

## 論文 (Original article)

# Predicting annual trends in leaf replacement and $^{137}\text{Cs}$ concentrations in *Cryptomeria japonica* var. *japonica* plantations with radioactive contamination from the Fukushima Daiichi Nuclear Power Station accident

Yoshiyuki KIYONO<sup>1)\*</sup> and Akio AKAMA<sup>2)</sup>

### Abstract

*Sugi* (*Cryptomeria japonica* var. *japonica*) plantations are the predominant evergreen coniferous forest ecosystems in Japan. *Sugi* is a unique evergreen tree with no abscission layer in the leaf. However, the lifespan of *sugi* leaves is unknown. In this study, we described *sugi* leaves, investigated the age structures of living and newly deceased leaves, and estimated the lifespan and death process of the leaves. The lifespan of *sugi* leaves was 1–8 years, with an estimated mean of 4.3–5.3 years. Then, we modeled patterns of leaf replacement and cesium-137 ( $^{137}\text{Cs}$ ) concentrations and estimated the potential  $^{137}\text{Cs}$  supply to the forest floor through leaf shedding based on  $^{137}\text{Cs}$  concentration data from leaves of various ages sampled after the Fukushima Daiichi Nuclear Power Station (FDNPS) accident. Around 90% of leaves that had sprouted before March 11, 2011 (*ante*-3.11 leaves) were predicted to die within 4 years of the FDNPS accident. Moreover, ~90% of the  $^{137}\text{Cs}$  in *ante*-3.11 leaves was predicted to be removed within 3 years of the FDNPS accident. The predicted trends of stand leaf  $^{137}\text{Cs}$  concentrations were verified with  $^{137}\text{Cs}$  concentrations measured in four permanent sample plots in Fukushima Prefecture. This study revealed that  $^{137}\text{Cs}$  translocated from the canopy to the forest floor at a faster rate than the *ante*-3.11 leaves that had been directly contaminated by the FDNPS accident. Including *sugi* leaf mortality and replacement in future models will allow for more accurate predictions of the fate and persistence of radiocesium in *sugi* forests affected by the FDNPS accident.

**Key words** : death process of leaves, evergreen conifer, fallout, leaf lifespan, leaf replacement, simulation, *sugi*

### 1. Introduction

Radioactive material deposited onto forests after the Tokyo Electric Power Company (TEPCO) Fukushima Daiichi Nuclear Power Plant Station (FDNPS) accident has moved through these ecosystems following the flow of forest materials. Radiocesium fallout is initially delivered to forest ecosystems predominantly on plant surfaces, including tree canopies, and gradually moves towards the soil (Hisadome et al. 2013, Kajimoto et al. 2015). Various factors, such as plant species, climate, and disturbance regime, can influence radiocesium movement through ecosystems. Therefore, it is important to monitor and model radiocesium dynamics (Kato et al. 2015) to forecast future contamination of forests affected by the FDNPS accident.

One month after the FDNPS accident, >80% of the total deposited cesium-137 ( $^{137}\text{Cs}$ ) from the fallout had been deposited onto evergreen coniferous forest canopies (Kato and Onda 2014).  $^{137}\text{Cs}$  deposition on forest floors occurred mainly via throughfall from July to September 2011, and the subsequent transfer of  $^{137}\text{Cs}$  from the canopy to the forest floor occurred mainly through litterfall (Kato et al. 2015). Moreover, litterfall was determined to have contributed 45% of  $^{137}\text{Cs}$  transfer from forest canopies to forest floors within

~5 months of the FDNPS accident, which was comparable to the contribution of throughfall (53%) in an evergreen hinoki cypress (*Chamaecyparis obtusa*) forest (Teramaga et al. 2014). In forests 26–134 km west or southwest of the FDNPS, in August to September 2011, 5–6 months after the FDNPS accident, 23–43% of stand total radiocesium ( $^{134}\text{Cs}$  +  $^{137}\text{Cs}$ ) was distributed on the crowns of the evergreen conifer *sugi* (*Cryptomeria japonica* var. *japonica*), whereas more than half was distributed in the litter and surface soil (Forestry Agency 2011, 2014b).

*Sugi* forests comprised the largest area of forested land in Japan in 2012 (18%), and occupied 19% of the total forested land in Fukushima Prefecture in 2012 (Forestry Agency 2012). However, no data are available on *sugi* leaf death (Kiyono et al. 2012). *Sugi* is a unique evergreen tree, as its leaves have no abscission layer. It bears needle-like leaves 2–15 mm long and 0.5–2 mm wide at the base, arranged in spirals around young stems and branches. In this study, we defined *sugi* leaves as young stems and branches covered with living needle-like leaves. In this definition, some *sugi* leaves grow into the stem or branches following the development of the xylem. Relatively old leaves may have a greater mass per unit leaf area than young leaves

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1) General Affairs Department, Forestry and Forest Products Research Institute (FFPRI), Japan

2) Research Planning and Coordination Department, FFPRI, Japan

\* Research Expert, FFPRI, 1 Matsunosato, Tsukuba, Ibaraki, 305-8687 Japan, e-mail: kiono@ffpri.affrc.go.jp

with the same leaf area. The lifespan of *sugi* leaves is unknown. Most studies that have used destructive biomass sampling techniques to estimate primary productivity have assumed that *sugi* leaves have a lifespan of 4 years (e.g., Cannel 1982). In general, leaf lifespan differs depending on tree species, tree age, and environmental conditions (Hatiya et al. 1966, Reich et al. 1992). However, there is no information available on *sugi* leaf death and the transfer process of radiocesium via leaf shedding from the canopy to the forest floor. Clarifying leaf replacement patterns (death and birth) in *sugi* will reduce uncertainties regarding radiocesium fate and persistence predictions in *sugi* forests affected by the FDNPS accident.

First, we examined the age structures of living and newly deceased leaves to estimate the lifespan of *sugi* leaves. In addition, we investigated  $^{137}\text{Cs}$  concentrations in various age classes of *sugi* leaves sampled between 1 year 8 months and 3 years 8 months after the FDNPS accident. Using these data, we modeled the dynamics of *sugi* leaves and  $^{137}\text{Cs}$  concentrations in plantation canopies using leaf age structure as a parameter to estimate the persistence of  $^{137}\text{Cs}$  in canopies. We verified the *sugi* leaf  $^{137}\text{Cs}$  concentration predictions in the canopy using data independent from ours obtained from four *sugi* forest permanent sample plots (PSPs) in Fukushima Prefecture.

## 2. Materials and methods

### 2.1 *Sugi* leaf age structure determination

**Leaf age structure method:** We measured the diameter at breast height (DBH) and tree height of all *sugi* in the Kunugidaira PSP (400 m<sup>2</sup>), a 33-year-old *sugi* plantation in Ishioka City, Ibaraki Prefecture (Table 1). In March 1983, seven trees near the PSP were felled for destructive biomass sampling. They covered the entire area and corresponded to the frequency distribution of tree DBH classes in the plot. In addition, two trees were large (Nos. 1 and 2), four trees were medium-sized (Nos. 3–6), and one tree was small (No. 7) (Table 2). The crown of each tree was divided vertically into 1-m sections. Branch diameter was measured at the branch base for all branches primarily derived from stems (primary branches). In each layer, four primary branches with leaves were selected as samples (primary branch samples). Leaves were separated according to leaf age class (e.g., current-year (0 years old), 1 year old, 2 years old, etc.). Green stems and branches were included as leaves based on the description above. However, the stems and main branches of primary branches were categorized as stems and branches, respectively, even though they had green tips. With the approach of winter, *sugi* leaves usually die as a module composed of a small branch with leaves (Tange et al. 1989, Kaneko et al. 1997). Sometimes,

