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Topographic heterogeneity effect on the accumulation of Fukushima-derived radiocesium on forest floor driven by biologically mediated processes

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The accident at the Fukushima Daiichi nuclear power plant caused serious radiocesium (^{137}Cs) contamination of forest ecosystems located in mountainous and hilly regions with steep terrain. To understand topographic effects on the redistribution and accumulation of ^{137}Cs on forest floor, we investigated the distribution of Fukushima-derived ^{137}Cs in forest-floor litter layers on a steep hillslope in a Japanese deciduous forest in August 2013 (29 months after the accident). Both leaf-litter materials and litter-associated ^{137}Cs were accumulated in large amounts at the bottom of the hillslope. At the bottom, a significant fraction (65%) of the ^{137}Cs inventory was observed to be associated with newly shed and less degraded leaf-litter materials, with estimated mean ages of 0.5–1.5 years, added via litterfall after the accident. Newly emerged leaves were contaminated with Fukushima-derived ^{137}Cs in May 2011 (two months after the accident) and ^{137}Cs concentration in them decreased with time. However, the concentrations were still two orders of magnitude higher than the pre-accident level in 2013 and 2014. These observations are the first to show that ^{137}Cs redistribution on a forested hillslope is strongly controlled by biologically mediated processes and continues to supply ^{137}Cs to the bottom via litterfall at a reduced rate.

The accident at the Fukushima Daiichi nuclear power plant (NPP), triggered by a catastrophic earthquake (M9.0) and resulting tsunami on March 11, 2011, caused serious radioactive contamination over a wide area of eastern Japan[1]. Of the radionuclides found in the atmospheric fallout from the accident, ^{137}Cs with a physical half-life of 30.1 years is the largest source of concern because of its potential impact on humans and ecosystems over the coming decades.

Among the terrestrial ecosystems, forest ecosystems have received the most attention because a majority (~70%) of the land area contaminated by the accident is covered by forests[2]. Forest ecosystems consist of tree biomass (aboveground: boles, branches, and leaves; belowground: roots), forest-floor litter (fallen dead leaves and branches, and their decomposed materials), and underlying mineral soil[2]. Fukushima-derived ^{137}Cs was first deposited on forest-floor litter directly[3,4], or intercepted by forest canopies and subsequently deposited on the forest floor through processes such as throughfall (precipitation wash-off) and stemflow[5,6]. As a result, ^{137}Cs deposited on the forest floor was observed mostly in litter layers at forest sites in 2011 (ref. 3, 7), although some of the ^{137}Cs was observed to be retained by abiotic components (minerals and organic materials) of the underlying soil at shallow depths[8,9]. Studies conducted in European forests after the accident at the Chernobyl NPP showed that a large part of Chernobyl-derived ^{137}Cs persisted in forest-floor litter layers over a decade, and was a prolonged source for ^{137}Cs recycling in plants[10–12]. Therefore, knowledge about the behavior of Fukushima-derived ^{137}Cs in Japanese forest ecosystems—particularly in forest surface environments—is of great importance in the assessment of associated radiological risks from both external and internal (via consumption of forest products) radiation exposure.

Forest-floor litter is a dynamic component of forest ecosystems in Japan, with a mean residence time of a few years[13,14]. The temporal pattern of litter accumulation is a function of litter input and decomposition[15]. In temperate deciduous forests, a bulk of deciduous leaf fall (litterfall) occurs in autumn (October and November), which is the largest source of litter input on a forest floor. Litter decomposition on a forest floor involves a complex set of processes including physical, chemical, and biological breakdown of leaf-litter materials[16]. These decomposition processes operate simultaneously at varying rates depending on the litter quality and

environmental conditions by which the mass (and size) and chemical composition (e.g., carbon and nitrogen content and organic carbon structure) of leaf litter change during decomposition[14,17]. Because of variations in input and decomposition behavior, the forest-floor litter comprises a variety of organic materials of different degrees of degradation, from undecomposed and partially decomposed leaves to finely fragmented and macroscopically unrecognizable materials.

