論　文（Original article）

^{137}Cs concentrations in the pollen of sugi (*Cryptomeria japonica* var. *japonica*) over 5 years following the 2011 Fukushima Daiichi Nuclear Power Station accident in Fukushima Prefecture

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Abstract

We aimed to estimate the amount of radiocesium re-dispersed by sugi (*Cryptomeria japonica* var. *japonica*) pollen release accurately based on measurements of sugi male flowers before pollen release. We measured the biomass and cesium-137 (^{137}Cs) concentrations in male flowers and sugi pollen in November (shortly after pollen maturity), and in the following February (shortly before pollen release), in forests impacted by the 2011 Fukushima Daiichi Nuclear Power Station accident. Pollen accounted for *ca*. ⅓ of male flower biomass and the ^{137}Cs concentrations were not significantly different between male flowers and pollen in November in the Koriyama sugi forest. The regression relationship between male flower and pollen ^{137}Cs concentrations was used to calculate pollen ^{137}Cs concentrations for every November–December period in the years 2011–2015 at 21 localities in Fukushima Prefecture. The sugi pollen ^{137}Cs concentration declined year on year. The concentration in the spring of 2016 was *ca*. 8% of the concentration in spring 2012. The calculated ^{137}Cs T_{ag} (aggregated transfer factor) for the transmission of ^{137}Cs from the environment to pollen was 0.0203 m^2 dry kg^{-1} in February 2012 and 0.00168 m^2 dry kg^{-1} in February 2016, based on data available in the MEXT ^{137}Cs deposition quantity distribution map.

Key words: Deposition, distribution map, male flower, phenology, radiocesium, T_{ag} (aggregated transfer factor), yearly variation in pollen production

1. Introduction

Radiocesium is contained in flowers (Barišić et al. 1992, Molzahn and Assmann-Werthmüller 1993). After the 2011 TEPCO Fukushima Daiichi Nuclear Power Station (FDNPS) accident, airborne pollen (Bunzl et al. 1993) was identified as a dispersal vector of radiocesium (Ministry of Agriculture Forestry and Fisheries [MAFF] 2011, Tokuma Shoten 2011, TV Asahi 2011). Igarashi et al. (2019) also reported fungal spore involvement in the re-suspension of cesium-137 (^{137}Cs) during summer. Many sugi (*Cryptomeria japonica* var. *japonica*) trees grow in Fukushima Prefecture, which is of concern because their pollen can be inhaled by humans within the vicinity of the trees. The internal radiation dosage that results from breathing inhalation has been calculated for people living in Tokyo (MAFF 2011, Tsuruoka et al. 2015). Male sugi flowers are formed on current-year leaves in the June–August period. Pollen matures in mid to late October (Forestry Agency 2007) and becomes nutritionally independent in male flowers by November. Subsequently, male flowers enter dormancy and mature after this dormancy is broken by winter cold. Pollen is usually released from mid to late February in Fukushima Prefecture (Ministry of the Environment 2018). Pollen production fluctuates greatly from year to year (Yokoyama and Kanazashi 1999, Kajimoto and Fukushima 2015); the ability to predict pollen production is an important element of measures used to combat sugi-pollinosis (hay fever). Male flowers are usually observed in monitored forests from November to December (Teiten-sugi-rin, National Forestry Extension Association in Japan 2017, Ministry of the Environment 2016), when they are readily observable in tree crowns. The quantity of male flowers that will release pollen in the following spring was estimated during the monitoring procedure. However, the relationship between the observed

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male flower mass and the ^{137}Cs content of these flowers when the pollen is released in the following spring remains unclear. MAFF (2011) assumed that pollen mass accounts for *ca*. half of the observed male flower biomass, and that the ^{137}Cs concentrations of male flowers and pollen were identical. However, there are no data to verify these assumptions. Quantification of pollen/male flower biomass and ^{137}Cs concentration ratios will enable accurate calculation of the re-dispersal of ^{137}Cs mass by pollen based on measurements of male flowers prior to pollen release.

We conducted measurements of sugi male flowers in a forest located in Koriyama, Fukushima Prefecture on six occasions (November 2011, February and November 2012, February 2013, November 2015, February 2016) (Fig. 1, Δ). This forest was exposed to radiation released in the FDNPS accident. Subsequently, we examined the relationships among biomass and ^{137}Cs concentrations in male flowers and mature pollen in November. We estimated the ^{137}Cs concentration of sugi pollen dispersed in the spring in the 2012–2016 period based on these relationships and the measured ^{137}Cs concentrations in male flowers collected in the sugi forests (Fig. 1, ●) at diverse locations in Fukushima Prefecture, in the November–December period in each year between 2011 and 2015 (2011 data provided by the Forestry and Forest Products Research Institute (FFPRI) research project grant 'Methodology development for estimating radiocesium concentration in sugi pollen, #201128'; data for 2012–2015 provided by the Forestry Agency research project grant 'Understanding actual condition of radioactive substance in forest'). We also estimated the ^{137}Cs T_{ag} (aggregated transfer factor) for the transmission of ^{137}Cs from the environment to sugi pollen.

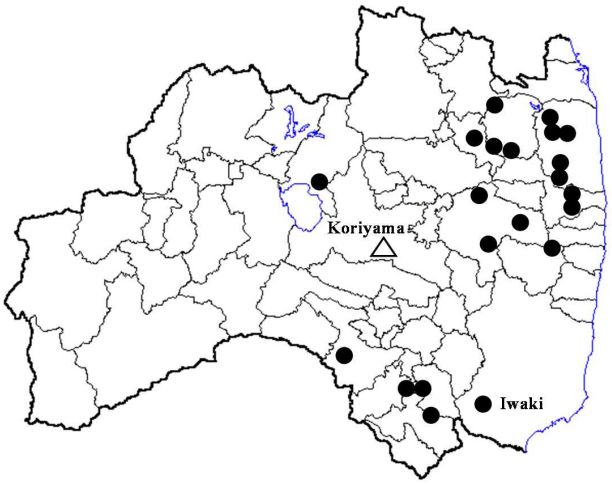

Fig. 1. Locations of sugi (*Cryptomeria japonica* var. *japonica*) forests that were surveyed.
Δ, location (Koriyama) at which the cesium-137 (^{137}Cs) concentrations and biomasses of sugi male flower parts were measured. ●, locations at which yearly changes in sugi male flower ^{137}Cs concentrations were measured.

Table 1. Measured biomasses and ^{137}Cs concentrations in sugi (*Cryptomeria japonica* var. *japonica*) male flower parts.

		2011.11	2012.2	2012.11	2013.2	2016.11	2017.2
Pollen/male flower biomass ratio (g g^{-1})		-	-	-	-	0.32 ± 0.05^{a}	0.48 ± 0.06^{b}
Male flower biomass (g 100 male flowers^{-1})		-	-	-	-	1.756	1.180 ± 0.181
Pollen biomass (g 100 male flowers^{-1})		-	-	-	-	0.561	0.561 ± 0.074
Male flower husk[1] biomass (g 100 male flowers^{-1})		-	-	-	-	1.196	0.620 ± 0.149
^{137}Cs concentration (Bq dry kg^{-1})	Male flower	565	1792 ± 487^{a}	$^{A}231 \pm 85^{b}$	-	51	59 ± 52^{c}
	Pollen	-	686 ± 292^{a}	-	$^{A}284 \pm 210^{b}$	-	$^{A}53 \pm 55^{c}$
	Male flower husk[1]	-	3256 ± 1256^{a}	-	379 ± 310^{b}	-	64 ± 48^{c}
	Current-year leaf	1430	-	$^{A}275 \pm 86^{a}$	$^{A}349 \pm 227^{a}$	-	$^{A}40 \pm 27^{b}$
Ratio of ^{137}Cs concentration in February pollen/ ^{137}Cs concentration in previous November male flowers		1.21		1.19		1.04	
Cryptomeria japonica var. *japonica* clone		Combined	Soma 5, Soma 9, Futaba 4, Iwaki 2, Yama 1, and Minamiaizu 5	Soma 1, Iwaki 1, Iwaki 7, Minamiaizu 7, and Ken-misho	Soma 1, Iwaki 1, Iwaki 7, Minamiaizu 7, and Ken-misho	Soma 1, Soma 9, Futaba 4, Iwaki 1, Iwaki 7, and Yama 1. Combined for ^{137}Cs	Soma 1, Soma 9, Futaba 4, Iwaki 1, Iwaki 7, and Yama 1

Different lower case superscript letters identify significant pairwise differences between means in rows (*t*-test; $P < 0.05$). Different upper case letters identify significant pairwise differences between means in columns (paired *t*-test; $P < 0.05$).
[1], including anther.