Table 1. Summary of the research plots and *Cryptomeria japonica* var. *japonica* trees used in this study

| Plot type             | Plot or site name | Administrative district   | Location                          | Altitude, m | Distance and direction from the FDNPS     | Tree age in 2011, y | Stand structure <sup>2</sup>  | Number of sample trees | Time of measurement  | Measurement items  | Remarks      |
|-----------------------|-------------------|---|-----------------------------------|-------------|---|---------------------|---|------------------------|----------------------|--|--------------|
| Permanent sample plot | Kunugidaira PSP   | Kunugidaira National Forest, former Kasama District Forest Office, presently Ibaraki District Forest Office, Ishioka City | 36.17 N–140.17 E                  | 298         | 140 km Southwest                          | 61                  | DBH 19.6 ± 6.3, H 15.3 ± 2.2, $\rho$ 1,625 ha <sup>-1</sup> in 1983 | 7                      | Mar 1983             | DBH, tree leaf mass and area, and leaf age structure         |              |
| Temporary plot        | Chiyoda plot      | Chiyoda Experimental Station of FFPRI, Kasumigaura City   | 36.18 N–140.22 E                  | 41          | 140 km Southwest                          | 19                  | DBH 22.0 ± 3.5, H 10.5 ± 2.2, $\rho$ 492 ha <sup>-1</sup> in 2011   | 20                     | Nov 2011–Jan 2012    | Leaf age structure of withered small branch and leaf modules |              |
| Temporary plot        | Tsukuba plot      | The First Arboretum of FFPRI, Tsukuba City  | 36.01 N–140.13 E                  | 25          | 160 km Southwest                          | ~40                 | DBH 34.0 ± 8.0, $\rho$ 716 ha <sup>-1</sup> in 2011                 | 20                     | Nov 2011–Jan 2012    | Leaf age structure of withered small branch and leaf modules |              |
| Temporary plot        | Fukushima 23      | National forests and private forests, Fukushima Prefecture  | 36.94–37.73 N and 140.15–141.01 E | 24–678      | Within 78 km North-northwest to southwest | -                   | -   | 2–3 at each site       | Nov 2012, 2013, 2014 | Leaf age structure   | 23 locations |
| Permanent sample plot | Kawauchi PSP      | Okayama National Forest of Iwaki District Forestry Office, Kawauchi Village   | 37.29 N–140.80 E                  | 665         | 26 km Southwest                           | 43                  | DBH 18.9, H 14.3, $\rho$ 975 ha <sup>-1</sup> in 2011               | 3                      | Aug 2013             | Leaf age structure   |              |
| Permanent sample plot | Kamikawauchi PSP  | Kawauchi Village forest, Togenomori, Kamikawauchi, Kawauchi Village   | 37.38 N–140.72 E                  | 693         | 27 km West                                | 57                  | DBH 30.9, H 19.2, $\rho$ 733 ha <sup>-1</sup> in 2011               | 3                      | Aug 2013             | Leaf age structure   |              |
| Permanent sample plot | Otama PSP         | Maegatahokaichi National Forest of Fukushima District Forest Office, Otama Village  | 37.57 N–140.31 E                  | 734         | 66 km West                                | 42                  | DBH 24.8, H 17.8, $\rho$ 1,117 ha <sup>-1</sup> in 2011             | 3                      | Aug 2013             | Leaf age structure   |              |
| Permanent sample plot | Tadami PSP        | Tabanematsuyama National Forest of Minamiaizu Sub-district Forest Office in Aizu District Forest Office, Tadami Town      | 37.32 N–139.52 E                  | 717         | 134 km Southwest                          | 38 <sup>1</sup>     | DBH 19.9, H 14.3, $\rho$ 1,105 ha <sup>-1</sup> in 2011             |                        |                      |  |              |

<sup>1</sup> Kajimoto et al. (2014) revised. <sup>2</sup> DBH, cm, mean ± SD; H, m, mean ± SD;  $\rho$ , tree density, ha<sup>-1</sup> (the data source of the Kawauchi, Kamikawauchi, Otama, and Tadami PSPs is Kajimoto et al. 2014).

there are fragmented green leaves in withered branch and leaf modules, but these were not included as living leaves because they were expected to die soon. We collected leaf subsamples to calculate the specific leaf area (SLA). Since *sugi* exhibits spiral phyllotaxis, we flattened and ironed the spiral leaves into an alternate pattern (Fig. 1). The leaf samples were used to measure one-sided leaf area with an automated area meter (AAM-7; Hayashi Denko Co. Ltd., Tokyo, Japan). The same samples were oven-dried at 80°C for at least 72 h to determine the dry mass. In each layer, the leaf area and mass were estimated for every leaf age class using the sampled primary branch results and the ratio of branch base cross-sectional area of all primary branches to that of the sampled primary branches in the layer. Then, the sum of each tree layer of the seven sampled trees was calculated. Following the methods of Satoo (1973), the ratio of stem cross-sectional area at breast height (*ba*) of the total trees in the plot to that of the seven sampled trees ((sum of *ba* of all trees in a plot)/(sum of *ba* of sampled trees)) was used to estimate stand leaf area and leaf mass for every leaf age class. Finally, the results of the plot were converted to



Fig. 1. A *Cryptomeria japonica* var. *japonica* leaf with the spiral phyllotaxis flattened into an alternate leaf pattern

per unit land area.

We assumed that the leaf area distribution of each age class in a stand was annually stable and considered the difference between two successive years to represent the quantity of deceased leaves per year. Based on these assumptions, we estimated the age structure of dead leaves produced per year. In this estimation, we also assumed that current-year leaves arose from 1-year-old leaves, not 2-year-old or older leaves. In reality, dormant buds of primary branches in the lower crown layer sometimes become active. While the mass of current-year leaves arising from older leaves is non-negligible when the main stem is broken, it likely to be insignificant in undamaged trees. Therefore, we assumed that the chronological order of

Table 2. Description of destructively sampled trees in the 33-year-old *Cryptomeria japonica* var. *japonica* forest in the Kunugidaira PSP

| No. | DBH<br>cm | Total<br>height<br>m | Hb <sup>1</sup><br>m | Db <sup>2</sup><br>cm | Branch<br>mass<br>kg | Leaf<br>mass<br>kg | Leaf<br>area<br>m <sup>2</sup> |
|-----|-----------|----------------------|----------------------|-----------------------|----------------------|--------------------|--------------------------------|
| 1   | 29.6      | 19.0                 | 9.6                  | 19.0                  | 12.4                 | 23.0               | 92.2                           |
| 2   | 24.9      | 18.6                 | 12.5                 | 13.6                  | 8.6                  | 16.1               | 50.4                           |
| 3   | 21.2      | 17.8                 | 12.0                 | 13.5                  | 5.8                  | 11.1               | 44.5                           |
| 4   | 19.0      | 14.8                 | 9.5                  | 11.1                  | 4.8                  | 8.1                | 27.9                           |
| 5   | 14.9      | 15.9                 | 11.6                 | 7.4                   | 1.5                  | 4.1                | 13.7                           |
| 6   | 14.5      | 13.5                 | 8.9                  | 8.5                   | 1.8                  | 2.9                | 14.1                           |
| 7   | 12.2      | 13.1                 | 8.3                  | 6.5                   | 0.83                 | 1.5                | 8.4                            |

Nos. 1 and 2, large trees; 3–6, medium-sized trees; and 7, small tree. <sup>1</sup> Height of the lowest living branch. <sup>2</sup> Stem diameter below the lowest living branch.

leaf age was preserved within branches.

**Withered branch and leaf module method:** In the period November 2011–January 2012, we sampled two withered small branch and leaf modules from 20 trees in a 19-year-old *sugi* plantation at the Chiyoda Experimental Station of the Forestry and Forest Products Research Institute (FFPRI), Kasumigaura City, Ibaraki Prefecture (Chiyoda plot, Table 1), and 20 trees in a ~40-year-old plantation at the First Arboretum of FFPRI, Tsukuba City, Ibaraki Prefecture (Tsukuba plot, Table 1). The sampled modules were mostly collected from the lower crown layer <4 m above the ground. Each withered small branch and leaf module was collected after determining its leaf age range by comparing it with the leaf age ranges of nearby living branch and leaf modules on the tree. *Sugi* sprouts long needle-like leaves up to 15 mm long from spring to around the summer solstice, after which it sprouts short needle-like leaves (2–4 mm long) until around September (Kiyono, unpublished data of observations of *sugi* trees at the nursery of FFPRI, Tsukuba City in 2007–2009). Therefore, we considered the boundary of *sugi* leaf birth year to be where leaf formation switched from short needle-like leaves to long needle-like leaves toward the tip in the two successive years.