Local topography, notably hillslopes, can further influence litter accumulation on a forest floor through its effect on microclimate and differential lateral transport of litter materials[15,16]. Litter decomposition is generally faster at hillslope bottoms than at upper-slope positions because of the moderated environmental conditions at the bottom; however, the downslope movement of litter materials more than offsets the enhanced decomposition and increases the accumulation of litter materials at the bottom[15,18]. The topographically controlled accumulation of litter materials at hillslope bottoms should be a rapid ecological process; it is thus hypothesized that this process causes a rapid topographic heterogeneity in the distribution of Fukushima-derived ^{137}Cs on the forest floor. Once ^{137}Cs reaches the soil, it can be rapidly and strongly adsorbed by fine soil particles[19] and subsequently redistributed within the landscape primarily through physical processes[20,21]. Therefore, it is well documented that tracing the soil-associated ^{137}Cs provides a very effective tool for estimating long-term (20–50 years) rates of ^{137}Cs (soil) redistribution after deposition[21,22]. However, this technique is difficult to apply to litter layers as ^{137}Cs cannot be strongly fixed by forest-floor litter materials, probably because cesium forms weak bonds with natural organic matter[11]. There is still insufficient understanding of the short-term dynamic processes that influence the distribution, migration, and accumulation of ^{137}Cs in litter layers on a forested hillslope.

Here we investigated the distribution of ^{137}Cs in litter layers on a steep hillslope in a Japanese deciduous forest, the Ogawa Forest Reserve (Fig. 1), affected by the Fukushima NPP accident. We collected litter samples at three slope positions (bottom, and 8 and 12 m above the bottom) on the hillslope in August 2013 (before the bulk of litterfall occurred in 2013), fractionated the samples into four litter fractions (F1 to F4) that were characterized by different sizes and physical status, and then determined the concentrations and inventories of Fukushima-derived ^{137}Cs in the fractions. Furthermore, mean ages since litterfall for the fractions were estimated based on their chemical (carbon and nitrogen) composition. We also observed the yearly change in ^{137}Cs concentrations of fresh (newly emerged) leaves at the site. Based on these observations, we show evidence for a significant, short-term topographic heterogeneity in the accumulation of Fukushima-derived ^{137}Cs on the forest floor, which is driven by biologically mediated processes.

Results

Litter accumulation. The inventory of leaf-litter materials (excluding coarse woody debris such as fallen branches and twigs) on the forest floor was significantly greater at the bottom of the hillslope (2.6 ± 0.4 kg m^{-2}; mean ± standard deviation of triplicate samples) than at 12 m (0.26 ± 0.06 kg m^{-2}) and 8 m (0.56 ± 0.16 kg m^{-2}) above the bottom (see Table S1 in Supplementary information). The inventory of coarse woody debris was 0.14 ± 0.07 kg m^{-2} (range: 0.07–0.31 kg m^{-2}, corresponding to 4.8%–31.2% of the total amount of litter materials) and showed no significant difference between the slope positions.

The leaf-litter materials were fractionated into four fractions (Fig. 2): leaves showing no visible signs of degradation (F1); chipped or degraded leaves > 3 cm × 3 cm (F2) and < 3 cm × 3 cm (F3) in size; and fine leaf fragments < 1 cm × 1 cm in size, including macroscopically unrecognizable materials (F4). The fractionation method recovered 95.7% ± 2.6% (by weight) of leaf-litter materials (Table S1). The results revealed that the bottom of the hillslope accumulated more leaf-litter materials in all fractions than the upper parts of the hillslope (Fig. 3a). However, the distribution pattern of leaf-litter materials among the fractions was quite different between the slope positions. The F4 fraction represented the largest fraction (nearly half of the total mass) at the upper parts of the slope, whereas leaf-litter materials were approximately equally distributed among the four fractions at the bottom of the slope.

