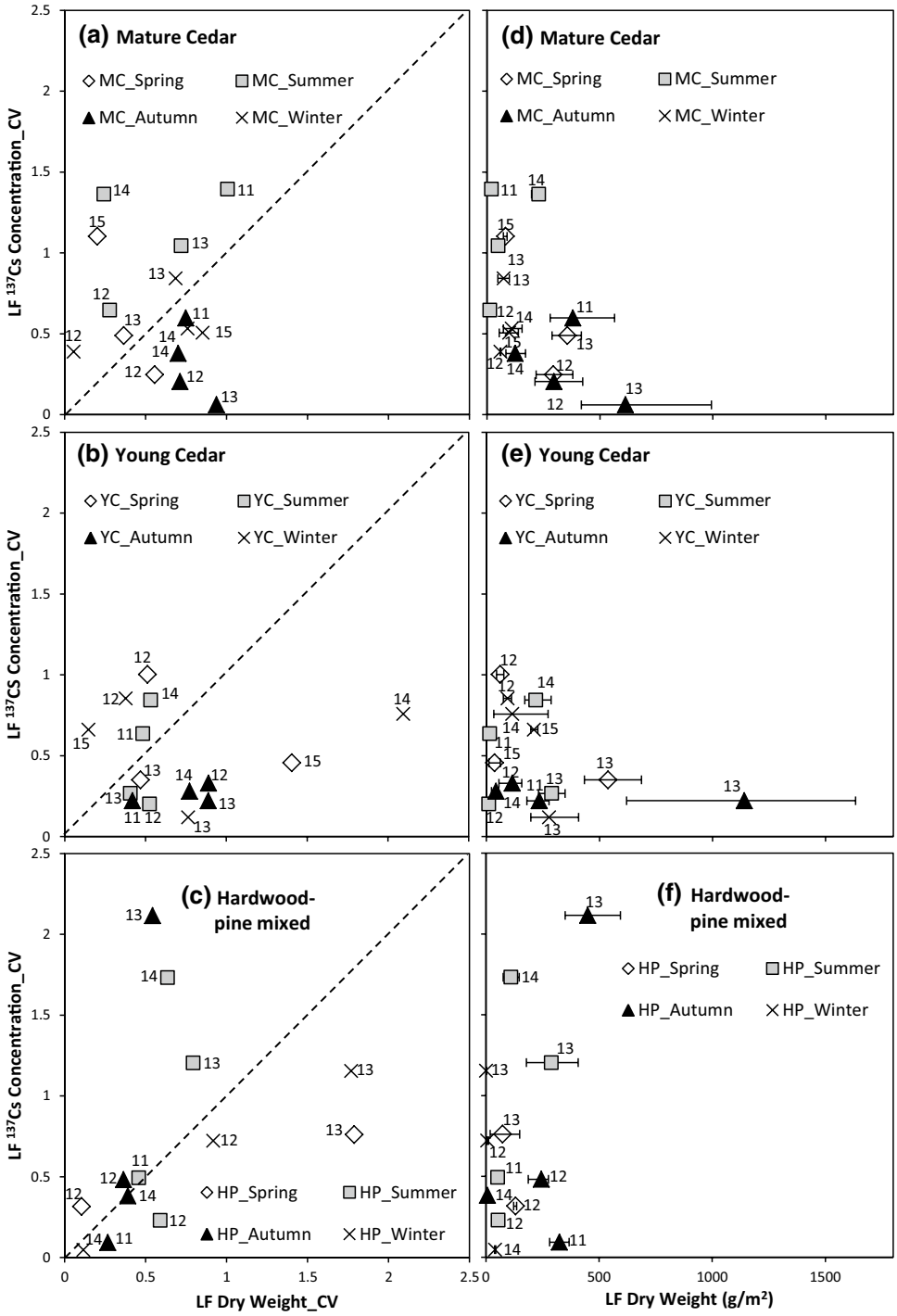

Fig. 8 Relationship between litterfall amount and ^{137}Cs concentration by season. **a–c** For both the litterfall amount and ^{137}Cs concentration, the coefficient of variation (CV) was obtained by dividing the difference between the maximum and minimum values by the average value. This measure was used to assess the variation in values for each collector. The average ^{137}Cs concentration was calculated based on the weight of the sample. **d–f** Litterfall amount shown as the average, maximum and minimum values, and ^{137}Cs concentration represented by the CV. The number on each symbol indicates the year