2. Materials and methods

2.1 Biomass and ^{137}Cs concentrations in sugi male flowers and pollen in November and the following February

Male flowers and current-year leaves were collected in November 2011 from several flowering plus-tree clones of sugi (Table 1) that had been planted in the progeny test demonstration forest [established in 1969, Kawakami (2000)] of the Fukushima Prefectural Forestry Research Centre (Koriyama City, Fig. 1, ∆). Here, we define sugi leaves as young stems and branches covered with live needle-like leaves (Kiyono and Akama 2016). Under this definition, some sugi leaves grow into stems or branches following the development of xylem. Male flowers were collected in February 2012 from six clones: Soma 5, Soma 9, Futaba 4, Iwaki 2, Yama 1, and Minamiaizu 5 (Table 1). Sun-exposed lower branches of trees on the forest edge *ca*. 0.6 m long bearing leaves and male flowers were collected (as on all subsequent occasions) and transferred to water-filled plastic crates for separate collection of pollen and other male flower organs (referred to hereafter as "male flower husk") (Fig. 2). Male flowers and current leaves were collected in November 2012 from four clones, Soma 1, Iwaki 1, Iwaki 7, and Minamiaizu 7, and from a non-clone sugi tree grown from a seedling (Ken-misho, referred to hereafter as a "clone temporarily"). In February 2013, pollen, male flower husks, and current-year leaves were collected from the same five clones via the procedure used in February 2012. In November 2016 and February 2017, male flowers were collected from six clones: Soma 1, Soma 9, Futaba 4, Iwaki 1, Iwaki 7 and Yama 1 (Table 1). In February 2017, the branches of each clone were cut and stood in water for collection of pollen, male flower husks, and current-year leaves. We counted the number of male flower husks on each clone on this occasion. We sampled six sugi clones in February 2012, five during the second procedure (November 2012/February 2013), and six during the third procedure (November 2016/February 2017). None of the clones bloomed well on all three occasions, likely a reflection of the annual fluctuation in sugi male flower formation and genetic variability. However three clones bloomed to the same extent on the first and third occasions, and on the second and third occasions. The provenances of the 10 clones were as follows: 7 from Hamadori, 1 from Nakadori, and 2 from Aizu, i.e., primarily from Hamadori, but spread throughout Fukushima Prefecture. The clones selected were considered representative of the sugi trees impacted by the FDNPS accident.

Collected and dried male flowers, pollen, male flower husks, and current-year leaves of each clone were packed separately in U-8 containers. The ^{137}Cs concentrations were measured using a high-purity germanium (HPGe) coaxial detector system at the FFPRI. In 2011 and 2016, the male flowers from multiple clones were combined into single samples. The ^{137}Cs concentrations in the samples prepared in November 2011 and February 2012 were measured by the Japan Frozen Foods Inspection Corporation (JFFIC); measurements of the remaining samples were obtained by the FFPRI. Measured values were standardized to zero moisture content based on our measurements of moisture content in the samples. We selected three medium-sized male flowers as samples from each clone in November 2016 and February 2017, and dissected out the pollen and male flower husk with needles under a stereoscopic microscope (SMZ 800-1; Nikon). Anthers were included with the male flower husks. In November, a large amount of resin was observed in the resin paths of the husks, and the removed pollen grains were often smeared with this resin. In February, the amount of resin was reduced and pollen/male flower husk separation was relatively easy. The masses of the pollen and male flower husks were measured using an electronic balance (XS 205 DU; Mettler Toledo; max weight, 81/220 g, d = 0.01 mg/0.1 mg) and then standardized to zero moisture content.

The numbers of male flowers were not counted in November 2011, February 2012, November 2012, or February 2013. The measured values for pollen, male flower husks, and male flower biomasses per male flower obtained in November 2016 were used as proxy data for November 2011 and November 2012. Likewise, measurements made in February 2017 were used as proxy data for February 2012 and February 2013. Since the pollen biomasses and ^{137}Cs concentrations were not different between November and the following February, the ^{137}Cs concentrations of male flower husks in November 2011 and November 2012 were calculated from the ^{137}Cs concentrations

Fig. 2. Procedure for collecting *Cryptomeria japonica* var. *japonica* pollen from branches held standing in water-filled crates.
Branches were separately wrapped in bags of glassine paper (used to reduce static electricity) (18 × 38 cm, Rizo, Inc.; http://www.rizo.co.jp/crossingbag.html). The bag openings were closed with cotton and copper wire, after which the branches were transferred to water-filled crates in a laboratory held at *ca*. 23°C during working hours (8:30–17:15) on weekdays. The water level in the crates was maintained over time. Most pollen was released into the bags within 3 weeks.

of male flowers (measured in each November) and pollen (measured in each following February), and the pollen/male flower biomass ratio measured in November 2016. The male flower ^{137}Cs concentration in February 2013 was estimated from (i) the pollen and male flower husk ^{137}Cs concentrations at that time and (ii) the pollen/male flower biomass ratio measured in February 2017. Using these calculated values, the pollen, male flower husk, and male flower ^{137}Cs contents (product of biomass and ^{137}Cs concentration) were estimated for November 2011, February and November 2012, and February 2013.

2.2 Estimating the ^{137}Cs concentration in sugi pollen released in spring during the period 2012–2016

FFPRI and MAFF collected male sugi flowers in the November–December period of every year between 2011 (132 localities in sugi forests) and 2015 (21 localities) in Fukushima Prefecture, and measured the ^{137}Cs concentrations in the tissues (Akama et al. 2013, Akama et al. 2017). Leaves have also been collected since 2012. In our work, we selected 21 localities (Fig. 1, ●) in which the survey continued for 5 years, to identify inter-annual changes in^{137}Cs concentrations. Sugi trees from which samples were collected were not necessarily those sampled in the previous years.

The 21 localities in our study had 14 granite and other plutonic rock types, 3 sedimentary rocks, 2 metamorphic rocks, 1 igneous rock type, and 1 Jurassic accretionary complex (Geological Survey of Japan 2014). The pollen/male flower ^{137}Cs concentration ratio obtained in the Fukushima Prefectural Forestry Research Centre forest was used in the calculations to estimate the pollen ^{137}Cs concentrations over 5 years. A few male flowers dropped during the period from male flower maturity to pollen release, but we did not consider the implications of this phenomenon.

2.3 Transmission of ^{137}Cs to sugi pollen

The ^{137}Cs deposition data after July 2, 2011 in the 21 localities (Fig. 1) were obtained from a distribution map of ^{137}Cs deposition in the region (Ministry of Education, Culture, Sports, Science and Technology, MEXT 2018). The mean value between the upper and lower limits of a given deposition class constituted the deposition value and was used in the analyses. The sugi pollen ^{137}Cs concentration (see **2.2**)/^{137}Cs deposition ratio was determined for each year in the period 2012–2016 to provide ^{137}Cs aggregated transfer factors (T_{ag}s) for ^{137}Cs transmission from the environment to sugi pollen.

Decay corrections for all ^{137}Cs concentrations were made on 1 February 2017. We used an online tool (http://www.civilworks.jp/freetool/freetool.htm#normal) to test the normality of the data distribution.

3. Results

3.1 Biomass and ^{137}Cs concentrations in sugi male flowers and pollen in each November and each following February

Biomass per male flower decreased between each November and each following February (Fig. 3a). Since we assumed the pollen status to be unchanged, the male flower husk biomass therefore decreased. The ratio of pollen biomass to male flower biomass increased as male flowers matured: 0.32 ± 0.05 in November and 0.48 ± 0.06 in February (Table 1) ($P < 0.0001$, $n = 6$). The male flower ^{137}Cs concentration was higher in each February than in each previous November (Fig. 3b) due to the increase in ^{137}Cs concentration in the male flower husk. The increase in the period November 2011–February 2012 was especially large. The quantities of ^{137}Cs per male flower and per male flower husk increased in the period November 2011–February 2012 (Fig. 3c), but were lower in the periods November 2012–February 2013 and November 2016–February 2017. We found no significant difference in the current-year leaf ^{137}Cs concentration between November 2012 and February 2013 (Fig. 3d, Table 1).

Both the concentrations and quantities of ^{137}Cs decreased (Fig. 3b,c; Table 1) year on year. The decline was rapid immediately after the accident. Similar trends in ^{137}Cs concentration occurred in the current-year leaves (Fig. 3d, Table 1).

The ^{137}Cs concentrations were not significantly different between (a) November 2012 male flowers and current-year leaves and (b) February 2013 pollen (paired *t*-tests, $P = 0.152$–0.898, $n = 5$, Fig. 3b,d).

The pollen/male flower ^{137}Cs concentration ratios were high in November and low in the following February (Fig. 4), reflecting the increase in ^{137}Cs concentration in male flower husks from November through the following February. The difference in ^{137}Cs concentration ratio between November and the following February was smaller in later years. The pollen/November male flower ^{137}Cs concentration ratio was *ca*. 1.2 in the first and second years after the FDNPS accident and close to 1.0 in the fifth year (Table 1). Using the duration of time after the accident as a parameter, we constructed a regression model to calculate the pollen/November male flower ^{137}Cs concentration ratio:

$$\text{Pollen/November male flower } ^{137}\text{Cs concentration ratio} = 1.27 \exp(-0.0000895T); (R^2 = 0.9292, P = 0.171, n = 3) \quad (1)$$

where T is the number of days since March 11, 2011.

Since there were so few data points ($n = 3$), the slope of the regression line was not significant, but the coefficient of determination (R^2) was large.