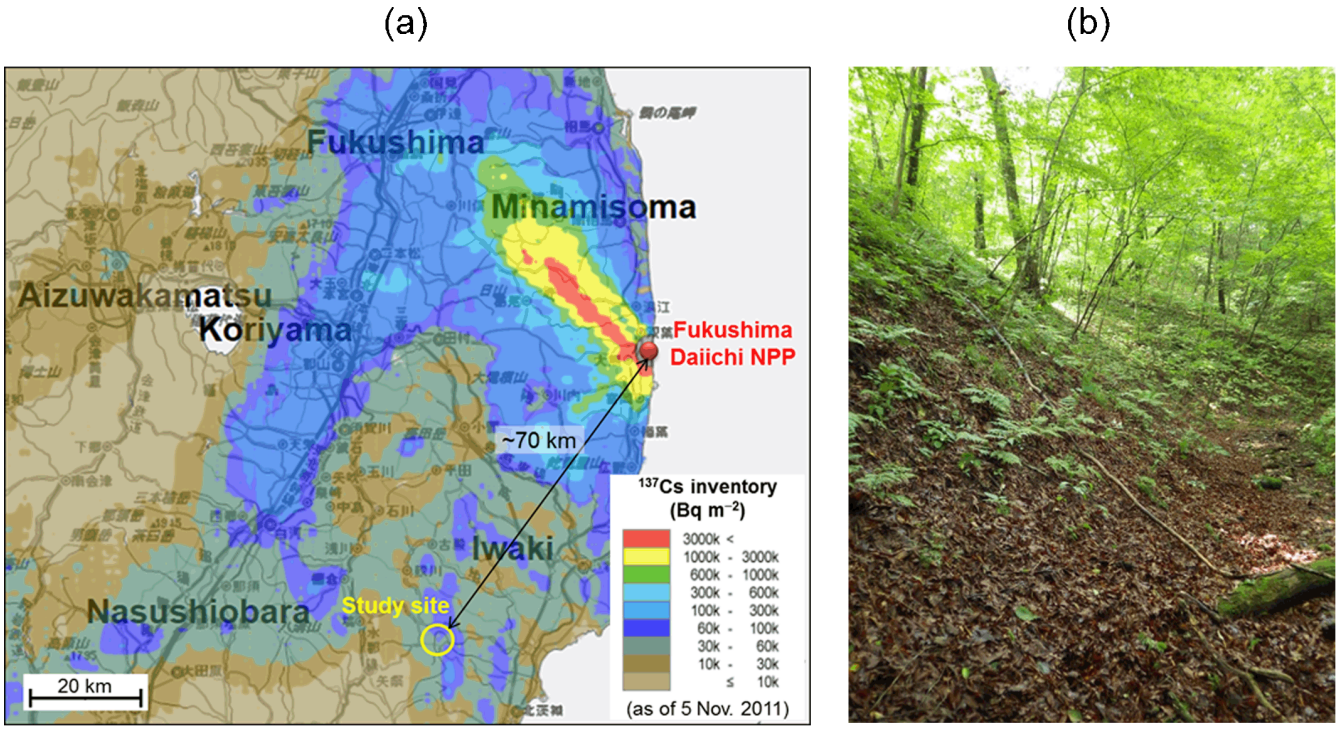

Figure 1 | Location of the Ogawa Forest Reserve (a) and a photograph of the forested hillslope (b) investigated in this study. The ^{137}Cs inventory map (a) was generated using the website "Extension Site of Distribution Map of Radiation Dose, etc." prepared by MEXT, Japan[40]. Photograph by E. Takeuchi.