mature cedar forest, where the variation was slightly larger than in the young cedar forest. In the young cedar forest, the CVs of litterfall amount were 1.4–2.1 in winter 2014 and spring 2015, showing large variations. The CV of ^{137}Cs concentration was relatively high in summer and relatively low in autumn in the mature cedar forest. In both cases, the CV of ^{137}Cs concentration tended to be smaller when the CV of litterfall amount was large. In the hardwood–pine mixed forest, the CV of the ^{137}Cs concentration tended to be larger than those in cedar forests; the CV of the ^{137}Cs concentration was three times larger and the CV of litterfall amount was twice as large as for cedar forests. In both cases, the CV exceeded 1 and the variations tended to be larger than those recorded in cedar forests.

In Fig. 8d–f, the horizontal axis represents the average, maximum, and minimum litterfall amount, with error bars, according to the CV of the ^{137}Cs concentration. In the mature and young cedar forests, the variance in samples with large litterfall amounts was large. When the litterfall amount and its variation were large, the CV of the ^{137}Cs concentration was small, and when the CV of the ^{137}Cs concentration was large and variable, the litterfall amount and its variation tended to be small. In contrast to the cedar forests, the litterfall amount and its variation in the hardwood–pine mixed forest varied independently of the CV of the ^{137}Cs concentration.

A weak inverse correlation was found between the variances of litterfall amount and ^{137}Cs concentration, especially in the cedar forests, but this relationship was not apparent for ^{137}Cs deposition. The ^{137}Cs concentration varied less when the litterfall amount was large, as the total amount of sample increased, resulting in smoothing of the values. When the litterfall amount was small, the ^{137}Cs concentration varied substantially if a particular material from a specific tree (e.g., cedar bark or seeds) containing a high ^{137}Cs concentration was collected. The weak inverse correlation between litterfall amount and ^{137}Cs concentration was one reason why the variation in ^{137}Cs deposition did not increase with increasing variation in litterfall amount. These results lead to the homogeneity of ^{137}Cs distribution in the forest floor as a result.

Relationship between *α* and CC

In the mature cedar forest, young cedar forest, and hardwood–pine mixed forest, the CC value increased (i.e., the canopy closed) and decreased (i.e., the canopy opened) when α was small and large (generally $\alpha > 10°$), respectively. After averaging, CC showed a tendency toward closure (Fig. 6). Litterfall collectors in locations with large CC values recorded large litterfall amounts and high ^{137}Cs deposition. The range of α over which a significant relationship existed between CC and litterfall amount/^{137}Cs deposition varied by individual tree species. For example, litterfall is affected by canopy height and wind [41], and the timing and scattering distance of fallen leaves differ between cedar and hardwood [42]. Moreover, patterns of leaf fall and scattering distance may differ among tree species due to differences in the shape, size, and weight of branches and leaves among species. For example, cedars have large, heavy leaves that tend to fall straight down from the canopy. Thus, in cedar forests, when α is relatively small ($\leq 10°$), the amount of litterfall would be associated with CC. Conversely, in the hardwood–pine mixed forest, the leaves disperse widely as they fall from the canopy; therefore, the magnitude of CC and litterfall amount would not be associated, regardless of the value of α.

To further investigate this hypothesis, the associations of CC, litterfall amount, and ^{137}Cs deposition were analyzed for each forest (Table 3; Fig. 9).

In the young cedar forest, we found positive correlations among CC, litterfall amount, and ^{137}Cs deposition when α was small ($\leq 5°$). Meanwhile, in the mature cedar forest, only positive correlations were observed when α was in the range of 8° to 14°, and α values were slightly larger than those in the young cedar forest. Such differences may be caused by variations in tree height, given the greater height of the lowest branch in mature cedars compared to young cedars. In the hardwood–pine mixed forest, CC showed associations with litterfall amount and ^{137}Cs deposition at $\alpha = 1°$, 14°, and 30°. Interestingly, litterfall amount and ^{137}Cs deposition showed the same tendencies in relation to CC in both cedar forests (Fig. 9a, b, d, e), despite the variation of ^{137}Cs deposition being lower than that of litterfall amount. Therefore, our hypothesis could be accepted for both the cedar and hardwood–pine mixed forests, and the range of α values over which the correlation between CC and litterfall amount or ^{137}Cs deposition was significant could be determined from the deposition and scattering pattern of fallen leaves.