Each sampled module was placed on the ground horizontally with the tip facing outward (Fig. 2) and *sugi* leaves were identified by age, which formed a U-shaped band pattern (yellow lines, Fig. 2). Each leaf area in the sampled module was approximated from the area of the pipe-shaped outline of the leaf (dotted lines in inset, Fig. 2). The ratio of the sum of the projected leaf space areas of each leaf age class to the total area of the module was measured by visual observation and considered to be the age distribution of dead leaves. Using small portion of the Chiyoda plot leaf samples, the area of leaves based on the outlined area did not differ significantly from the one-sided



Fig. 2. A newly deceased small branch and leaf module of *Cryptomeria japonica* var. *japonica*

Leaves of the same age are located within the curved bands denoted by the solid yellow lines. Leaf area was approximated from the area of the pipe-shaped outline of the leaf, as shown by the dotted lines in the inset. The ratio of the sum of the projected outlined area of each leaf age class to total leaves was measured from visual observations. The ratio of the projected outlined areas of 6-, 5-, 4-, and 3-year-old leaves was approximately 4:4:2:0.

leaf area of the flattened leaves (paired *t*-test,  $P = 0.955$ ,  $n = 32$ ). The age distribution of the dead leaves of all sampled modules in a stand was averaged and considered to be the age distribution of newly deceased leaves.

## 2.2 $^{137}\text{Cs}$ concentration measurements in various leaf age classes

The 23 *sugi* forest locations (Table 1, Akama et al. 2013, Forestry Agency 2014a) were distributed at almost regular intervals in areas with various air dose rates (0.10–23.2  $\mu\text{Sv h}^{-1}$  at a height of 1 m in November 2011) in Fukushima Prefecture (hereafter referred to as the Fukushima 23). In November 2012, 2013, and 2014, leaves in branches 0.3–0.5 m long at a height of several meters in direct sun (Akama et al. 2013) were collected from two or three *sugi* trees at each site and divided by age class. The branches contained leaves of varying ages, from those that had sprouted before the FDNPS accident (*ante*-3.11) up to current-year (*post*-3.11, sprouted after the FDNPS accident) leaves. The sampled trees were not consistent among sample years. The sampled trees were considered to be  $\geq 20$  years old, as they had

produced male flowers. In August 2013, leaves in branches 1–2 m long at a height of several to  $>10$  m in the sun were sampled in nine *sugi* trees from the three PSPs, Kawauchi, Kamikawauchi, and Otama (Table 1).

The leaves were divided by age class. *Post*-3.11 leaves were washed with tap water, rinsed with distilled water, and dried by draining and blotting with a paper towel. The *ante*-3.11 and *post*-3.11 leaf samples were weighed to determine their fresh mass, oven-dried at  $75^\circ\text{C}$  for at least 48 h, and then weighed again. The samples were minced with a household blender into pieces  $<3\text{-mm}$ . The processed samples were packed into 100-mL polystyrene containers for gamma-ray spectrometry measurements. The samples were measured at the FFPRI with an HPGe coaxial detector system (GEM20-70, DS-P600 Gamma Studio; Seiko EG&G Co. Ltd., Tokyo, Japan) for at least 1,800 s. The detection limit for the radioactive element measurement was calculated based on three-sigma rule in DS-P600 Gamma Studio (Seiko EG&G Co. Ltd.). Gamma-ray peaks of 604 and 662 keV were used for the  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  measurements, respectively. Calibrations were conducted using a standard gamma-ray source (MX033U8PP; Japan Radioisotope Association, Tokyo, Japan). We corrected the coincidence-sum effect of gamma rays from  $^{134}\text{Cs}$ .  $^{137}\text{Cs}$  values were adjusted to those of September 1, 2013.

## 2.3 Modeling *sugi* leaf and $^{137}\text{Cs}$ dynamics in the canopy

We generated a simple model to predict the potential supply of leaves and accompanying  $^{137}\text{Cs}$  from the canopy to the forest floor via leaf shedding based on leaf replacement and  $^{137}\text{Cs}$  concentrations.

To model leaf replacement, we used the data from the leaf age structure method because it was more detailed than that from the withered branch and leaf module method. To estimate chronosequential changes in the proportion of *ante*-3.11 leaves and *post*-3.11 leaves, the relationship between leaf age class and percentage of one-sided leaf area (*LAR*, leaf area ratio) in each leaf age class was approximated using the Gompertz equation. The results were converted into an equation approximating the relationship between leaf age class and percentage of leaf mass in each leaf age class using the relationship between leaf age class and mean *SLA* ( $\text{cm}^2 \text{g}^{-1}$ ) of the same leaf samples.

For leaf  $^{137}\text{Cs}$  concentrations, we assumed that  $^{137}\text{Cs}$  concentrations in *ante*-3.11 leaves (*CON*  $^{137}\text{Cs}$  *ante*-3.11 *leaves*) had exponentially decreased with time after the FDNPS accident (*Years after FDNPS accident*) according to empirical rates (*RATE*) obtained from the Fukushima 23 (*CON*  $^{137}\text{Cs}$  *ante*-3.11 *leaves* =  $a \exp(\text{RATE} \times \text{Years}$

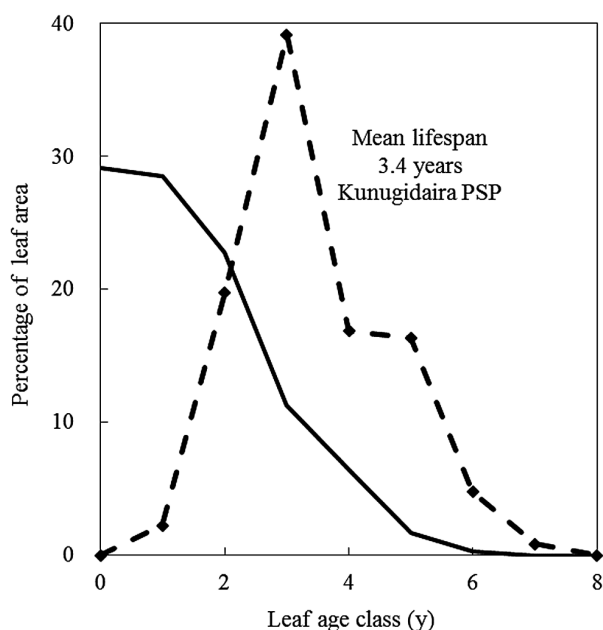


Fig. 3. Percentage of living and yearly deceased leaf area by leaf age class in the 33-year-old *Cryptomeria japonica* var. *japonica* forest in the Kunugidaira PSP. Solid line, leaf area; broken line and diamond, yearly deceased leaf area with a mean lifespan of 3.4 years.

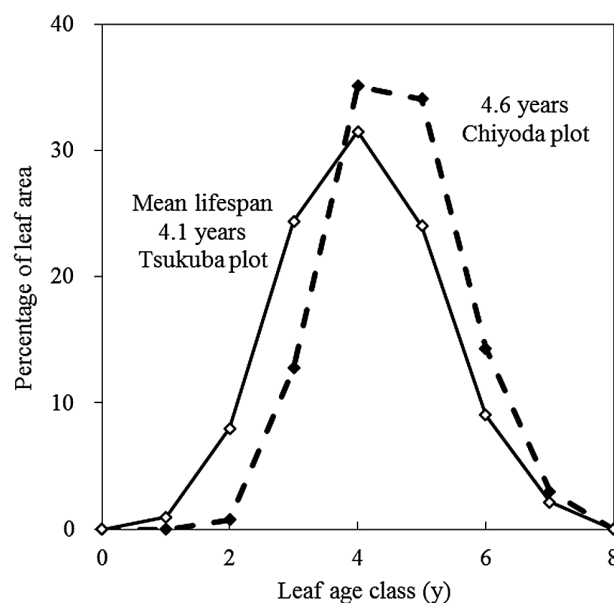


Fig. 4. Percentage of dead leaf area by leaf age class in withered small branch and leaf modules of *Cryptomeria japonica* var. *japonica* forests. Solid line and open diamond, 40-year-old stand in the Tsukuba plot (mean lifespan, 4.1 years); broken line and closed diamond, 19-year-old stand in the Chiyoda plot (mean lifespan, 4.6 years).

after FDNPS accident)). In addition, we assumed that  $^{137}\text{Cs}$  concentrations in *post*-3.11 leaves (*CON*  $^{137}\text{Cs}$  *post*-3.11 leaves) had changed according to the multi-regression equation derived from the Fukushima 23 with independent variables of leaf  $^{137}\text{Cs}$  concentrations in March 2011 (*CON*  $^{137}\text{Cs}$  *ante*-3.11 leaves immediately after fallout) estimated from  $^{137}\text{Cs}$  levels measured after the FDNPS accident and *RATE*, leaf age (*Leaf age*), and time after the FDNPS accident (*CON*  $^{137}\text{Cs}$  *post*-3.11 leaves =  $(b - c \text{ Years after FDNPS accident} - d \text{ Leaf age}) \times \text{CON } ^{137}\text{Cs ante-3.11 leaves immediately after fallout}$ ).