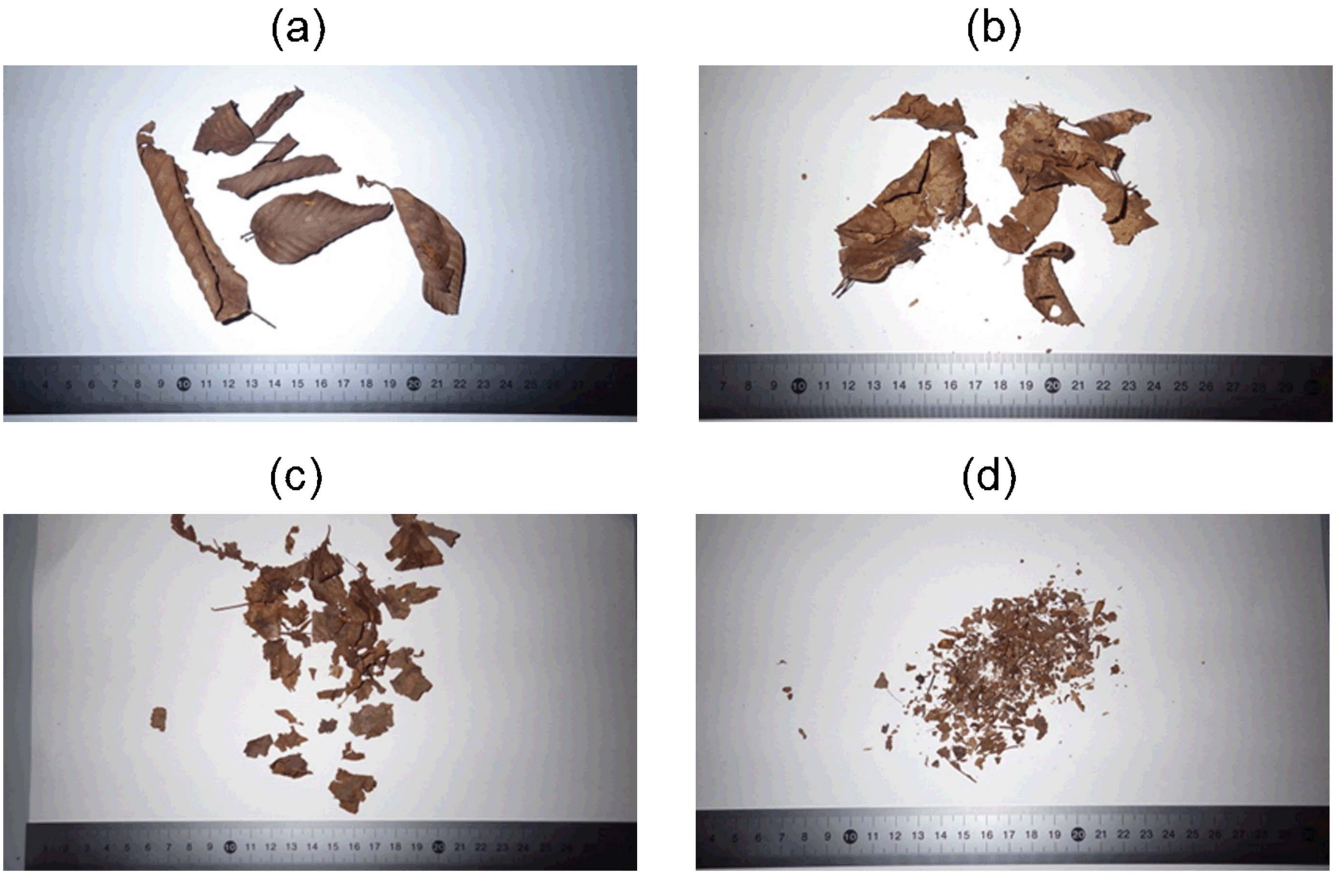

Figure 2 | Examples of litter materials in four litter fractions. (a) Leaves showing no visible signs of degradation (F1); (b) chipped or degraded leaves >3 cm × 3 cm in size (F2) and (c) <3 cm × 3 cm in size (F3); and (d) fine leaf fragments <1 cm × 1 cm in size, including macroscopically unrecognizable materials (F4).

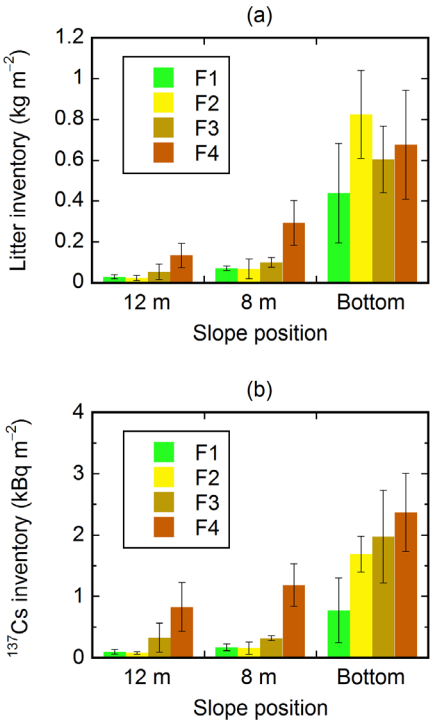

Figure 3 | Inventories of leaf-litter materials (a) and ^{137}Cs (b) in litter fractions (F1 to F4) at three slope positions.

C and N content of litter fractions. Overall, the C content of the litter fractions decreased with decreasing size of leaf-litter materials in the fraction (F1 to F4), but the N content remained nearly constant (Table 1). As a result, the C/N ratio of the litter fractions decreased with decreasing size of leaf-litter materials. Considering the general trend that the C/N ratio progressively decreases with litter mass loss during an early stage of decomposition[17,23], the C/N ratios obtained in this study indicate that the degree of degradation of leaf-litter materials increases in the order of F1 < F2 < F3 < F4. There were no consistent changes in the C and N content and the C/N ratio along the three slope positions.