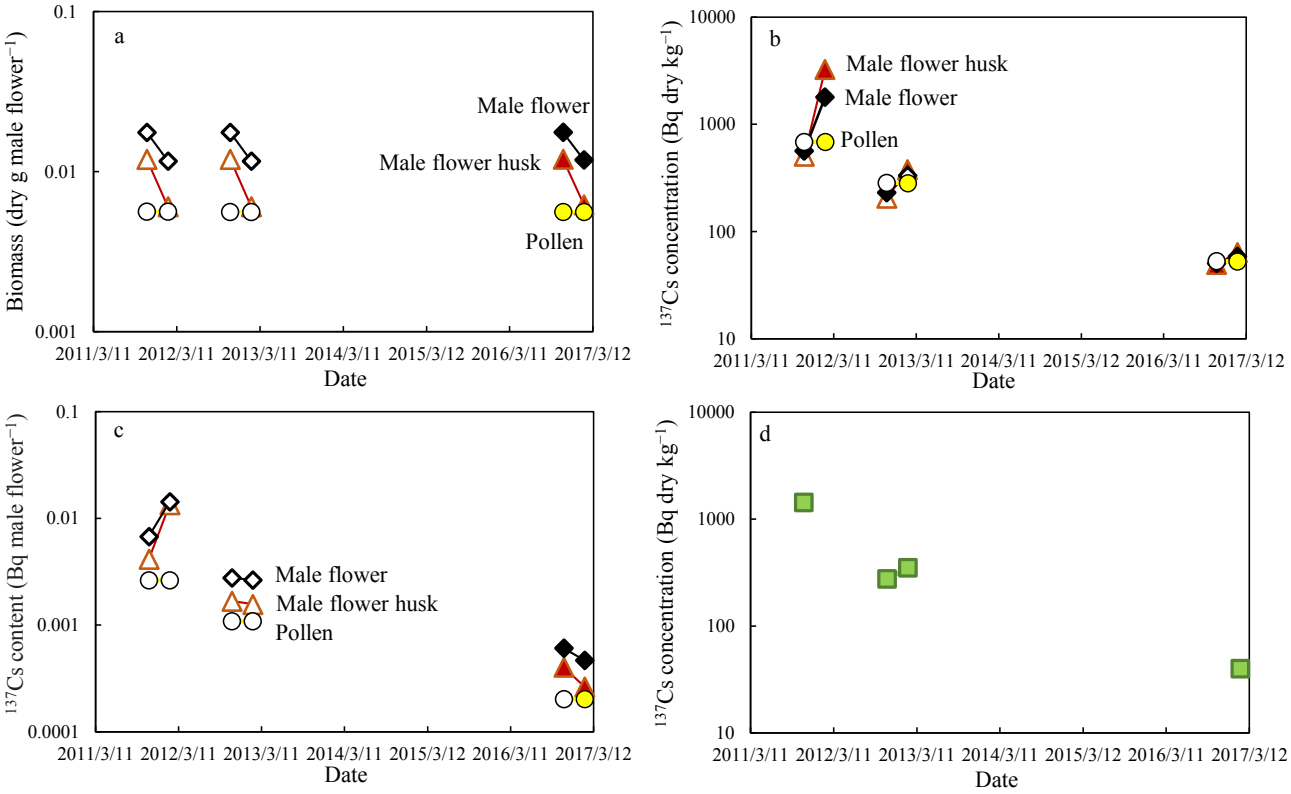

Fig. 3. Temporal changes in (a) biomass, (b) ^{137}Cs concentration, and (c) the contents of ^{137}Cs in pollen, male flower husks, and male flowers; (d) current-year leaf ^{137}Cs concentration in *Cryptomeria japonica* var. *japonica*.
● ○ Pollen, ▲ △ Male flower husk, ◆ ◇ Male flower, ■ Current-year leaf. Closed symbols represent measured values and open symbols are estimates.

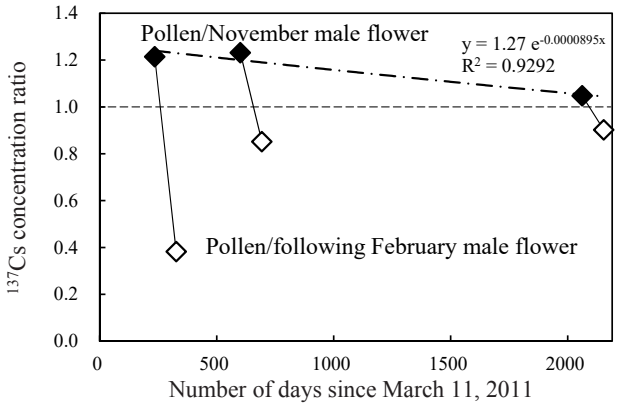

Fig. 4. Temporal changes in *Cryptomeria japonica* var. *japonica* pollen/male flower ^{137}Cs concentration ratios.
◆ Pollen/November male flower ^{137}Cs concentration ratio, ◇ pollen/following February male flower ^{137}Cs concentration ratio.

3.2 Estimating the ^{137}Cs concentration in sugi pollen released in spring during the period 2012–2016

The sugi male flower and current-year leaf ^{137}Cs concentration data for Fukushima Prefecture were not normally distributed in any of the years; the distributions may have been lognormal (D'Agostino-Pearson test, $K^2 < 5.991$, $P > 0.050$; Anderson and Darling test, $A^2 < 0.752$).

When we divided the 21 localities into (a) 11 with high male flower ^{137}Cs concentrations (2,600–130,000 Bq dry kg^{-1}) in the period November–December 2011 and (b) 10 with low ^{137}Cs concentrations (97–2,500 Bq dry kg^{-1}), we found that the logarithmic-mean ^{137}Cs concentrations of male flowers and current-year leaves decreased over time (Fig. 5a,b). The trends were similar between the two categories of localities: the mean concentration decreased exponentially with time. We therefore calculated the logarithmic means and standard deviations (SD) of the male flower ^{137}Cs concentrations across the pooled set of localities (Fig. 6).

We predicted the logarithmic means (± SD) of current-year leaf ^{137}Cs concentrations, the male flower ^{137}Cs concentrations, and the estimated pollen ^{137}Cs concentrations by inserting the male flower ^{137}Cs concentrations into equation (1). The ^{137}Cs concentrations of current-year leaves, male flowers, and pollen declined over time in the November–December periods from 2011 through 2015 (Fig. 6). The ^{137}Cs concentrations in the three plant components varied significantly, except in 2013: male flower < pollen ($P = 0.008$–0.021, $n = 21$), current-year leaf < pollen ($P = 0.006$–0.038, $n = 21$).

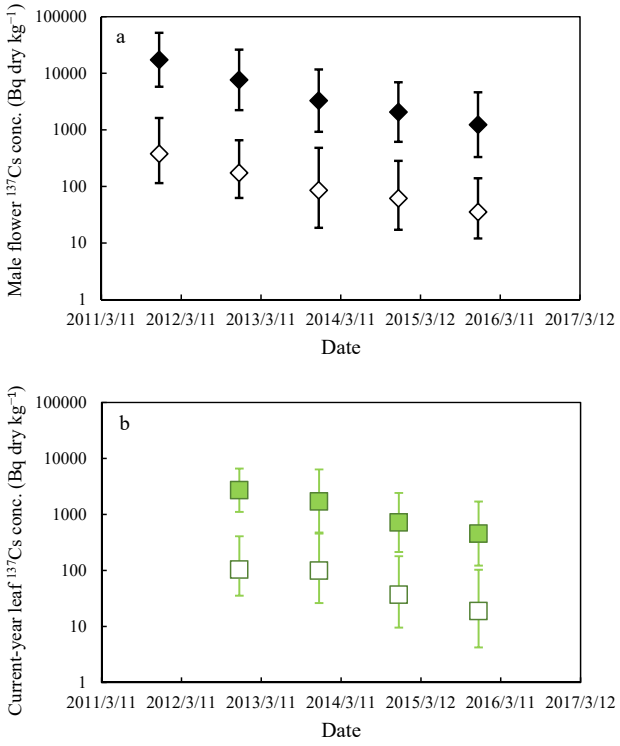

Fig. 5. Inter-annual changes in ^{137}Cs concentrations in (a) male flowers and (b) current-year leaves of *Cryptomeria japonica* var. *japonica*.
The plot provides logarithmic means ± standard deviation (SD) of ^{137}Cs concentrations in flowers and leaves in 11 localities with high male flower ^{137}Cs concentrations (◆ ■), and in 10 localities with low ^{137}Cs concentrations (◇ □).

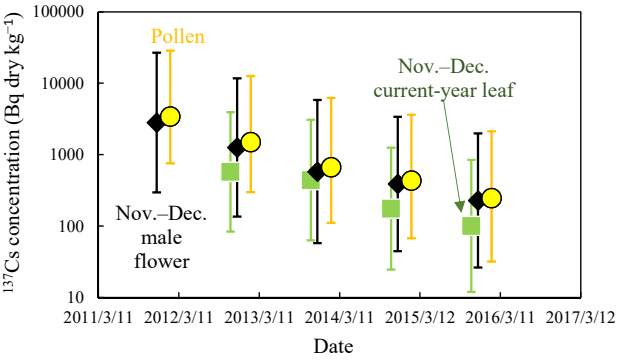

Fig. 6. Inter-annual changes in the ^{137}Cs concentrations in *Cryptomeria japonica* var. *japonica* male flowers, current-year leaves, and pollen.
The plot provides logarithmic means ± SD of ^{137}Cs concentrations in male flowers (◆) and current-year leaves (■) collected in the November–December period, and in pollen (●) collected in the following February in 21 sugi forest localities in Fukushima Prefecture.