## 2.4 Verifying the predicted *sugi* leaf $^{137}\text{Cs}$ concentrations in the canopy

Using the model described above and the leaf  $^{137}\text{Cs}$  concentrations measured in 2011 (Forestry Agency 2014b), leaf  $^{137}\text{Cs}$  concentrations in the following 2 years were predicted and verified with leaf  $^{137}\text{Cs}$  concentrations obtained from measurements independent from ours (Forestry Agency 2014b) in the four PSPs in Fukushima Prefecture (Kawauchi, Kamikawauchi, Otama, and Tadami) (Table 1). The data from a Forestry Agency (2014b) press release were only the sum of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  concentrations and leaves not divided by leaf age class. Therefore, the ratios of leaf  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  concentration in the original

leaf samples (unpublished data for a FFPRI grant project) were used to obtain the leaf  $^{137}\text{Cs}$  concentrations. Leaf mass was divided into current-year (*post*-3.11) leaf mass and other (*ante*-3.11) leaf mass using the above-mentioned Gompertz equation to predict leaf mass distribution by leaf age class.

Using the first leaf  $^{137}\text{Cs}$  concentration measurement and assuming that the leaf concentration decreased at an empirical rate in *ante*-3.11 leaves, leaf  $^{137}\text{Cs}$  concentrations were estimated for March 2011 immediately after the fallout from the FDNPS accident for each PSP.  $^{137}\text{Cs}$  concentrations in *ante*-3.11 leaves on September 1, 2012 and September 1, 2013 were predicted using the estimated leaf concentrations in March 2011 and the empirical rate of  $^{137}\text{Cs}$  decrease in *ante*-3.11 leaves. Concentrations in *post*-3.11 leaves were predicted using the empirical multi-regression equation using estimated leaf concentrations in March 2011, leaf age, and time after FDNPS accident for each PSP. The stand leaf mass was assumed to be 20  $\text{Mg ha}^{-1}$ . The stand leaf concentrations were compared to the measured  $^{137}\text{Cs}$  concentrations in the PSPs based on destructive biomass sampling (Kajimoto et al. 2014) and converted into concentrations on September 1, 2012 and September 1, 2013 (Forestry Agency 2014b). We calculated the estimation error (%) as:  $(\text{predicted concentration} - \text{measured concentration}) / \text{predicted concentration} \times 100$ .



Table 3. Percentage of leaf area of each leaf age class in 33-year-old *Cryptomeria japonica* var. *japonica* trees in the Kunugidaira PSP

| No. | Percentage of leaf area of each leaf age class |           |     |           |     |     |       |
|-----|--|-----------|-----|-----------|-----|-----|-------|
|     | 0-y  | 1-y       | 2-y | 3-y       | 4-y | 5-y | 6–7-y |
| 1   | <b>31</b>                                      | 26        | 22  | 12        | 7   | 2   | 0.1   |
| 2   | <b>38</b>                                      | 31        | 18  | 8         | 4   | 1   | 0.1   |
| 3   | 25   | <b>28</b> | 26  | 13        | 7   | 1   | 0.5   |
| 4   | 27   | <b>36</b> | 25  | 7         | 4   | 1   | 0.0   |
| 5   | 27   | <b>35</b> | 23  | 11        | 4   | 0.3 | 0.0   |
| 6   | 21   | <b>28</b> | 25  | 16        | 8   | 2   | 0.2   |
| 7   | 14   | 21        | 23  | <b>23</b> | 15  | 3   | 2     |

Nos. 1 and 2, large trees; 3–6, medium-sized trees; and 7, small tree. The largest percentage of leaf area in each leaf age class is shown in italicized text for every tree.

### 3. Results

#### 3.1 Lifespan of *sugi* leaves

**Leaf age structure method:** The stand leaf mass and leaf area of the 33-year-old *sugi* planted forest were estimated to be 20.3 Mg ha<sup>-1</sup> and 5.70 (one-sided) ha ha<sup>-1</sup>, respectively. Age distribution in the leaf area of dead leaf produced in a year (broken line, Fig. 3) was estimated based on the age distribution in leaf area (solid line, Fig. 3). The dead leaf age was 1–8 years and the distribution of dead leaf area of each leaf age class was considered to follow a normal distribution. The mean leaf at death was 3.4 years. Since *sugi* sprouts new leaves around April in the study area and the destructive leaf sampling was conducted in March, we adjusted the age by 0.9 years to obtain a mean leaf lifespan of 4.3 years.

The one-sided leaf area of dead leaves of the stand produced in 1 year was estimated to be 1.59 ha ha<sup>-1</sup> y<sup>-1</sup>. For each tree (Table 3), the percentage of current-year leaves of total leaves was the highest in large trees (Nos. 1 and 2), indicating that the dominant trees were expanding their crowns at the time of sampling. The smallest tree (No. 7) had the highest percentage of 3-year-old leaves, and its crown was considered to be decreasing. The medium-sized trees (Nos. 3–6) had the highest percentage of 1-year-old leaves, indicating that their crown development was intermediate among large and small trees. The vertical distribution of the values of leaf area by leaf age class in the stand is shown in Supplementary Figures 1 and 2.

**Withered branch and leaf module method:** Using the withered small branch and leaf module, we determined leaf age to be 1–8 years (Tables 4 and 5) and the distribution of dead leaf area by age class was considered to be almost normal (Fig. 4). The mean leaf age at death was 4.6 years

in the Chiyoda plot and 4.1 years in the Tsukuba plot. Since the withered small branch and leaf modules were sampled in November 2011–January 2012, we adjusted the age by 0.7 years to obtain mean leaf lifespans of 5.3 and 4.8 years, respectively.

*Sugi* leaf survival was estimated to be 1–8 years old with a mean lifespan of 4.3–5.3 years based on the leaf age structure and withered branch and leaf modules methods. The leaf age structure method appeared to provide younger ages than the withered branch and leaf module method; however, the results of the two methods differed by only 0.5–1.0 year.

In addition, the oldest leaf ages of nine *sugi* in the three PSPs in Fukushima Prefecture (three trees per PSP) were 4–6 years old ( $5.0 \pm 1.0$  years, mean and standard deviation (SD)) in the Kawauchi PSP, 8 years ( $8.0 \pm 0.0$  years) in the Kamikawauchi PSP, and 6–7 years ( $6.7 \pm 0.6$  years) in the Otama PSP.

#### 3.2 *Sugi* leaf and <sup>137</sup>Cs dynamics in the canopy

The relationship between leaf age class and *LAR* in each leaf age class (solid line, Fig. 3) was approximated using the following Gompertz equation, where the leaf age class was 0–7 years:

$$LAR = 31.9 \times 0.9156^{(1.99^{\text{Leaf age class}})} \quad (R^2 = 0.9827, P < 0.001, n = 8) \quad (1)$$

where *LAR* is the one-sided leaf area ratio (%) by leaf age class. In addition, the relationship between the mean *SLA* and leaf age class was approximated as the following equation:

$$SLA = -3.86 \text{ Leaf age class} + 42.4 \quad (R^2 = 0.5652, P < 0.001, n = 48) \quad (2)$$

The leaf area of *ante*-3.11 leaves in the canopy was predicted to decrease rapidly to 46% of the total ~2 years after the FDNPS accident, 10% ~4 years after the FDNPS accident, and 0.1% 6 years after the FDNPS accident (Fig. 5a). Similarly, the leaf mass of *ante*-3.11 leaves in the canopy was predicted to decrease over time, although at a slightly delayed rate compared to leaf area, with a decrease to 53% at 2 years, 13% at 4 years, and 0.2% at 6 years after the FDNPS accident (Fig. 5b). Therefore, some *ante*-3.11 leaves are predicted to remain for up to 6 years after the FDNPS accident, until March 2017. However, most (~90%) of the *ante*-3.11 leaves were predicted to die within ~4 years of the FDNPS accident (March 2015). Therefore, the potential supply of *ante*-3.11 leaves to the forest floor was considered