Characterization of litter fractions based on their C/N ratios. A three-year litterbag experiment[14] previously conducted in the Ogawa Forest Reserve showed that the C/N ratio of decomposing leaf litter decreased exponentially with time for two dominant species (beech and oak). A relationship between the C/N ratio and the incubation period (or the age of leaf-litter materials) was derived using the litterbag experiment data[14] (see Fig. S1 in Supplementary information), and was employed to estimate the mean ages of litter materials in the four fractions. The mean ages increased with decreasing size of litter materials (F1 to F4), ranging from 0.5 to >3.0 years (Table 1). The mean ages of litter materials in the F1 fraction were generally less than one year, demonstrating that the leaves showing no visible evidence of degradation in this fraction were for the most part newly shed leaves originating from most recent litterfall events (i.e., October–November 2012). The mean ages for the F2 and F3 fractions suggest that the fractions mainly contained litter materials added to the forest floor in October–November 2011

Table 1 | Concentrations of ^{137}Cs, carbon (C), and nitrogen (N), and estimated mean ages of litter fractions at three slope positions

Slope position	Litter fraction	^{137}Cs concentration (Bq kg^{-1} dw)	C content (%)	N content (%)	C/N ratio	Mean age of litter materials[a] (yr)
12 m	F1	3203 ± 667[b]	44.1 ± 0.3[b]	1.51 ± 0.19[b]	29.6 ± 4.1[b]	0.5–1.2
	F2	3416 ± 765	41.9 ± 3.1	1.58 ± 0.10	26.5 ± 0.4	1.0–1.1
	F3	5973 ± 1994	37.3 ± 4.9	1.58 ± 0.17	23.5 ± 0.8	1.3–1.6
	F4	6085 ± 2217	27.2 ± 8.6	1.23 ± 0.34	22.0 ± 1.4	1.5–2.4
8 m	F1	2454 ± 920	35.9 ± 2.4	1.45 ± 0.10	24.8 ± 1.3	1.1–1.5
	F2	2543 ± 710	34.3 ± 2.3	1.50 ± 0.08	23.0 ± 1.7	1.3–2.1
	F3	3309 ± 1069	30.3 ± 0.7	1.46 ± 0.03	20.8 ± 0.3	2.2–2.5
	F4	4163 ± 969	17.7 ± 1.6	1.00 ± 0.05	17.7 ± 0.9	>3.0
Bottom	F1	1606 ± 442	46.0 ± 0.3	1.40 ± 0.04	32.8 ± 0.9	0.5–0.6
	F2	2178 ± 856	44.9 ± 1.6	1.57 ± 0.10	28.7 ± 2.6	0.6–1.1
	F3	3232 ± 550	40.0 ± 3.0	1.59 ± 0.01	25.2 ± 1.9	1.0–1.5
	F4	3819 ± 1347	35.4 ± 7.8	1.49 ± 0.26	23.6 ± 1.9	1.2–1.9

[a]Estimated based on the C/N ratios of litter fractions. See "Methods" for details.
[b]Mean and standard deviation for three replicate samples (n = 3).

(after the Fukushima NPP accident) at the bottom and at 12 m above the bottom of the hillslope. The finely fragmented F4 fraction showed mean ages of 1.2–2.4 years (except for the fraction obtained at 8 m above the bottom), indicating that the fraction was dominated by litter materials added via litterfall in 2010 (before the accident), as well as those added via litterfall in 2011 (after the accident).

The leaf-litter materials collected at 8 m above the bottom gave consistently lower C/N ratios (and thus, older mean ages) in all fractions than those collected at the other slope positions. Furthermore, a lower C content was observed in the fractions at this slope position, suggesting adhesion of soil mineral particles interacting with highly microbially transformed organic materials (humus) to leaf-litter materials[24].

Radiocesium concentrations of litter fractions. The litter fractions largely varied in ^{137}Cs concentration from 1,610 to 6,090 Bq kg^{-1} dry weight (dw) (Table 1). The concentrations were generally higher in fractions comprising smaller-size litter materials and at higher slope positions. Of particular interest was the observation of high ^{137}Cs contamination (1,610–3,200 Bq kg^{-1} dw), even in the F1 fraction at all slope positions.