For CC values, we used survey results obtained in early summer. Although the degree of openness of the canopy is nearly constant regardless of season in cedar forests, hardwood–pine mixed forests exhibit differences in openness among autumn, winter, spring, and summer [43]. Moreover, in hardwood–pine mixed forests, CC and litterfall amount show differing characteristics among the sprouting, full-leaf, and fallen leaf stages. Because the range of α values over which significant associations can occur is not constant, it is necessary to evaluate CC, litterfall amount, and ^{137}Cs deposition in different seasons. Future studies should determine the amount of ^{137}Cs deposition based on forest status when evaluating the degree of ^{137}Cs transfer from forests to the surrounding environment.

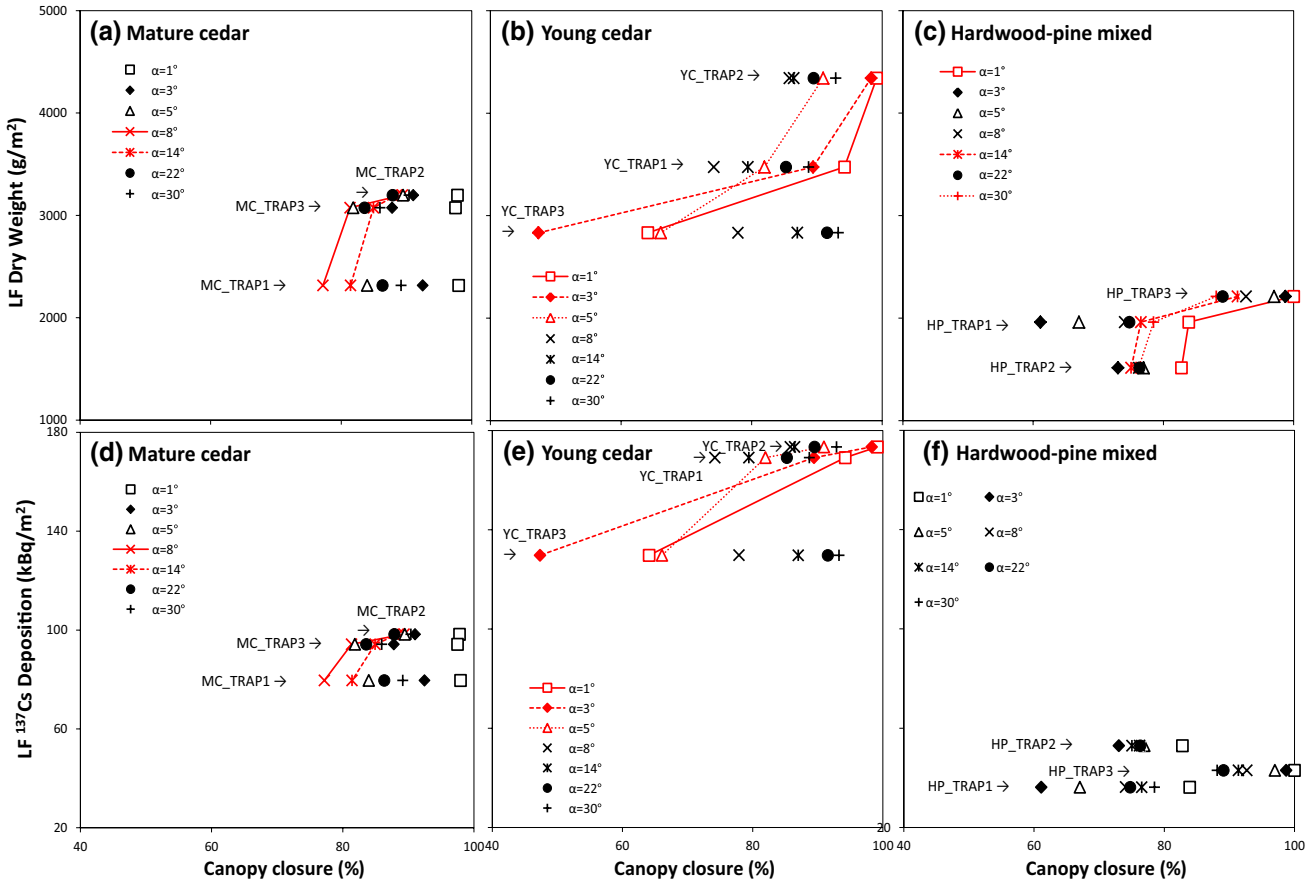

Fig. 9 Scatter diagrams showing CC against the angle of view (α) obtained from hemispherical photographs taken for each litterfall collector in the **a**, **d** mature cedar, **b**, **e** young cedar, and **c**, **f** hardwood–pine experimental plots. The litterfall mass **a**–**c** and **d**–**f** ^{137}Cs deposition are shown for each case. Hemispherical photographs were taken on June 22, 2015. Total litterfall amount is shown from July 3, 2011, to June 22, 2015. In **a** to **c**, when the symbol α is shown in red, CC and litterfall amount were positively correlated