3.3 ^{137}Cs aggregated transfer factors (T_{ag}s) for ^{137}Cs transmission from the environment to sugi pollen

The coefficient ^{137}Cs T_{ag} decreased exponentially over time after the accident (Fig. 7). The coefficient of variation (CV) of T_{ag} was unrelated to time since the accident (P = 0.737, n = 5). The relationship between the T_{ag} and the quantity of ^{137}Cs deposited was not significant in any year (P = 0.091–0.567, n = 21). The T_{ag} was not significantly different between plutonic (n = 14) and sedimentary rocks (n = 3) (P = 0.217–0.995), and no trends were detected for igneous rocks (n = 1). The mean (± SD) T_{ag} value was 0.0203 ± 0.0121 m^2 dry kg^{-1} in February 2012 and 0.00168 ± 0.00926 m^2 dry kg^{-1} in February 2016.

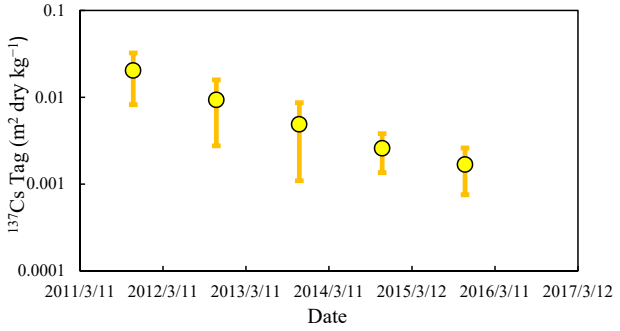

Fig. 7. ^{137}Cs point-in-time aggregated transfer factor (T_{ag}) for ^{137}Cs transmission from the environment to *Cryptomeria japonica* var. *japonica* pollen.
The plot provides logarithmic means ± SD for the pollen ^{137}Cs concentration/deposition (MEXT 2018) ratios across 21 *C. japonica* var. *japonica* forests in Fukushima Prefecture.

4. Discussion

4.1 ^{137}Cs in sugi male flowers during the period between pollen maturity and pollen release

Old leaves and branches of sugi usually die in the fall (Tange et al. 1989, Kaneko et al. 1997). Before leaf death, compounds in the withering leaves are transported to the surviving organs. Since the biomass per male flower was reduced in the periods from November through the following February (Fig. 3a, Table 1), it is likely that material resorption occurred in male flowers[1)] as well as in leaves. The amount of resin in male flowers fell between November and the following February.

In *Osmunda japonica* (Kiyono et al. 2018a), *Petasites japonicus* (Kiyono et al. 2018b), and *Eleutherococcus sciadophylloides* (Akama and Kiyono 2015, Kiyono et al. 2019), the ^{137}Cs tends to remain in dead leaves. This has not been reported for sugi. Potassium and Cs is thought to behave similarly in sugi leaves (Yoshihara et al. 2016); we found that K concentrations in male flowers, female flowers, and leaves were higher than in trunks, branches, and roots (Appendices 1 and 2). Between November and the following February, the male flower K concentration appeared to increase in comparison with other elements, such as N (Appendix 3). We therefore propose that ^{137}Cs likely remained in the dying tissues of sugi (see Fig. 2 in Yoshihara et al. 2016).

The ^{137}Cs concentration of the male flower husks was higher in February than in the previous November (Fig. 3b, Table 1). The increase in concentration was especially large in the period

from November 2011 through the following February. The amount of ^{137}Cs in male flower husks increased over this period (Fig. 3c, Table 1). Since less than a year had passed following the accident, new ^{137}Cs absorption likely occurred through both the roots and above-ground plant surfaces (Mahara et al. 2014, Sekiyama et al. 2016, Wang et al. 2016). Consequently, the quantity of ^{137}Cs in whole individuals, and in leaves that were nutritionally close to the male flower husks, may have increased during that time. ^{137}Cs adhering to the *ante*-3.11 leaves that existed before the accident (Kiyono and Akama 2016) was almost entirely lost in the 3 years following the accident due to rain wash-off and leaf death (Kiyono and Akama 2016, Yoshihara et al. 2016). A proportion of the ^{137}Cs adhering to the leaf surfaces in the November 2011–February 2012 period would have been transferred to the plant body, and probably accounts for increases in the ^{137}Cs concentration in the male flower husks after November. The increases in ^{137}Cs concentration in male flower husks between November and the following February declined over time, becoming negligible in the period from 2016 to 2017, likely because the transfer of new ^{137}Cs from the environment to sugi was reduced. Thus, the concentration in each component of the trees appeared to have stabilized.

Note 1) A similar phenomenon was observed in the fruit of *Akebia trifoliata* (Kiyono and Akama, unpublished data). The moisture content proportion in the fruit was 0.24, and the biomass was 19.6 dry g fruit^{-1}, immediately before the fruit opened. The moisture content proportion of opened fruit was 0.12 and the biomass was 9.7 dry g fruit^{-1}.

4.2 Evaluation of previous assumptions on the quantitative relationship between sugi male flowers and pollen

In a press release issued in late 2011, the MAFF (2011) assumed that pollen biomass accounted for 50% of male flower biomass. According to our findings, this was an overestimate for male flowers in the November–December period, when pollen accounted for about one third of flower biomass (Table 1). However, the assumption of the MAFF that the ^{137}Cs concentration in sugi pollen was the same as the concentration in male flowers during these months was reasonable (Table 1). Thus, the amount of ^{137}Cs contained in airborne pollen was 60–70% of the value proposed by the MAFF (2011).

4.3 Relationship between ^{137}Cs concentrations in sugi male flowers and pollen in November

We found no significant difference in ^{137}Cs concentration between male flowers and pollen in November (Fig. 3b, Table 1). Since pollen becomes nutritionally independent in the male flower by November, the ^{137}Cs concentrations in male flower husks and pollen were considered to be similar. After the male flowers had matured, and as the resorption of materials from male flower husks to leaves progressed, the difference in ^{137}Cs concentration between pollen and male flower husks widened. We applied equation (1) (using values for male flowers in November in Fig. 6) to the male flower data for the November–December period. The pollen concentration predicted by equation (1) may be an overestimation. The coldness of winter is related to the breaking of dormancy, and warming after the dormancy break is related to the subsequent maturation of male flowers (Kanazashi et al. 2016). Since some male flowers might end dormancy in the November–December period, the rate of subsequent maturation depends to some extent on weather conditions. Hence, the pollen concentrations in Figs. 6 and 7 incorporate uncertainties resulting from differences in meteorological conditions in the sugi habitat and the year in which male flowers bloomed.

4.4 Time since the accident and the radiocesium concentration in sugi pollen

The concentration of ^{137}Cs in the current-year leaves (Appendix 4) and male flowers declined over the years after the accident (Akama et al. 2017). We found that the decline in concentration in mature pollen was similar to that in male flowers (Fig. 6). The supply of ^{137}Cs can vary by habitat soil (Saunton et al. 2003, Salt et al. 2004, Yamaguchi 2014) and geology, as found for *Eleutherococcus sciadophylloides* (Kiyono et al. 2019): shoot ^{137}Cs concentrations become higher in habitats with plutonic (Kawamata) and igneous rocks (Kaneyama) than in those with sedimentary rocks (Hirono). However, we did not detect similar habitat effects on sugi pollen in our study. The pollen ^{137}Cs concentration in February 2016 was 8% of that in February 2012 (Fig. 6). Differences in the initial ^{137}Cs concentration had little effect on the rate of decrease over time (Fig. 5). It was predicted that over 90% of ^{137}Cs in *ante-3.11* leaves would be removed within 3 years of the FDNPS accident (Kiyono and Akama 2016). Transfer from ^{137}Cs deposits in bark would have continued, even after transfer from the leaves over the short term had decreased. However, within 3 years of the FDNPS accident, the amount of ^{137}Cs in bark was less than in leaves (Kajimoto et al. 2015) and the amounts of ^{137}Cs transferred from bark would therefore be relatively small. By about March 2014, root absorption would have been the main supply source of ^{137}Cs to the plant body of sugi. The ^{137}Cs distribution in sugi wood approached an equilibrium state within 3 years of the accident (Ogawa et al. 2016), but such equilibrium state was not observed in the same period in some other sugi forests (Imamura et al. 2017, Ohashi et al. 2017).

The decrease in ^{137}Cs concentration in male flowers and pollen matched the decrease (Mahara et al. 2014, Imamura et al. 2017) in sugi leaf ^{137}Cs concentrations. However, pollen production varies greatly from year to year; summer weather conditions in

the two preceding years greatly influence the amount of pollen produced (Takahashi et al. 1996, Takahashi and Kawashima 1999, Kiyono 2010). The amount of pollen captured in Fukushima City between 2012 and 2016 was highest in 2013 (Hanako, Ministry of the Environment 2016), but we found no clear trends in the ^{137}Cs concentrations in sugi male flowers and pollen in 2013 (Figs. 3–7). The influence of fluctuations in pollen production on pollen ^{137}Cs concentration was not determined in detail in our study (see Appendix 3).