The measured ^{137}Cs and ^{134}Cs concentrations showed a similar pattern of distribution for all the litter fractions (see Table S1). The ^{134}Cs/^{137}Cs activity ratios of the litter fractions were 0.48 ± 0.02 (mean ± standard deviation), independent of the litter fraction category. The ratios corresponded well to the ratio (0.47) theoretically predicted for Fukushima-derived radiocesium at the time of sample collection (August 2013), the initial ratio being unity in March 2011 (ref. 25) and decreasing according to different rates of radioactive decay (the physical half-lives of ^{137}Cs and ^{134}Cs are 30.1 and 2.1 years, respectively). Therefore, the observed ^{137}Cs in the present study was considered to originate from the Fukushima NPP accident.

Radiocesium inventory in litter layers. The total inventory of ^{137}Cs in litter layers was significantly greater at the bottom (6.8 ± 0.6 kBq m^{-2}) than at the upper-slope positions (1.3 ± 0.6 and 1.8 ± 0.4 kBq m^{-2}) (Fig. 3b). At the upper-slope positions, approximately two-thirds of the total ^{137}Cs inventory was apportioned to the F4 fraction; the other three fractions (F1, F2, and F3) retained only a small amount (~0.5–0.6 kBq m^{-2} in total) of ^{137}Cs. In contrast, a large proportion (~65%) of Fukushima-derived ^{137}C was observed in the F1, F2, and F3 fractions at the bottom of the hillslope, which amounted to ~4.4 kBq m^{-2}, far larger than the total inventory of ^{137}Cs at the upper-slope positions.

Radiocesium concentrations of fresh leaves. Concentrations of ^{137}Cs in fresh leaf samples were 286–3,310 Bq kg^{-1} dw (Table 2), high in May 2011 (two months after the Fukushima NPP accident) and low in October 2013 and May 2014. The ^{134}Cs/^{137}Cs activity ratios were close to the theoretically predicted ratios (0.95, 0.45, and 0.38 for samples collected in 2011, 2013, and 2014, respectively), indicating that the radiocesium isotopes observed here came from the Fukushima NPP accident. There was no

Table 2 | Radiocesium concentrations of an archived litter sample and fresh beech leaf samples

Sampling date	Sample type[a]	Height (m)	^{137}Cs concentration (Bq kg^{-1} dw)	^{134}Cs concentration (Bq kg^{-1} dw)	^{134}Cs/^{137}Cs	Detailed conditions
January 23, 2007	Litter	NA[b]	5.0 ± 1.2[c]	ND[d]	ND[d]	
May 17, 2011	Leaves	1.5	3310 ± 100	3160 ± 175[c]	0.95	
	Leaves	<0.5	1790 ± 27	1760 ± 49	0.98	
October 1, 2013	Leaves	1–3	557 ± 9	260 ± 8	0.47	
May 2, 2014	Leaves	3–5	792 ± 39	333 ± 31	0.42	At 12 m above the bottom
	Leaves	3–5	875 ± 47	309 ± 33	0.35	At 12 m above the bottom, washed with water
	Leaves	3	286 ± 24	113 ± 20	0.40	At 8 m above the bottom
	Leaves	3	316 ± 27	131 ± 22	0.42	At 8 m above the bottom, washed with water
	Leaves	2–3	362 ± 33	128 ± 27	0.35	At the bottom
	Leaves	2–3	433 ± 37	152 ± 28	0.35	At the bottom, washed with water

[a]Samples were collected on the hillslope in May 2014, around the hillslope (not on the hillslope) in October 2013, and ~180 m away from the hillslope in January 2007 and May 2011.
[b]Not available because this is a litter sample collected from a forest floor.
[c]Errors represent counting errors in the radiation measurement.
[d]Not determined: ^{134}Cs concentration was less than the lowest detectable concentration, and therefore ^{134}C/^{137}Cs ratio was not determined.