Table 4. Percentage of the outlined leaf area of each leaf age class of withered small branch and leaf modules in the *Cryptomeria japonica* var. *japonica* forest in the Chiyoda plot

| Tree–<br>module no. | Leaf age (y) and percentage of the outlined leaf area <sup>1</sup> of each leaf age class |   |    |     |      |      |     |     |   |
|---------------------|---|---|----|-----|------|------|-----|-----|---|
|                     | 0   | 1 | 2  | 3   | 4    | 5    | 6   | 7   | 8 |
| 1–1                 |   |   |    | 0   | 0    | 20   | 40  | 40  | 0 |
| 1–2                 |   |   | 0  | 50  | 50   | 0    |     |     |   |
| 2–1                 |   |   |    | 10  | 10   | 40   | 40  | 0   |   |
| 2–2                 |   |   |    | 0   | 50   | 50   | 0   |     |   |
| 3–1                 |   |   |    | 20  | 40   | 40   |     |     |   |
| 3–2                 |   |   |    | 20  | 40   | 40   |     |     |   |
| 4–1                 |   |   |    | 0   | 33   | 33   | 33  |     |   |
| 4–2                 |   |   |    | 0   | 50   | 50   |     |     |   |
| 5–1                 |   |   |    | 20  | 40   | 40   |     |     |   |
| 5–2                 |   |   |    | 0   | 20   | 40   | 40  |     |   |
| 6–1                 |   |   |    | 0   | 20   | 40   | 40  |     |   |
| 6–2                 |   |   | 0  | 20  | 40   | 40   |     |     |   |
| 7–1                 |   |   | 0  | 50  | 50   | 0    |     |     |   |
| 7–2                 |   |   |    | 0   | 20   | 40   | 40  |     |   |
| 8–1                 |   |   |    | 0   | 20   | 40   | 40  |     |   |
| 8–2                 |   |   |    |     | 50   | 50   | 0   |     |   |
| 9–1                 |   |   |    |     |      | 50   | 50  |     |   |
| 9–2                 |   |   |    |     | 0    | 100  | 0   |     |   |
| 10–1                |   |   |    |     |      | 0    | 20  | 80  | 0 |
| 10–2                |   |   |    | 0   | 80   | 20   |     |     |   |
| 11–1                |   |   |    | 0   | 50   | 50   |     |     |   |
| 11–2                |   |   |    |     | 20   | 40   | 40  |     |   |
| 12–1                |   |   | 10 | 80  | 10   |      |     |     |   |
| 12–2                |   | 0 | 10 | 40  | 40   | 10   |     |     |   |
| 13–1                |   |   | 0  | 0   | 50   | 50   |     |     |   |
| 13–2                |   |   |    |     | 50   | 50   | 0   |     |   |
| 14–1                |   |   |    | 0   | 20   | 40   | 40  | 0   |   |
| 14–2                |   |   |    | 0   | 40   | 40   | 20  |     |   |
| 15–1                |   |   |    |     | 20   | 40   | 40  |     |   |
| 15–2                |   |   |    |     | 20   | 40   | 40  |     |   |
| 16–1                |   |   | 0  | 10  | 40   | 40   | 10  |     |   |
| 16–2                |   |   | 0  | 20  | 40   | 40   |     |     |   |
| 17–1                |   |   | 0  | 50  | 50   |      |     |     |   |
| 17–2                |   | 0 | 10 | 30  | 20   | 20   | 20  |     |   |
| 18–1                |   |   | 0  | 50  | 50   |      |     |     |   |
| 18–2                |   |   |    | 20  | 80   | 0    |     |     |   |
| 19–1                |   |   |    |     | 50   | 50   |     |     |   |
| 19–2                |   |   |    | 20  | 40   | 40   |     |     |   |
| 20–1                |   |   |    | 0   | 100  | 0    |     |     |   |
| 20–2                |   |   |    |     | 0    | 80   | 20  |     |   |
| Total               | 0   | 0 | 30 | 510 | 1403 | 1363 | 573 | 120 | 0 |
| Percentage          | 0   | 0 | 1  | 13  | 35   | 34   | 14  | 3   | 0 |

<sup>1</sup> Measured from visual observations (%). Zero (0) indicates an area of <5%.

Table 5. Percentage of the outlined leaf area of each leaf age class of withered small branch and leaf modules in the *Cryptomeria japonica* var. *japonica* forest in the Tsukuba plot

| Tree–<br>module no. | Leaf age (y) and percentage of the outlined leaf area <sup>1</sup> of each leaf age class |    |     |     |      |     |     |    |
|---------------------|---|----|-----|-----|------|-----|-----|----|
|                     | 0   | 1  | 2   | 3   | 4    | 5   | 6   | 7  |
| 1–1                 |   | 0  | 20  | 40  | 40   |     |     |    |
| 1–2                 |   | 0  | 20  | 40  | 40   |     |     |    |
| 2–1                 |   |    | 0   | 50  | 50   |     |     |    |
| 2–2                 |   |    |     | 0   | 50   | 50  |     |    |
| 3–1                 |   |    | 10  | 40  | 40   | 10  |     |    |
| 3–2                 |   |    | 10  | 80  | 10   |     |     |    |
| 4–1                 |   | 7  | 7   | 7   | 27   | 27  | 27  | 0  |
| 4–2                 |   |    |     | 10  | 80   | 10  |     |    |
| 5–1                 |   |    | 33  | 33  | 33   |     |     |    |
| 5–2                 |   |    | 10  | 80  | 10   |     |     |    |
| 6–1                 |   |    |     | 10  | 10   | 40  | 40  | 0  |
| 6–2                 |   |    | 10  | 30  | 30   | 30  | 0   |    |
| 7–1                 |   |    |     | 0   | 20   | 40  | 40  |    |
| 7–2                 |   |    |     |     | 20   | 80  | 0   |    |
| 8–1                 |   | 5  | 5   | 5   | 80   | 5   |     |    |
| 8–2                 |   | 0  | 7   | 7   | 7    | 40  | 40  | 0  |
| 9–1                 |   |    | 0   | 33  | 33   | 33  |     |    |
| 9–2                 |   |    |     | 25  | 25   | 25  | 25  |    |
| 10–1                |   |    | 0   | 20  | 40   | 40  | 0   |    |
| 10–2                |   |    | 0   | 20  | 40   | 40  | 0   |    |
| 11–1                |   | 10 | 40  | 40  | 10   |     |     |    |
| 11–2                |   |    |     | 20  | 40   | 40  | 0   |    |
| 12–1                |   | 10 | 20  | 20  | 20   | 20  | 0   |    |
| 12–2                |   | 0  | 10  | 10  | 40   | 40  | 0   |    |
| 13–1                |   | 7  | 7   | 7   | 40   | 40  |     |    |
| 13–2                |   |    | 7   | 7   | 80   | 7   |     |    |
| 14–1                |   |    |     | 10  | 10   | 27  | 27  | 27 |
| 14–2                |   |    | 7   | 7   | 40   | 40  | 7   |    |
| 15–1                |   | 0  | 10  | 40  | 40   | 10  |     |    |
| 15–2                |   |    | 20  | 27  | 27   | 27  | 0   |    |
| 16–1                |   |    |     | 25  | 25   | 25  | 25  |    |
| 16–2                |   | 0  | 25  | 50  | 25   |     |     |    |
| 17–1                |   |    |     | 10  | 30   | 30  | 30  | 0  |
| 17–2                |   |    |     |     | 25   | 25  | 25  | 25 |
| 18–1                |   |    |     |     | 0    | 33  | 33  | 33 |
| 18–2                |   |    | 0   | 20  | 40   | 40  | 0   |    |
| 19–1                |   |    | 7   | 80  | 7    | 7   |     |    |
| 19–2                |   |    | 27  | 27  | 27   | 0   |     |    |
| 20–1                |   |    |     | 33  | 33   | 33  | 0   |    |
| 20–2                |   |    | 7   | 7   | 7    | 40  | 40  | 0  |
| Total               | 0   | 38 | 317 | 968 | 1250 | 953 | 358 | 85 |
| Percentage          | 0   | 1  | 8   | 24  | 31   | 24  | 9   | 2  |

<sup>1</sup> Measured from visual observations (%). Zero (0) indicates an area of <5%