We found no significant difference in the current-year leaf ^{137}Cs concentration between November and the following February (Fig. 3d, Table 1). During this period, the current-year leaf ^{137}Cs concentration appeared more stable than the concentration in male flowers. The current-year leaf ^{137}Cs concentration in sugi forest was higher than that of pollen in November 2011, but the concentrations were not significantly different after November 2012 (Fig. 3b,d; Table 1). However, data obtained in our broad area survey over 21 localities in Fukushima prefecture showed that ^{137}Cs concentrations were low in the current year leaves in most cases since November 2012 (Figs. 5 and 6). Since concentrations in current year leaves and pollen showed similar trends over the years, the current year leaf ^{137}Cs concentration in November may be useful for estimating the ^{137}Cs concentration in pollen released in the following February. Kanasashi et al. (2015) found that within the period from November 2012 to February 2013, in regions partially overlapping those included in our study and extending over a wide area of eastern Japan (mostly in Fukushima Prefecture), the ^{137}Cs concentration in leaves was lower than concentrations in male flowers and pollen. However, the ^{137}Cs concentration at the tips of the leaves that produced male flowers was higher than the concentrations in male flowers and pollen. ^{137}Cs concentrations differ among leaf parts (Burger and Lichtscheidl 2017). Studies of leaf parts that could be used as an appropriate indicator of pollen ^{137}Cs concentration should be undertaken in future investigations.

4.5 ^{137}Cs aggregated transfer factors (T_{ag}s) for ^{137}Cs transmission from the environment to sugi pollen

The ^{137}Cs T_{ag} declined year on year from February 2012 through February 2016, and had not reached an equilibrium state by the fifth year after the FDNPS accident (Fig. 7). After the Chernobyl accident, it took 5–10 years for the movement of ^{137}Cs dispersed among trees and soils of the polluted forests to stabilize (IAEA 2009). The coefficients calculated for the unbalanced stage were accompanied by large errors (Yoshida 2012). Therefore, the values in Fig. 7 are temporal and applicable only to the relevant year. However, there were no significant inter-annual trends in ^{137}Cs T_{ag} within a deposition range of 5–4,500 kBq m^{-2} (as of July 2, 2011). Since the data on ^{137}Cs deposition covered almost all sugi forests in eastern Japan, including Fukushima Prefecture, pollen ^{137}Cs concentrations in eastern Japan can be estimated from ^{137}Cs deposition data in the Extension Site of Distribution Map of Radiation Dose, etc.,/GSI Maps (MEXT 2018) and the T_{ag} for the relevant year (Fig. 7). Multiplication of these data and pollen biomass (about ⅓ of the male flower biomass) provides an estimate of the ^{137}Cs content of sugi pollen in a single year over a wide area before pollen release. For example, sugi pollen released in spring 2016 from a sugi forest where the ^{137}Cs deposition was 800 kBq m^{-2} on July 2, 2011 was calculated to contain *ca*. 1,180 Bq dry kg^{-1} of ^{137}Cs based on the known ^{137}Cs deposition (704 kBq m^{-2} after attenuation correction) and a T_{ag} of 0.00168 m^2 dry kg^{-1} as of 2016.

5. Conclusions

There are few data on ^{137}Cs in flower organs. Based on the measurements made on male flowers before pollen release, we were able to more precisely estimate the amount of ^{137}Cs released by sugi pollen than previous investigations. It might be possible to estimate pollen ^{137}Cs concentration from the current-year leaf ^{137}Cs concentration, but this will require rigorous verification in future studies. The transfer of ^{137}Cs from the environment to sugi pollen had not stabilized 5 years after the accident, and further monitoring will be required to determine the ^{137}Cs T_{ag} value in the equilibrium state. The ^{137}Cs concentration of sugi pollen should be lower in the future, but pollen production fluctuates year on year, and the ^{137}Cs mass released by pollen fluctuates accordingly. The relationship between sugi pollen production and pollen ^{137}Cs content is unknown and awaits future investigations.

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Appendix 1. C, N, P, K, Ca, Mg, Na, and Mn concentrations in organs of *Cryptomeria japonica* var. *japonica* and *Chamaecyparis obtusa*.

In July 2001, trees in the young forests of *Cryptomeria japonica* var. *japonica* (sugi) and *Chamaecyparis obtusa* (hinoki) (Table A1-1) in the nursery of the Chiyoda Experimental Station of FFPRI in Kasumigaura City, Ibaraki Prefecture were divided into three size classes per species. A sample tree was dug out from each compartment containing two species and trees of three size classes. Simultaneously, GA_3 (gibberellic acid) was sprayed on the leaves to promote flowering in some of the live standing trees; similar numbers of trees were used as controls. In the period February–March 2002, three sprayed trees and three control trees in three size classes of both species were sampled. The sampled trees were divided into organs: male flowers, female flowers (since male and female hinoki flowers were difficult to distinguish, they were combined into a single flower category), leaves, branches, stems, and roots. The concentrations of C, N, P, K, Ca, Mg and Mn were measured (Table A1-2, Vegetation Control Laboratory, FFPRI). A CN elemental analyzer was used to measure C and N; P was measured by inductively coupled plasma emission spectroscopy (ICP); flame photometry was used to measure K and Mn; atomic absorption spectrometry was used to measure Ca and Mg (Ringyo Kagaku Gijutsu Shinkosho).

Table A. 1-1. General description of surveyed trees.

Sampling date	Species	Treatment	DBH cm	Tree height m	Tree biomass dry kg tree^{-1}
2001.7	Sugi		2.7 ± 0.8	2.9 ± 0.7	2.8 ± 1.3
2002.2	Sugi	2001.7 GA_3	2.9 ± 0.8	3.2 ± 0.6	4.1 ± 1.4
2002.2	Sugi		3.1 ± 0.9	3.1 ± 0.6	4.3 ± 2.0
2001.7	Hinoki		2.0 ± 0.3	2.7 ± 0.2	3.3 ± 0.6
2002.3	Hinoki	2001.7 GA_3	2.8 ± 0.5	2.9 ± 0.9	3.5 ± 1.1
2002.3	Hinoki		2.6 ± 0.3	2.8 ± 1.5	3.5 ± 0.7

Values are mean ± SD. Sugi, *Cryptomeria japonica* var. *japonica*. Hinoki, *Chamaecyparis obtusa*. DBH, diameter at breast height.

Table A1-2. C, N, P, K, Ca, Mg, Na, and Mn concentrations in organs of *Cryptomeria japonica* var. *japonica* and *Chamaecyparis obtusa*.