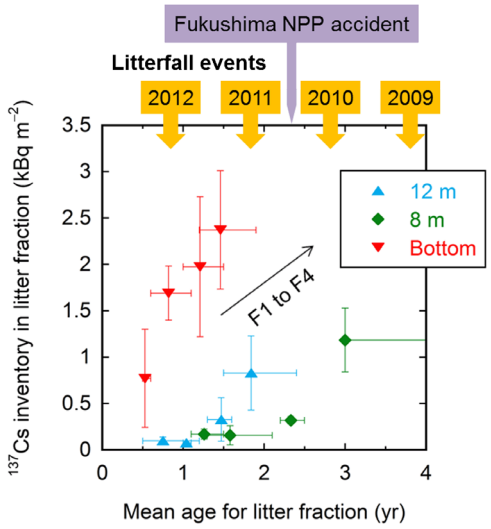

Figure 4 | Relationship between mean age and ^{137}Cs inventory for litter fractions (F1 to F4) at three slope positions.

difference in ^{137}Cs concentrations between the washed and non-washed leaf samples in 2014.

No fresh leaf samples were collected from the site before the accident. To determine the pre-accident level of ^{137}Cs in beech leaves at the site, an archived litter sample collected from the forest floor in 2007 was analyzed for radiocesium. The ^{137}Cs concentration of the archived litter sample was 5.0 Bq kg^{-1} dw, which was three orders of magnitude lower than that of fresh leaves collected in May 2011. As expected, ^{134}Cs was not detected in the sample.

Discussion

The results of the present study showed that hillslope topography has a great effect on the accumulation of litter materials and consequently of Fukushima-derived ^{137}Cs on the forest floor. A similar topographic pattern of surface litter accumulation along a hillslope has been observed throughout a growing season (April to October) in an American beech and maple forest[15]. The total inventory of Fukushima-derived ^{137}Cs in the litter layer varied by a factor of five, from 1.3 kBq m^{-2} at the highest position to 6.8 kBq m^{-2} at the bottom position (Fig. 3b). More importantly, a significant fraction (65%) of the ^{137}Cs inventory was associated with the younger litter materials (F1 to F3 fractions: mean ages of 0.5–1.5 years, dominated by litter materials added after the Fukushima NPP accident) at the bottom of the hillslope (Fig. 4).

The picture that emerges is that biological recycling of ^{137}Cs (i.e., uptake of ^{137}Cs by trees and subsequent re-deposition on the forest floor via litterfall) plays an important role in causing topographic heterogeneity in the accumulation of Fukushima-derived ^{137}Cs on the forest floor. The fresh leaves collected in May 2011 were highly contaminated (1,790–3,310 Bq kg^{-1} dw) with Fukushima-derived ^{137}Cs (Table 2). The leaves were newly emerged ones; such a contamination therefore cannot be explained without invoking mechanisms such as uptake of ^{137}Cs by roots and translocation of ^{137}Cs from tree stems[6,26,27]. The contamination of leaf surfaces by adhering resuspended soil particles may be possible; however, the ^{137}Cs concentration of fresh leaf samples was not reduced by washing (Table 2), suggesting that this process is of minor importance compared with root uptake[28]. Given the finding in the Ogawa Forest Reserve that leaves disperse along hillslopes within 20 m of source trees by lateral transport driven by wind action while falling[29], it is possible that newly emerged leaves contaminated with "biologically recycled" ^{137}Cs were carried to the bottom of the hillslope via litterfall. This is consistent with the observation that ^{137}Cs concentrations (2,180–3,230 Bq kg^{-1} dw) of the litter materials in the F2 and F3 fractions (considered mainly from the 2011 litterfall events) at the bottom of the hillslope were similar to those of the fresh leaves collected in May 2011.

With an annual litterfall input of 0.43 kg m^{-2} y^{-1} at this site[30], we estimate the annual input of biologically recycled ^{137}Cs on the forest floor to be 0.77–1.42 kBq m^{-2} y^{-1} in 2011. This input corresponds to ~21%–39% of the ^{137}Cs inventory in the F2 and F3 fractions at the bottom of the hillslope in August 2013, which is sufficiently large to possibly explain the observed accumulation of Fukushima-derived ^{137}Cs on the forest floor at the bottom of the hillslope.

Furthermore, our observations suggest that the biologically recycled ^{137}Cs has been supplied, but at a reduced rate, to the bottom of the hillslope via litterfall until the present. At the bottom of the hillslope, a substantial amount (0.77 kBq m^{-2}) of Fukushima-derived ^{137}Cs was found in the youngest F1 fraction, the nearly intact leaf-litter materials originating mainly from the 2012 litterfall events (Fig. 4). The fresh leaves collected in 2013 and 2014 still had ^{137}Cs concentrations ranging from 286 to 875 Bq kg^{-1} dw; the concentrations were much lower than that of fresh leaves collected in May 2011 (two months after the accident), but were far higher than the pre-accident level (5.0 Bq kg^{-1} dw) (see Table 2). The contaminated leaves could still mediate ~0.13–0.38 kBq m^{-2} of biologically recycled ^{137}Cs via annual litterfall.