Conclusions

In this study, we characterized the spatial and seasonal changes in litterfall amount, ^{137}Cs concentration in litterfall, and ^{137}Cs deposition from the period immediately after the FDNPP accident in 2011 until 2015. We studied three forest types (mature cedar, young cedar, and hardwood–pine mixed forests) with differing characteristics. Focusing on the relationships among CC, litterfall amount, and ^{137}Cs deposition, we examined the range of α values over which the extent of CC affected the litterfall amount and ^{137}Cs deposition for each forest type. In general, the variation in litterfall amount was greater than that of ^{137}Cs deposition. In cedar forests, the degree of variation in ^{137}Cs deposition was 10–13% lower than that of litterfall amount. Moreover, a weak inverse correlation was found between the variation in litterfall amount and ^{137}Cs concentration in cedar forests. The ^{137}Cs concentration may fluctuate more when smaller amounts of litterfall are collected, as samples are more likely to be affected by individual tree parts (e.g., bark and seeds) with high ^{137}Cs concentrations. Moreover, the range of α values over which CC was most strongly correlated with litterfall amount or ^{137}Cs deposition was smallest in the young cedar forest and greatest in the hardwood–pine mixed forest due to differing leaf deposition and scattering patterns; heavy cedar leaves fall straight to the ground while light hardwood leaves are scattered widely. In addition, differences among tree species in terms of height, likely cause differences in the falling patterns of branches and leaves from the tree canopy. Overall, the results of this study show that temporal and spatial changes in litterfall amount and ^{137}Cs deposition from the tree canopy are highly heterogeneous in cedar forests, but show a homogeneous tendency in hardwood–pine mixed forest. This pattern is due to the difference in falling patterns of leaves among tree species, causing different patterns of secondary contamination.

Table 3 Canopy closure values by angle of view (α) obtained from hemispherical photographs taken at the site of each litterfall collector

$\alpha(°)=$	2015/6/22							2011/7/3–2015/6/22	
	Canopy closure (%)							Litterfall dry-weight (g/m^2)	^{137}Cs deposition (kBq/m^2)
	30	22	14	8	5	3	1		
(a) Mature cedar									
MC_TRAP1	89.18	86.33	**81.43**	**77.20**	83.98	92.48	97.98	**2317.66**	79.6
MC_TRAP2	90.40	87.88	**88.48**	**89.38**	89.48	91.03	97.80	**3197.45**	98.3
MC_TRAP3	85.95	83.58	**84.95**	**81.35**	81.85	87.80	97.50	**3076.09**	94.3
(b) Young cedar									
YC_TRAP1	88.73	85.25	79.43	74.20	**81.98**	**89.43**	**94.23**	**3474.58**	169.1
YC_TRAP2	92.90	89.50	86.45	85.80	**90.95**	**98.30**	**99.15**	**4342.60**	173.4
YC_TRAP3	93.28	91.55	87.00	77.90	**66.08**	**47.35**	**64.10**	**2833.08**	129.7
(c) Hardwood pine mixed									
HP_TRAP1	**78.50**	74.78	**76.55**	74.08	67.05	61.13	**83.90**	**1960.54**	37.0
HP_TRAP2	**76.18**	76.33	**75.05**	76.23	76.95	73.03	**82.78**	**1514.55**	53.4
HP_TRAP3	**88.13**	89.10	**91.40**	92.70	97.00	98.73	**100.00**	**2211.44**	44.0

The litterfall amount and radiocesium (^{137}Cs) deposition through litterfall are shown. The hemispherical photograph was taken on June 22, 2015. The litterfall amount and ^{137}Cs deposition are shown as cumulative values from July 3, 2011, to June 22, 2015. In the mature and young cedar forests, litterfall amount and ^{137}Cs deposition were positively correlated. For the α values outlined in bold, CC and litterfall amount were positively correlated

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