Sampling date	Species	GA_3 treatment	Organ	Sample trees	C %	N %	P %	K %	Ca %	Mg %	Na %	Mn %
2002.2	Sugi	2001.7	Male flower	3	51.8 ± 1.0	0.74 ± 0.04	0.20 ± 0.01	0.84 ± 0.04	0.50 ± 0.06	0.20 ± 0.05	0.0050 ± 0.0010	0.0036 ± 0.0007
2002.2	Sugi	-	Male flower	1	52.6	0.74	0.17	0.91	0.37	0.17	0.085	0.0025
2002.2	Sugi	2001.7	Female flower	3	49.3 ± 1.1	1.15 ± 0.13	0.27 ± 0.01	0.95 ± 0.08	1.23 ± 0.34	0.30 ± 0.05	0.0026 ± 0.0006	0.0057 ± 0.0004
2002.2	Sugi	-	Female flower	3	49.7 ± 0.3	1.04 ± 0.59	0.18 ± 0.13	0.57 ± 0.38	0.91 ± 0.30	0.20 ± 0.12	0.0079 ± 0.0027	0.0034 ± 0.0012
2001.7	Sugi	-	Leaf	3	52.3 ± 0.6	0.97 ± 0.04	0.19 ± 0.08	0.64 ± 0.19	1.13 ± 0.12	0.18 ± 0.02	0.0054 ± 0.0011	0.0024 ± 0.0003
2002.2	Sugi	2001.7	Leaf	3	51.1 ± 0.4	0.78 ± 0.07	0.22 ± 0.03	0.69 ± 0.13	1.06 ± 0.20	0.20 ± 0.02	0.0012 ± 0.0007	0.0019 ± 0.0004
2002.2	Sugi	-	Leaf	3	51.3 ± 0.2	0.80 ± 0.01	0.22 ± 0.03	0.65 ± 0.08	1.20 ± 0.19	0.19 ± 0.03	0.0045 ± 0.0005	0.0020 ± 0.0001
2001.7	Sugi	-	Branch	3	49.8 ± 0.5	0.47 ± 0.05	0.069 ± 0.020	0.29 ± 0.07	0.90 ± 0.29	0.103 ± 0.050	0.0004 ± 0.0006	0.0008 ± 0.0001
2002.2	Sugi	2001.7	Branch	3	49.5 ± 0.3	0.44 ± 0.03	0.050 ± 0.005	0.22 ± 0.02	0.73 ± 0.10	0.086 ± 0.013	0.0047 ± 0.0027	0.0007 ± 0.00003
2002.2	Sugi	-	Branch	3	49.9 ± 0.3	0.47 ± 0.02	0.059 ± 0.006	0.28 ± 0.03	0.78 ± 0.05	0.084 ± 0.013	0.0023 ± 0.0040	0.0009 ± 0.00005
2001.7	Sugi	-	Stem	3	50 ± 1	0.34 ± 0.1	0.023 ± 0.001	0.16 ± 0.03	0.35 ± 0.06	0.038 ± 0.004	ND	0.00056 ± 0.00015
2002.2	Sugi	2001.7	Stem	3	50 ± 0.2	0.34 ± 0.02	0.022 ± 0.01	0.12 ± 0.02	0.38 ± 0.03	0.041 ± 0.007	ND	0.00051 ± 0.00005
2002.2	Sugi	-	Stem	3	50 ± 0.3	0.34 ± 0.02	0.028 ± 0.01	0.11 ± 0.01	0.43 ± 0.04	0.039 ± 0.002	ND	0.00053 ± 0.00007
2001.7	Sugi	-	Root	3	49.2 ± 0.1	0.35 ± 0.04	0.032 ± 0.003	0.23 ± 0.02	0.49 ± 0.10	0.057 ± 0.008	0.0067 ± 0.0027	0.0040 ± 0.0008
2002.2	Sugi	2001.7	Root	3	49.6 ± 0.3	0.33 ± 0.03	0.024 ± 0.008	0.13 ± 0.05	0.45 ± 0.05	0.048 ± 0.005	0.0058 ± 0.0020	0.0022 ± 0.0004
2002.2	Sugi	-	Root	3	50.5 ± 1.1	0.68 ± 0.36	0.119 ± 0.097	0.41 ± 0.31	0.77 ± 0.35	0.110 ± 0.010	0.0040 ± 0.0037	0.0041 ± 0.0034
2002.3	Hinoki	2001.7	Flower	2	51 ± 2	2.1 ± 0.7	0.36 ± 0.14	0.84 ± 0.29	0.57 ± 0.09	0.17 ± 0.02	0.0057 ± 0.0011	0.0051 ± 0.0019
2002.3	Hinoki	-	Flower	2	49.9 ± 0.01	1.6 ± 0.04	0.31 ± 0.06	0.64 ± 0.12	0.89 ± 0.03	0.20 ± 0.05	0.0097 ± 0.0114	0.0097 ± 0.0029
2001.7	Hinoki	-	Leaf	3	52.2 ± 0.6	1.21 ± 0.27	0.16 ± 0.02	0.59 ± 0.08	1.1 ± 0.3	0.17 ± 0.02	0.0052 ± 0.0019	0.0114 ± 0.0011
2002.3	Hinoki	2001.7	Leaf	3	51.0 ± 0.5	1.01 ± 0.09	0.23 ± 0.11	0.53 ± 0.06	1.1 ± 0.2	0.15 ± 0.01	0.0037 ± 0.0030	0.0073 ± 0.0002
2002.3	Hinoki	-	Leaf	3	51.2 ± 1.0	0.86 ± 0.06	0.20 ± 0.08	0.51 ± 0.08	1.2 ± 0.3	0.14 ± 0.02	0.0028 ± 0.0010	0.0098 ± 0.0013
2001.7	Hinoki	-	Branch	3	50.6 ± 0.3	0.42 ± 0.02	0.051 ± 0.012	0.24 ± 0.01	0.72 ± 0.16	0.045 ± 0.006	ND	0.0020 ± 0.0003
2002.3	Hinoki	2001.7	Branch	3	50.1 ± 0.1	0.47 ± 0.02	0.048 ± 0.009	0.18 ± 0.02	0.76 ± 0.09	0.038 ± 0.001	0.0067 ± 0.0017	0.0015 ± 0.0002
2002.3	Hinoki	-	Branch	3	50.4 ± 0.2	0.44 ± 0.02	0.041 ± 0.007	0.16 ± 0.01	0.81 ± 0.05	0.039 ± 0.001	0.0080 ± 0.0067	0.0021 ± 0.0006
2001.7	Hinoki	-	Stem	3	49.8 ± 0.3	0.33 ± 0.02	0.024 ± 0.007	0.15 ± 0.01	0.33 ± 0.04	0.020 ± 0.003	ND	0.00012 ± 0.00005
2002.3	Hinoki	2001.7	Stem	3	49.6 ± 0.1	0.39 ± 0.01	0.046 ± 0.006	0.13 ± 0.02	0.42 ± 0.04	0.025 ± 0.001	0.0007 ± 0.0012	0.0012 ± 0.0002
2002.3	Hinoki	-	Stem	3	49.9 ± 0.2	0.35 ± 0.03	0.034 ± 0.005	0.12 ± 0.02	0.45 ± 0.08	0.022 ± 0.001	ND	0.0015 ± 0.0001
2001.7	Hinoki	-	Root	3	50.2 ± 0.3	0.40 ± 0.05	0.050 ± 0.015	0.21 ± 0.03	0.39 ± 0.10	0.070 ± 0.023	0.0053 ± 0.0021	0.0071 ± 0.0045
2002.3	Hinoki	2001.7	Root	3	49.9 ± 0.1	0.35 ± 0.01	0.036 ± 0.011	0.10 ± 0.02	0.31 ± 0.04	0.036 ± 0.005	0.0038 ± 0.0021	0.0025 ± 0.0002
2002.3	Hinoki	-	Root	3	49.5 ± 0.2	0.36 ± 0.01	0.041 ± 0.010	0.12 ± 0.03	0.37 ± 0.04	0.046 ± 0.002	0.0025 ± 0.0002	0.0036 ± 0.0008

Values are mean ± SD. Sugi, *Cryptomeria japonica* var. *japonica*. Hinoki, *Chamaecyparis obtusa*. GA3 treatment, sprayed with gibberellic acid.

Appendix 2. C, N, P, K, Ca, Mg, and Mn concentrations in organs of *Cryptomeria japonica* var. *japonica* (Bokasugi clone).

GA_3 was sprayed on the leaves (in July 2002, 2003, and 2004) of some of the live standing trees in a *Cryptomeria japonica* var. *japonica* (Bokasugi clone) forest (Kiyono et al. 2006) established in April 2002 in the nursery of FFPRI in Tsukuba City, Ibaraki Prefecture. The spray was applied to promote flowering (Table A2-1). Twelve individuals in February 2004 and ten individuals in February 2005 were harvested and separated into organs: male flowers, female flowers, leaves, branches, stems, and roots. The concentrations of C, N, P, K, Ca, Mg and Mn elements were measured for each organ (Table A2-2, Vegetation Control Laboratory, FFPRI). A CN elemental analyzer was used to measure C and N; P was measured by ICP; flame photometry was used to measure K and Mn; atomic absorption spectrometry was used to measure Ca and Mg (Ringyo Kagaku Gijutsu Shinkosho). A negligible number of flowers formed after the

GA_3 treatment in July 2004. The mean monthly temperature and monthly rainfall during the test period differed little from normal values, although precipitation in October 2004 was especially heavy.

Table A2-1. Schedule of GA_3 (gibberellic acid) spraying and subsequent sampling of *Cryptomeria japonica* var. *japonica* (Bokasugi clone) trees.

Abbrev.	2002.7 Treatment	2003.7 Treatment	2004.2 Sample trees	2004.7 Treatment	2005.2 Sample trees
-2-/4	GA_3		2		
--3/4		GA_3	4		
-23/4	GA_3	GA_3	2		
--- /4			4		
-2--/5	GA_3				1
--3-/5		GA_3			2
---4/5				GA_3	2
-2-4/5	GA_3			GA_3	1
--34/5		GA_3		GA_3	2
---- /5					2

Table A2-2. C, N, P, K, Ca, Mg, and Mn concentrations in organs of *Cryptomeria japonica* var. *japonica* (Bokasugi clone).