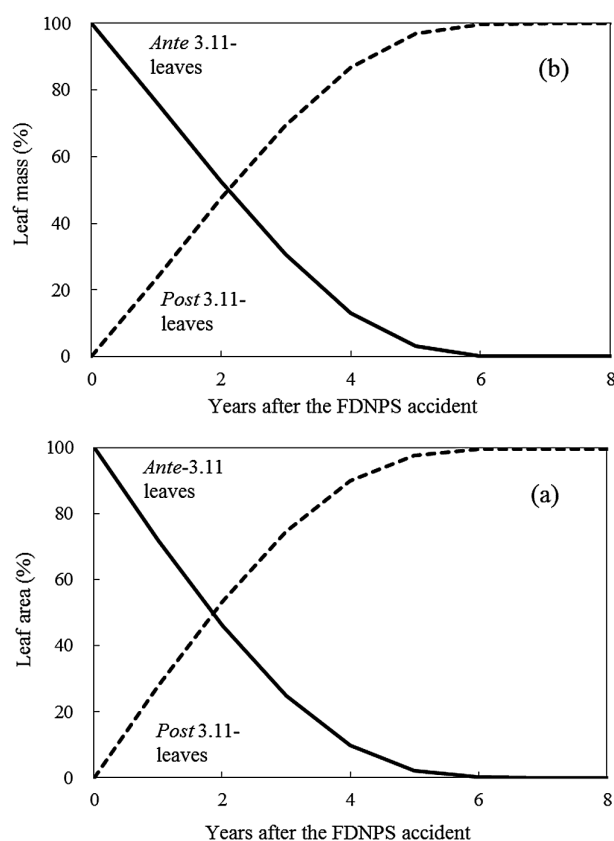


Fig. 5. Annual trends in the percentages of *ante*-3.11 leaves and *post*-3.11 leaves in the canopy of a *Cryptomeria japonica* var. *japonica* forest by (a) leaf area and (b) leaf mass

*Ante*-3.11 leaves (solid line) had sprouted before March 11, 2011 and were possibly contaminated directly with fallout from the FDNPS accident while *post*-3.11 leaves sprouted after March 11, 2011 and were not contaminated directly by the fallout.

to remain for up to ~5 years after the FDNPS accident (Fig. 5).

The  $^{137}\text{Cs}$  concentrations in the sampled leaves varied over time after the FDNPS accident (Fig. 6). The  $^{137}\text{Cs}$  concentrations in the sampled *ante*-3.11 leaves did not differ significantly (paired *t*-test,  $P = 0.260\text{--}0.902$ ,  $n = 2\text{--}9$ ) among leaves with different birth years (2005–2010) in nine *sugi* trees from the Kawauchi, Kamikawauchi, and Otama PSPs collected in August 2013. Therefore, the *ante*-3.11 leaves of the Fukushima 23 were pooled. The distribution of leaf  $^{137}\text{Cs}$  concentrations in the leaf samples of different birth years and observation years followed a logarithmic normal distribution (D'Agostino's K-squared test,  $K^2 = 0.3447\text{--}2.092 < 5.991$ ,  $P = 0.3513\text{--}0.8417 > 0.05$ ; Anderson–Darling test,  $A^2 = 0.1973\text{--}0.4776 < 0.752$ ,  $n = 23$ ). The geometric mean  $^{137}\text{Cs}$  concentration of Fukushima 23 *ante*-3.11 leaves ( $\bigcirc$ , Fig. 6) decreased between 1 year 8 months and 3 years 8 months after the FDNPS accident. The sampled *post*-3.11 leaves from 2011 ( $\times$ , Fig. 6) and

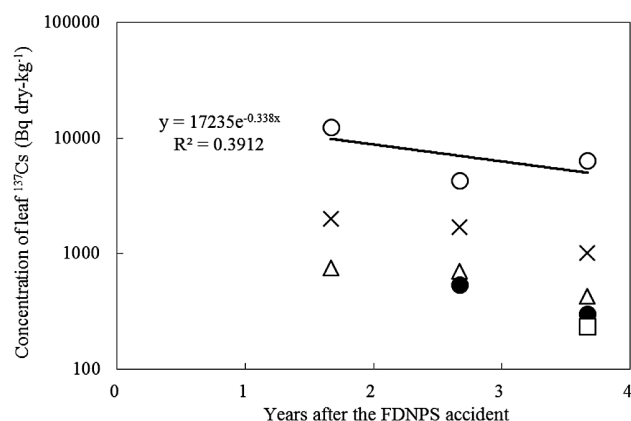


Fig. 6. Cesium-137 ( $^{137}\text{Cs}$ ) concentrations of *Cryptomeria japonica* var. *japonica* leaves between 1 year 8 months and 3 years 8 months after the FDNPS accident in 23 *C. japonica* var. *japonica* forests in Fukushima Prefecture (Akama et al. 2013; Forestry Agency 2014a)  $\bigcirc$ , leaves that sprouted in 2010 or earlier;  $\times$ , leaves that sprouted in 2011;  $\Delta$ , leaves that sprouted in 2012;  $\bullet$ , leaves that sprouted in 2013;  $\square$ , leaves that sprouted in 2014.

2012 ( $\Delta$ , Fig. 6) had  $^{137}\text{Cs}$  concentrations of 2,000 and 750 Bq dry-kg $^{-1}$  in November 2012, respectively, which changed little in November 2013 and then decreased. The concentrations in leaves that sprouted in 2013 ( $\bullet$ , Fig. 6) were lower than those of previous years.

The results of the Fukushima 23 *ante*-3.11 leaves ( $\bigcirc$ , Fig. 6) were used to obtain the empirical equation:

$$\text{CON } ^{137}\text{Cs ante-3.11 leaves} = 17,235 \exp(-0.338 \times \text{Years after FDNPS accident}) \quad (R^2 = 0.3912, n = 3) \quad (3)$$

where  $\text{CON } ^{137}\text{Cs ante-3.11 leaves}$  is in Bq dry-kg $^{-1}$ . The  $^{137}\text{Cs}$  concentration in *sugi* leaves immediately after the fallout from the FDNPS accident was estimated to be 17 kBq dry-kg $^{-1}$  with Eq. 3. In November 2012, the  $^{137}\text{Cs}$  concentrations in *ante*-3.11 and *post*-3.11 leaves were 72.6% and 4.3–11.6% of the initial 17 kBq dry-kg $^{-1}$  (Fig. 6). Assuming a stand leaf biomass of 20 Mg ha $^{-1}$ , equivalent to a one-sided leaf area of ~5.6 ha ha $^{-1}$ ,  $^{137}\text{Cs}$  deposition on *sugi* leaves was estimated to be 34 kBq m $^{-2}$  in the Fukushima 23 forests, which represented the leaf  $^{137}\text{Cs}$  concentration per unit of land area when the fallout from the FDNPS accident reached the site.

Although the  $^{137}\text{Cs}$  concentrations of *post*-3.11 leaves decreased minimally until 2013, they then decreased markedly and exhibited an overall annual decrease in  $^{137}\text{Cs}$  concentrations over time according to leaf birth year and time after the FDNPS accident (Fig. 6). We described these results with the empirical equation:

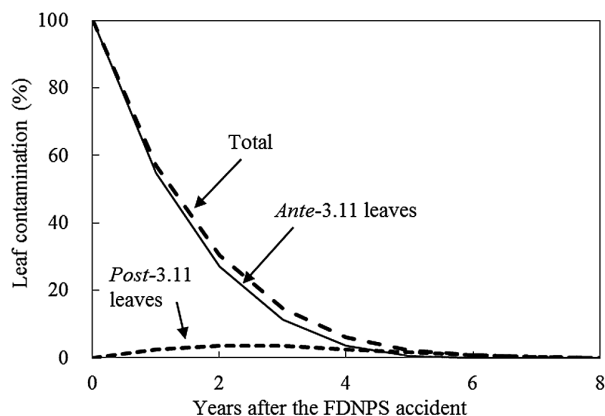


Fig. 7. Estimated leaf  $^{137}\text{Cs}$  contamination of *Cryptomeria japonica* var. *japonica* forests after the FDNPS accident based on the model

Leaf contamination, percentage of the amount of leaf  $^{137}\text{Cs}$  per unit land area when the fallout from the FDNPS accident reached the site. *Ante-3.11* leaves, leaves that sprouted before the FDNPS accident and were possibly directly contaminated with fallout material. *Post-3.11* leaves, leaves that sprouted after the FDNPS accident.

$CON^{137}\text{Cs post-3.11 leaves} = (0.116 - 0.0143 \times \text{Years after FDNPS accident} - 0.0233 \times \text{Leaf age}) \times CON^{137}\text{Cs ante-3.11 leaves immediately after fallout}$  ( $R^2 = 0.7596$ ,  $P = 0.014$ ,  $n = 9$ ) (4)

where  $CON^{137}\text{Cs post-3.11 leaves}$  is the  $^{137}\text{Cs}$  concentration of *post-3.11* leaves in  $\text{Bq dry-kg}^{-1}$ , *Years after FDNPS accident* is in years, *Leaf age* is the age of *post-3.11* leaves in years, and  $CON^{137}\text{Cs ante-3.11 leaves immediately after fallout}$  is *ante-3.11* leaf  $^{137}\text{Cs}$  concentration immediately after the fallout in  $\text{Bq dry-kg}^{-1}$ .

We assumed that the *ante-3.11* leaf  $^{137}\text{Cs}$  concentrations changed according to the empirical rate obtained from the Fukushima 23,  $\exp(-0.338 \times \text{Years after FDNPS accident})$ , and that the *post-3.11* leaf  $^{137}\text{Cs}$  concentrations changed according to Eq. 4 until leaf mortality. Furthermore, by incorporating the leaf replacement patterns (Fig. 5b), we predicted yearly changes in the leaf contamination which was expressed as percentage of the amount of leaf  $^{137}\text{Cs}$  per unit land area relative to the initial value when the fallout from the FDNPS accident reached the site (Fig. 7). The leaf contamination in *ante-3.11* leaves in the canopy, which were possibly directly contaminated with fallout material, decreased by ~90% within 3 years of the FDNPS accident (March 2014). The leaf contamination of *post-3.11* leaves slowly increased in the first 3 years and then decreased.

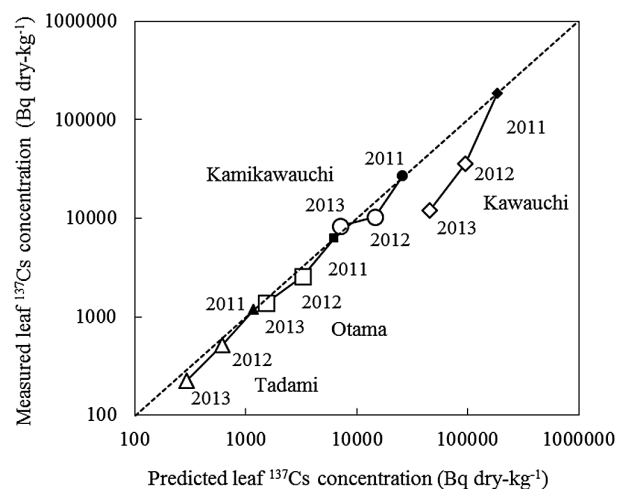


Fig. 8. Comparison of the chronosequential changes between predicted and measured leaf  $^{137}\text{Cs}$  concentrations in *Cryptomeria japonica* var. *japonica* forests  $\diamond$ , Kawauchi PSP;  $\circ$ , Kamikawauchi PSP;  $\square$ , Otama PSP;  $\triangle$ , Tadami PSP. The broken line represents equal predicted and measured concentrations. Closed symbols ( $\diamond$ ,  $\circ$ ,  $\square$ ,  $\triangle$ ) represent the measured concentrations in 2011 (the first measurement).

### 3.3 Verifying the predicted *sugi* leaf $^{137}\text{Cs}$ concentrations in the canopy

Assuming that the *ante-3.11* leaf concentrations decreased at a rate of  $\exp(-0.338 \times \text{Years after FDNPS accident})$  (Fig. 6, Eq. 3), the leaf  $^{137}\text{Cs}$  concentrations and amounts per land area when the fallout from the FDNPS accident reached each site (March 2011) were estimated to be  $276 \text{ kBq dry-kg}^{-1}$  and  $552 \text{ kBq m}^{-2}$  at the Kawauchi PSP,  $43.2 \text{ kBq dry-kg}^{-1}$  and  $86.3 \text{ kBq m}^{-2}$  at the Kamikawauchi PSP,  $9.35 \text{ kBq dry-kg}^{-1}$  and  $18.7 \text{ kBq m}^{-2}$  at the Otama PSP, and  $1.77 \text{ kBq dry-kg}^{-1}$  and  $3.54 \text{ kBq m}^{-2}$  at the Tadami PSP. The estimated *sugi* leaf  $^{137}\text{Cs}$  amounts per land area when the fallout from the FDNPS accident reached each site roughly corresponded to the amounts of  $^{137}\text{Cs}$  deposited on the ground surface,  $600\text{--}1,000 \text{ kBq m}^{-2}$  in the Kawauchi PSP and  $<300 \text{ kBq m}^{-2}$  in the Kamikawauchi, Otama, and Tadami PSPs; these values were obtained from an airborne monitoring survey (Ministry of Education, Culture, Sports, Science and Technology 2011a). The airborne monitoring survey used helicopters at ~300 m above ground level, and each data point of radiocesium deposition represented the mean of a circular area of ground with a 300-m radius (Torii et al. 2012). The values were then converted to estimate the values on April 29, 2011 (Ministry of Education, Culture, Sports, Science and Technology 2011a).

The predicted stand leaf  $^{137}\text{Cs}$  concentrations (Fig. 8) on September 1, 2012 were overestimated by 63% in the Kawauchi PSP, 29% in the Kamikawauchi PSP, 20%

in the Otama PSP, and 16% in the Tadami PSP, with a mean overestimation of  $27 \pm 29\%$ . On September 1, 2013, the predicted concentrations were overestimated by 74% in the Kawauchi PSP, underestimated by 17% in the Kamikawauchi PSP, overestimated by 10% in the Otama PSP, and 22% in the Tadami PSP, with a mean overestimation of  $32 \pm 21\%$ . Overall, the model simulated annual trends in stand leaf  $^{137}\text{Cs}$  concentrations for the PSPs in 2012 and 2013 within a  $22 \pm 38\%$  margin.

#### 4. Discussion

##### 4.1 *Sugi* canopy leaf survival

Most *ante*-3.11 leaves were likely to remain in the canopy for  $\sim 4$  years, and some could survive for  $\geq 6$  years after the FDNPS accident. For the first time, we determined the mean leaf lifespan of *sugi* leaves (4.3–5.3 years). The leaf age structure method estimated ages 0.5–1.0 younger than the withered branch and leaf module method. The best explanation of this difference is that it is due to the collection methods. Since *sugi* leaves mainly die in the autumn and winter, and the leaf samples used for the leaf age structure method in this study were collected in March near the end of the die-off season, we collected few senile leaves from the previous season. Accordingly, the estimate of mean leaf lifespan using the leaf age structure method (4.3 years) was considered an underestimate. By contrast, the samples for the withered branch and leaf module method were collected mostly from lower branches, which are often suppressed and have a slower leaf replacement rate. Therefore, the mean leaf lifespans using this method (5.3 and 4.8 years) were considered overestimated. Regardless, *sugi* has a longer leaf lifespan than *Pinus densiflora* of 1–2 years (Hatiya et al. 1966), and a similar leaf lifespan to that of *Chamaecyparis obtusa*: 2–6 years (Kiyono 1990) or 3–7 years (Miyamoto et al. 2013).

There is little information available on the interval between leaf death and shedding in *sugi*. Moreover, the duration of this interval could vary by forest conditions. In young *sugi* forests <20–30 years old, dead branches and leaves often require several years to be shed (Fujimori and Kiyono 1983). However, tall trees in >40-year-old forests are more easily disturbed by wind and form spaces between their crowns (Fujimori 1997) with fewer dead branches. Unlike young stands (17 and 31 years old), a 53-year-old *sugi* stand had the highest leaf litterfall rates in autumn–winter (see Fig. 3 in Miura 2000). The *sugi* trees in the Kawauchi, Otama, and Tadami PSPs were 38–43 years old in 2011 (Table 1). According to our observations, the small branch and leaf modules that died in the autumn and winter of 2011 had mostly been shed by May 2012 in all forests.

A few small modules comprised mainly of green leaves were also shed, likely due to wind and snow in autumn and winter. Therefore, it is reasonable to assume that most withered small branch and leaf modules are shed in the first autumn and winter after their death in *sugi* forests of the ages of those in the Kawauchi, Otama, and Tadami PSPs.