Sampling date	GA_3 treatment	Organ	Sample trees	C %	N %	P %	K %	Ca %	Mg %	Mn %
2004.2	2002.7	Root	2	50 ± 1	0.64 ± 0.12	0.069 ± 0.007	0.25 ± 0.01	0.46 ± 0.04	0.051 ± 0.008	0.0025 ± 0.0020
2004.2	2003.7	Root	4	50 ± 1	0.61 ± 0.04	0.067 ± 0.013	0.27 ± 0.10	0.52 ± 0.08	0.055 ± 0.006	0.0022 ± 0.0011
2004.2	2002.7, 2003.7	Root	2	49 ± 1	0.68 ± 0.12	0.068 ± 0.003	0.27 ± 0.02	0.53 ± 0.07	0.058 ± 0.005	0.0027 ± 0.0008
2004.2	-	Root	4	50 ± 1	0.64 ± 0.13	0.067 ± 0.013	0.28 ± 0.05	0.46 ± 0.04	0.057 ± 0.010	0.0027 ± 0.0013
2004.2	2003.7	Male flower	4	55 ± 0.4	1.3 ± 0.2	0.19 ± 0.02	0.92 ± 0.28	0.42 ± 0.20	0.10 ± 0.02	0.0015 ± 0.0001
2004.2	2002.7, 2003.7	Male flower	2	54 ± 0.2	1.3 ± 0.2	0.17 ± 0.02	0.97 ± 0.12	0.40 ± 0.03	0.11 ± 0.01	0.0018 ± 0.0001
2004.2	2003.7	Female flower	4	51 ± 0.5	1.7 ± 0.2	0.21 ± 0.02	0.89 ± 0.24	0.77 ± 0.11	0.14 ± 0.01	0.0026 ± 0.0005
2004.2	2002.7, 2003.7	Female flower	2	51 ± 0.2	1.8 ± 0.1	0.18 ± 0.01	0.83 ± 0.39	0.52 ± 0.27	0.12 ± 0.03	0.0021 ± 0.0007
2004.2	2002.7	Leaf	2	51 ± 0.05	1.58 ± 0.07	0.131 ± 0.022	0.78 ± 0.16	1.22 ± 0.04	0.090 ± 0.010	0.0021 ± 0.0004
2004.2	2003.7	Leaf	4	51 ± 0.3	1.48 ± 0.10	0.132 ± 0.013	0.89 ± 0.23	1.07 ± 0.05	0.068 ± 0.011	0.0018 ± 0.0001
2004.2	2002.7, 2003.7	Leaf	2	51 ± 0.1	1.50 ± 0.03	0.132 ± 0.027	0.80 ± 0.13	1.15 ± 0.22	0.088 ± 0.017	0.0016 ± 0.0004
2004.2	-	Leaf	4	52 ± 0.2	1.46 ± 0.12	0.115 ± 0.003	0.75 ± 0.14	1.14 ± 0.16	0.091 ± 0.014	0.0020 ± 0.0003
2004.2	2002.7	Branch	2	50 ± 0.3	0.98 ± 0.01	0.078 ± 0.010	0.47 ± 0.12	0.88 ± 0.07	0.060 ± 0.012	0.0010 ± 0.0001
2004.2	2003.7	Branch	4	50 ± 0.3	0.89 ± 0.15	0.073 ± 0.004	0.45 ± 0.09	0.80 ± 0.10	0.051 ± 0.010	0.0009 ± 0.0001
2004.2	2002.7, 2003.7	Branch	2	50 ± 0.5	0.86 ± 0.15	0.073 ± 0.002	0.45 ± 0.09	0.84 ± 0.03	0.057 ± 0.003	0.0010 ± 0.0001
2004.2	-	Branch	4	51 ± 0.2	0.86 ± 0.08	0.070 ± 0.014	0.42 ± 0.09	0.78 ± 0.12	0.066 ± 0.014	0.0010 ± 0.0002
2004.2	2002.7	Stem	2	50 ± 1	0.51 ± 0.12	0.053 ± 0.017	0.25 ± 0.05	0.47 ± 0.07	0.036 ± 0.004	0.00055 ± 0.00023
2004.2	2003.7	Stem	4	50 ± 1	0.51 ± 0.06	0.048 ± 0.007	0.22 ± 0.04	0.46 ± 0.08	0.034 ± 0.005	0.00065 ± 0.00022
2004.2	2002.7, 2003.7	Stem	2	50 ± 1	0.43 ± 0.01	0.033 ± 0.003	0.15 ± 0.06	0.45 ± 0.07	0.029 ± 0.007	0.00055 ± 0.00016
2004.2	-	Stem	4	50 ± 1	0.47 ± 0.06	0.041 ± 0.007	0.19 ± 0.03	0.41 ± 0.04	0.036 ± 0.004	0.00059 ± 0.00015
2005.2	2002.7	Leaf	1	50	1.29	0.091	0.66	1.30	0.088	0.0018
2005.2	2003.7	Leaf	2	50 ± 0.3	1.14 ± 0.31	0.122 ± 0.023	0.76 ± 0.07	1.11 ± 0.08	0.122 ± 0.026	0.0019 ± 0.0001
2005.2	2004.7	Leaf	2	50 ± 0.2	1.29 ± 0.04	0.107 ± 0.005	0.80 ± 0.03	1.39 ± 0.11	0.077 ± 0.005	0.0020 ± 0.0002
2005.2	2002.7, 2004.7	Leaf	1	50	1.34	0.099	0.74	1.24	0.073	0.0015
2005.2	2003.7, 2004.7	Leaf	2	50 ± 0.1	1.41 ± 0.10	0.116 ± 0.008	0.85 ± 0.02	1.23 ± 0.23	0.112 ± 0.056	0.0022 ± 0.0010
2005.2	-	Leaf	2	50 ± 0.3	1.35 ± 0.05	0.101 ± 0.008	0.84 ± 0.06	1.08 ± 0.09	0.112 ± 0.030	0.0017 ± 0.0001
2005.2	2002.7	Branch	1	49	0.54	0.044	0.29	0.77	0.041	0.00067
2005.2	2003.7	Branch	2	49 ± 0.2	0.43 ± 0.17	0.040 ± 0.003	0.31 ± 0.02	0.73 ± 0.002	0.050 ± 0.020	0.00068 ± 0.00016
2005.2	2004.7	Branch	2	45 ± 5	0.44 ± 0.06	0.039 ± 0.0002	0.29 ± 0.05	0.78 ± 0.06	0.037 ± 0.001	0.00057 ± 0.00001
2005.2	2002.7, 2004.7	Branch	1	49	0.49	0.038	0.28	0.76	0.039	0.00055
2005.2	2003.7, 2004.7	Branch	2	49 ± 0.3	0.53 ± 0.01	0.040 ± 0.002	0.30 ± 0.03	0.73 ± 0.01	0.053 ± 0.016	0.00070 ± 0.00010
2005.2	-	Branch	2	49 ± 0.1	0.50 ± 0.04	0.036 ± 0.001	0.27 ± 0.04	0.69 ± 0.10	0.047 ± 0.011	0.00063 ± 0.00006
2005.2	2002.7	Stem	1	50	0.35	0.032	0.20	0.35	0.024	0.00060
2005.2	2003.7	Stem	2	50 ± 0.3	0.29 ± 0.09	0.026 ± 0.006	0.18 ± 0.02	0.32 ± 0.03	0.028 ± 0.008	0.00053 ± 0.00011
2005.2	2004.7	Stem	2	50 ± 0.2	0.29 ± 0.002	0.028 ± 0.003	0.16 ± 0.01	0.36 ± 0.08	0.024 ± 0.004	0.00059 ± 0.00003
2005.2	2002.7, 2004.7	Stem	1	50	0.39	0.036	0.21	0.36	0.026	0.00073
2005.2	2003.7, 2004.7	Stem	2	50 ± 1	0.34 ± 0.06	0.030 ± 0.007	0.19 ± 0.07	0.38 ± 0.05	0.032 ± 0.010	0.00062 ± 0.000001
2005.2	-	Stem	2	50 ± 0.4	0.35 ± 0.01	0.029 ± 0.003	0.18 ± 0.01	0.38 ± 0.01	0.028 ± 0.002	0.00060 ± 0.00017
2005.2	2002.7	Root	1	49	0.41	0.044	0.28	0.40	0.039	0.00226
2005.2	2003.7	Root	2	50 ± 0.3	0.33 ± 0.12	0.031 ± 0.008	0.19 ± 0.03	0.38 ± 0.04	0.039 ± 0.005	0.00143 ± 0.00053
2005.2	2004.7	Root	2	49 ± 0.2	0.33 ± 0.14	0.035 ± 0.015	0.25 ± 0.09	0.45 ± 0.003	0.038 ± 0.003	0.00189 ± 0.00005
2005.2	2002.7, 2004.7	Root	1	49	0.47	0.043	0.24	0.48	0.041	0.00173
2005.2	2003.7, 2004.7	Root	2	49 ± 1	0.35 ± 0.02	0.032 ± 0.005	0.20 ± 0.02	0.35 ± 0.003	0.033 ± 0.001	0.00144 ± 0.00027
2005.2	-	Root	2	49 ± 0.2	0.32 ± 0.005	0.028 ± 0.003	0.18 ± 0.02	0.33 ± 0.09	0.031 ± 0.001	0.00166 ± 0.00014

Values are mean ± SD. GA_3 treatment, sprayed with gibberellic acid.

Appendix 3. Temporal changes in C, N, P, K, Ca, Mg, and Mn contents in *Cryptomeria japonica* var. *japonica* male flowers.

We sampled *Cryptomeria japonica* var. *japonica* (Kurakake 3 clone) in a forest established in April 2002 at the FFPRI nursery in Tsukuba City, Ibaraki Prefecture. Four trees of similar size were selected and male flowers were collected on February 14, 2008, November 21, 2008, and February 11, 2009. We measured concentrations of C, N, P, K, Ca, Mg, and Mn in the flowers. A CN elemental analyzer was used to measure C and N; P was measured by ICP; flame photometry was used to measure K and Mn; atomic absorption spectrometry was used to measure Ca and Mg (Ringyo Kagaku Gijutsu Shinkosho). The concentration ratios for February 2009/November 2008 were 0.99 ± 0.24 [mean ± SD] (n = 4) for N, 0.96 ± 0.24 (n = 4) for P, 1.06 ± 0.23 (n = 4) for Ca, 0.94 ± 0.11 (n = 4) for Mg, and 0.79 ± 0.27 (n = 4) for Mn. Concentrations were similar between November 2008 and February 2009. The ratio for K (1.25 ± 0.21 n = 4) exceeded those for the other elements; the February K concentration exceeded the November K concentration in all four individuals tested (Table A3-1). The concentration ratios for N and K were significantly different (paired t-test, P = 0.043, n = 4), but there were no other significant pairwise differences, likely because sample sizes were small. Compared to the other elements, the K content was maintained at relatively high levels throughout the pollen dispersal season (February). The February K contents were not significantly different between 2008 and 2009 (paired t-test, P = 0.160, n = 4). The Kurakake 3 clone produced a relatively large biomass of male flowers in every year. The quantity of male flowers was higher in 2008 than in 2009 (Fig. A3-1), corresponding to the increase/decrease pattern in airborne cedar pollen concentration (MAFF 2011) in the Kanto region. The data are insufficient to explain how the annual fluctuation in pollen affects the nutritional status of pollen.

Fig. A3-1. Monitored branch with leaves forming male flowers.
Left 2007.12.31, right 2008.11.21.

Table A3-1. Temporal changes in *Cryptomeria japonica* var. *japonica* cv. Kurakake 3 male flower C, N, P, K, Ca, Mg, and Mn contents. conc. ratio refers to the concentration ratios for 11 February 2009/21 November 2008.