##### 4.2 $^{137}\text{Cs}$ retention in *sugi* canopy leaves

After the FDNPS accident, radiocesium on leaves in the canopy was predominantly lost via washing with throughfall (Hayashi 2013, Kato et al. 2015) and in litterfall (Endo et al. 2015, Loffredo et al. 2015). Washing was considered to be the main cause of the decrease in  $^{137}\text{Cs}$  concentrations in *ante*-3.11 leaves initially. Between 4 and 5 months after the FDNPS accident, the  $^{137}\text{Cs}$  concentrations in throughfall and stemflow in *sugi* forests in Fukushima Prefecture were more than 50–200 times the  $^{137}\text{Cs}$  concentrations in rainfall (mean,  $0.4 \text{ Bq kg}^{-1}$ ) (Ministry of Education, Culture, Sports, Science and Technology 2011b). In addition, within 1 year of the FDNPS accident, the amount of  $^{137}\text{Cs}$  transferring from the canopy to the forest floor by leaching, washing, and throughfall was >10 times that of litterfall in a *sugi* plantation in Ibaraki Prefecture (Hayashi 2013).

According to our model (Fig. 5),  $\sim 90\%$  of the *ante*-3.11 leaves were predicted to die within 4 years of the FDNPS accident (March 2015), while  $\sim 90\%$   $^{137}\text{Cs}$  in *ante*-3.11 leaves in the canopy were predicted to disappear from the canopy within 3 years of the FDNPS accident (March 2014) (Fig. 7). During this period, the radiocesium deposited on *ante*-3.11 leaves could be the largest source of radiocesium transferred to soil, forest flora and fauna, and surface waters via weathering, leaf decomposition, foliar absorption, translocation, leaching in rainwater, and other processes. Therefore, tree thinning by the government to reduce the air dose in forests contaminated by radiation (Fukushima Minpo 2014) could be effective in the first 3 years.

Most of the  $^{137}\text{Cs}$  in *post*-3.11 leaves was predicted to have translocated from other parts of the same tree (Bunzl and Kracke 1989) and was the main source of  $^{137}\text{Cs}$  from the canopy to the forest floor via leaf shedding >4 years after the FDNPS accident (Fig. 7). Moreover, some of the total  $^{137}\text{Cs}$  will be cycled through the forest ecosystem for a long time (Yamaguchi et al. 2012, Ota et al. 2016).

According to our model, the predicted stand leaf  $^{137}\text{Cs}$  concentrations were mostly overestimates based on measured values (Fig. 8). There are several explanations for this. First, the *sugi* in the Kawauchi and Otama PSPs had the oldest leaves (Kawauchi, 5.0 years; Otama, 6.7 years), which were still younger than the age used in the simulation (8 years). Similar to leaf lifespan, the age of the oldest

leaves can differ depending on environmental conditions. For example, the *ante*-3.11 leaves in Kawauchi and Otama were lost earlier than in the model. In addition, the assumption that the rate of *post*-3.11 leaf  $^{137}\text{Cs}$  concentration decrease is constant (Eq. 4) may be an over-simplification. In reality, *post*-3.11 leaves may have more complex annual trends of decline in leaf  $^{137}\text{Cs}$  concentrations (Fig. 6). For example, the leaf  $^{137}\text{Cs}$  concentrations dropped rapidly in the last year of observation (2013).

The influence of seasonality on leaf  $^{137}\text{Cs}$  concentrations (Rantavaara et al. 2012) is unclear, as only a portion of the growing season was studied. There is limited information on the effects of exposure on leaf survival. Leaf lifespan seemed to be shorter in the Kawauchi PSP (5.0 years), the most contaminated of the three PSPs. However, further analyses are required to clarify this issue and determine whether it is an important factor in  $^{137}\text{Cs}$  transfer.

In general, *sugi* leaf  $^{137}\text{Cs}$  concentrations changed chronosequentially depending on the dynamics of  $^{137}\text{Cs}$  in forest ecosystems, including continued  $^{137}\text{Cs}$  deposition,  $^{137}\text{Cs}$  absorption by *sugi*, and the impacts of various environmental conditions. The results of our model indicate that radiocesium concentrations in leaves decrease over time (Fig. 7); however, longer-term observations are required to increase the robustness of this trend in *post*-3.11 leaves (Fig. 6). The assumptions for the *post*-3.11 leaf analyses, such as the constant rates of decrease of *post*-3.11 leaf  $^{137}\text{Cs}$  concentrations (Eq. 4), ignoring potential seasonality, root uptake, and the proportion of  $^{137}\text{Cs}$  in leaves transferred to branches or other tree organs before leaf mortality (Bunzl and Kracke 1989), should be verified by future studies using more samples over a longer period. Continual monitoring is required to determine the progress of future *post*-3.11 leaf  $^{137}\text{Cs}$  concentrations. Longer-term data will contribute to improving our understanding and predictions of  $^{137}\text{Cs}$  contamination in *sugi* forests.

## 5. Conclusions

Radiocesium transfer from forest canopies to forest floors has been evaluated mainly by direct measurements of throughfall, stemflow, and litterfall. However, ecological characteristics related to leaf mortality and replacement vary depending on plant species and habitat, which could affect radiocesium transfer rates. In this study, we determined the contamination patterns of *sugi* leaves. Radiocesium translocation from the canopy to the forest floor in *sugi* forests was evaluated using a model that considered *sugi* leaf replacement patterns. Based on the results, radiocesium translocated from the canopy to the forest floor at a faster rate than the *ante*-3.11 leaves that

had directly been contaminated by the FDNPS accident. Including *sugi* data on leaf mortality and *sugi* replacement will allow more accurate prediction of radiocesium fate and persistence in *sugi* forests affected by the FDNPS accident. Furthermore, a deeper understanding of plant life, including the growth and death patterns of plant organs not limited to leaves, is important for elucidating the movement of radiocesium through ecosystems.

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#### Supplementary data

Supplementary data can be found at <http://www.ffpri.affrc.go.jp/pubs/bulletin/438/index.html>

Fig. S1. Vertical distribution of leaf area of each leaf age class in the 33-year-old *Cryptomeria japonica* var. *japonica* forest canopy in the Kunugidaira PSP

Fig. S2. Vertical distribution of the percentage of leaf area of each leaf age class in the 33-year-old *Cryptomeria japonica* var. *japonica* forest canopy in the Kunugidaira PSP

# 福島第一原子力発電所事故で放射能汚染したスギ人工林における 葉の交代とセシウム 137 濃度の経年変化予測

清野 嘉之<sup>1)\*</sup>、赤間 亮夫<sup>2)</sup>

## 要 旨

スギ人工林は日本を代表する常緑針葉樹の生態系である。スギはユニークな常緑樹で葉に離層を作らず、葉の寿命が明らかでない。スギの生きている葉と枯死したばかりの葉の齢構成を調べ、寿命と枯死過程を推定した。スギの葉の寿命は 1 ～ 8 年で、平均は 4.3 ～ 5.3 年であった。葉の交代様式とセシウム 137 濃度をモデル化し、福島第一原発事故後に採取したさまざまな齢の葉のセシウム 137 濃度のデータにもとづいて、落葉によるセシウム 137 の林床への供給可能量を推定した。2011 年 3 月 11 日以前に生まれた葉（事故前葉）の約 9 割は事故から 4 年以内に枯死すると予測された。また、林冠の事故前葉のセシウム 137 の約 9 割は事故から 3 年以内に他に移動すると予測された。モデルで予測した林分当たりの葉のセシウム 137 濃度のトレンドを、福島県に設けた 4 つの長期調査プロットの計測値で検証した。本研究により、福島第一原発事故で直接汚染した葉のセシウム 137 は事故前葉よりも速いペースで林冠から林床に移動することが明らかになった。スギの葉の枯死と交代の情報の利用により、福島第一原発事故により被災したスギ林の放射性セシウムの将来や存続の程度をより正確に予測できると考えられる。

キーワード：葉の枯死過程、常緑針葉樹、放射性降下物、葉の寿命、葉の交代、シミュレーション、スギ

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1) 森林総合研究所総務部研究専門員

2) 森林総合研究所企画部研究専門員

\* 森林総合研究所総務部研究専門員 〒 305-8687 茨城県つくば市松の里 1