Sampling date	Tree no.	C %	N %	P %	K %	Ca %	Mg %	Mn %
2008.2.14	1	53.8	0.85	0.098	0.293	0.204	0.076	0.0023
2008.2.14	2	53.8	0.86	0.104	0.342	0.166	0.071	0.0010
2008.2.14	3	52.6	0.88	0.133	0.348	0.193	0.068	0.0013
2008.2.14	4	53.4	0.86	0.155	0.481	0.148	0.065	0.0010
2008.11.21	1	54.6	0.78	0.145	0.251	0.163	0.056	0.0015
2008.11.21	2	54.9	0.93	0.199	0.318	0.105	0.055	0.0010
2008.11.21	3	55.1	0.89	0.169	0.233	0.078	0.057	0.0006
2008.11.21	4	54.4	0.82	0.153	0.259	0.128	0.060	0.0015
2009.2.11	1	54.9	0.78	0.178	0.294	0.159	0.057	0.0012
2009.2.11	2	54.3	0.88	0.135	0.332	0.145	0.058	0.0007
2009.2.11	3	55.2	0.78	0.144	0.290	0.082	0.051	0.0006
2009.2.11	4	54.0	0.92	0.162	0.396	0.107	0.049	0.0007
2009.2.11/2008.11.21 conc. ratio	Mean ± SD	1.00 ± 0.01	0.99 ± 0.10	0.96 ± 0.24	1.25 ± 0.21	1.06 ± 0.23	0.94 ± 0.11	0.79 ± 0.27

Appendix 4. The quantity of ^{137}Cs per *Cryptomeria japonica* var. *japonica* leaves in 21 forests within Fukushima Prefecture after the Fukushima Daiichi Nuclear Power Station accident.

In 2016, we divided sugi leaves collected at Iwaki (Fig. 1) by their birth year. The ^{137}Cs concentrations in sugi male flowers and leaves collected at Iwaki have been measured annually since 2011. We measured the numbers, lengths, and dry weights of leaves collected in November 2016. Assuming (i) that the measured values can be applied to samples obtained in the period 2011–2015, and to the other 20 localities, and (ii) that the leaf ^{137}Cs concentration in 2011 (Bq dry kg^{-1}) can be estimated from the male flower ^{137}Cs concentration in 2011 (Bq dry kg^{-1}) using an unpublished regression by the Forestry and Forest Products Research Institute (FFPRI) based on the data from five prefectures between Nagano and Fukushima: Leaf ^{137}Cs concentration in 2011 = 0.3915 [Male flower ^{137}Cs concentration in 2011] $^{1.2022}$; (R^2 = 0.7812, P < 0.001, n = 13), we estimated the quantity of ^{137}Cs per current year leaf (Fig. A4-1, decay correction: 1 Dec 2011) using the following equation:

Quantity of ^{137}Cs per leaf = Leaf ^{137}Cs concentration × Leaf weight

When we examined leaves that formed in the same year, we found that the quantity of ^{137}Cs per leaf did not clearly increase or decrease as the leaves aged (within the data range) (Fig. A4-1). Kiyono and Akama (2016) predicted that the leaf ^{137}Cs concentration would decrease in the current-year leaves year-by-year, and also decrease with leaf aging based on the leaf dynamics model. The pattern of annual change in the amount of ^{137}Cs per current-year leaf in this study (Fig. A4-1) matched the model predictions of Kiyono and Akama (2016). As the leaves aged, the numbers thereof decreased (Fig. A4-2), and the dry weights of leaves in the same length category tended to increase (Fig. A4-3), likely due to leaf thickening as they grew into branches following the development of the xylem. Since the increase in dry weight in leaves (Fig. A4-3) offset the decrease in ^{137}Cs concentration per unit dry weight (Figs. 5 and 6), the quantity of ^{137}Cs per leaf did not clearly increase/decrease as the leaves aged (Fig. A4-1).

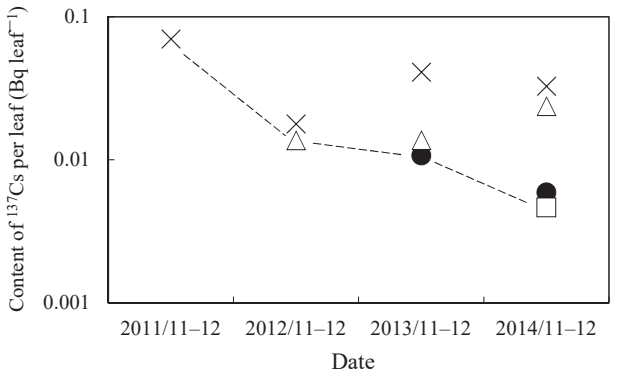

Fig. A4-1. The content of ^{137}Cs per *Cryptomeria japonica* var. *japonica* leaf (a) 8–9 months after the nuclear accident and (b) 3 years (plus 8–9 months) after the accident.
Data were collected in 21 *C. japonica* var. *japonica* forests in Fukushima Prefecture (Akama et al. 2013; Forestry Agency 2015, revised). ×, leaves that formed in 2011; Δ, leaves that formed in 2012; ●, leaves that formed in 2013; □, leaves that formed in 2014. Decay correction date: December 1, 2011. Content of ^{137}Cs per leaf = leaf ^{137}Cs concentration × leaf weight. ---, curve connecting the current-year leaves. The leaf ^{137}Cs concentration in 2011 (Bq dry kg^{-1}) was estimated from the male flower ^{137}Cs concentration in 2011 (Bq dry kg^{-1}) using an unpublished regression by the Forestry and Forest Products Research Institute (FFPRI) based on the data from five prefectures between Nagano and Fukushima: Leaf ^{137}Cs concentration in 2011 = 0.3915 [Male flower ^{137}Cs concentration in 2011] $^{1.2022}$; (R^2 = 0.7812, P < 0.001, n = 13).

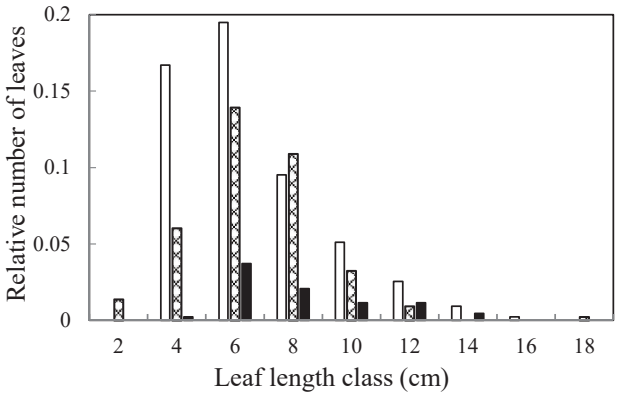

Fig. A4-2. Number of leaves of *Cryptomeria japonica* var. *japonica* in each leaf length class.
Leaf-bearing branches of several trees were collected and combined in Iwaki in November 2016. We added 2 cm to the length of each current-year leaf to take into account leaf elongation occurring between November and the following spring. □, current-year leaf; ☒, 1-year-old leaf; ■, 2-year-old leaf.

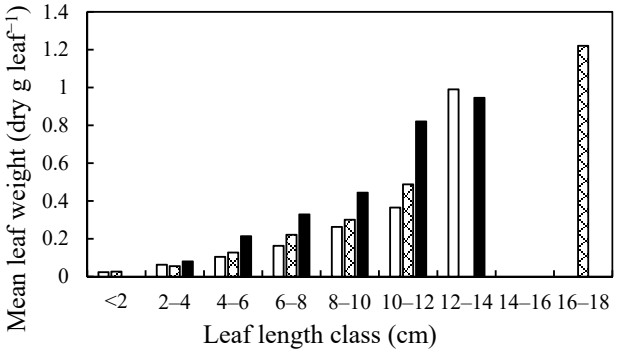

Fig. A4-3. Mean weight of *Cryptomeria japonica* var. *japonica* leaves in each leaf length class.
Leaf-bearing branches of several trees were collected in November 2016 in Iwaki and combined. □, current-year leaf; ☒, 1-year-old leaf; ■, 2-year-old leaf.

2011 年福島第一原子力発電所事故後 5 年間の、福島県のスギ（*Cryptomeria japonica* var. *japonica*）の花粉セシウム 137 濃度

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要旨

花粉飛散前のスギ（*Cryptomeria japonica* var. *japonica*）雄花の測定値をもとに、スギ花粉により再飛散される放射性セシウムの量を的確に推定するため、2011 年の東京電力福島第一原子力発電所事故の影響下にあるスギについて、花粉の完成直後（11 月）と飛散直前（翌年 2 月）の雄花と花粉の質量、セシウム 137（^{137}Cs）濃度を調べた。郡山市のスギ林では 11 月の雄花質量の約 1/3 が花粉で、雄花と花粉の ^{137}Cs 濃度に有意差はなかった。得られた雄花と花粉の関係を、福島県内の 21 か所で 2011–2015 年の毎年 11–12 月に得たスギ雄花の測定値にあてはめ、花粉 ^{137}Cs 濃度を推定した。スギ花粉 ^{137}Cs 濃度は年々低下し、2012 年春と比べ 2016 年春の濃度は約 8% であった。文部科学省の ^{137}Cs 沈着量の分布マップを利用して求めた、環境からスギ花粉への ^{137}Cs 面移行係数は 2012 年 2 月が 0.0203 m^2 dry kg^{-1}、2016 年 2 月は 0.00168 m^2 dry kg^{-1} であった。

キーワード：沈着、分布マップ、雄花、生物季節、放射性セシウム、面移行係数、花粉量年変化

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