On hillslopes, leaf-litter materials that have already been deposited on the forest floor may be redistributed by wind and gravity during snow-cover-free periods[15,31]. The trees at our site had no leaves in March 2011 when the Fukushima Daiichi NPP accident occurred; therefore, a majority of Fukushima-derived ^{137}Cs was very likely to have been directly deposited onto the forest-floor litter materials[4]. The larger accumulation of Fukushima-derived ^{137}Cs in the oldest F4 fraction at the bottom may indicate the redistribution (downslope movement) of litter materials contaminated with "directly deposited" ^{137}Cs to the bottom. However, the younger mean ages for the F4 fraction at the bottom compared with the upper positions suggests a supply of younger litter materials to the fraction at the bottom; this indicates that the ^{137}Cs accumulation observed in the F4 fraction at the bottom may be partly due to the supply of biologically recycled ^{137}Cs. At the upper-slope positions, Fukushima-derived ^{137}Cs was observed mainly in the oldest F4 fraction (Fig. 4). At the bottom of the hillslope, a large amount (4.4 kBq m^{-2}) of litter-associated ^{137}Cs was accumulated in the three younger (F1, F2, and F3) fractions. These results suggest that the short-term (within 2–3 years) topographic heterogeneity in the distribution of Fukushima-derived ^{137}Cs on the forest floor has been established primarily by the redistribution of biologically recycled ^{137}Cs, rather than by the redistribution of directly deposited ^{137}Cs.

These findings have major implications for the assessment of future impacts of radioactive contamination of forest ecosystems from the Fukushima NPP accident. The biologically mediated redistribution of ^{137}Cs on forested hillslopes significantly alters the distribution of litter-associated ^{137}Cs on the forest floor, and thus alters the radiation situation of not only external but also internal exposure to the population. The local (secondary) accumulation of litter-associated ^{137}Cs at hillslope bottoms is likely to be a main source for ^{137}Cs recycling in plants in the long term[11], which may result in unexpected ^{137}Cs contamination levels in some forest products. The redistribution may further influence the discharge of Fukushima-derived ^{137}Cs from forest ecosystems. Studies conducted after the Chernobyl NPP accident have shown that forest ecosystems act as effective long-term reservoirs of the deposited ^{137}Cs (ref. 32). However, the litter-associated ^{137}Cs allocated to hillslope bottoms probably has more opportunities to be carried away by stream flows (particularly those caused by heavy precipitation events), and thus to be transferred from forest ecosystems to downstream areas via aquatic pathways[33,34]. This biologically mediated redistribution

seems particularly important in Japan where forests are concentrated in mountainous and hilly regions with steep terrain. Clearly, this is worth further investigation (including field observations and modeling) to improve our understanding of the dynamics of ^{137}Cs in Japanese forest ecosystems. Finally, the results of this study suggest that even more than three years after the Fukushima NPP accident, the removal of forest-floor litter materials preferentially accumulated at hillslope bottoms is still an effective countermeasure option to reduce forest contamination[35].

Methods

Study site. The study was conducted on a steep, flat-bottomed hillslope (slope length: 28.5 m, slope angle: 35°–40°) in the Ogawa Forest Reserve (36°56′N, 140°35′E) in Ibaraki prefecture, Japan (Fig. 1). The Ogawa Forest Reserve is a temperate broad-leaved deciduous forest dominated by Japanese beech (*Fagus crenata*) and Japanese oak (*Quercus crispula*), located on an undulating plateau at the southern edge of the Abukuma mountain region. The area of the forest catchment is 58.4 ha and the elevation ranges from 588 to 724 m (ref. 36). The bulk of litterfall in the forest occurs during October and November, and the annual litterfall input on the forest floor is 0.43 kg m^{-2} y^{-1} (ref. 37). The trees had no leaves in March 2011 when the Fukushima Daiichi NPP accident occurred. The mean annual temperature and precipitation are 10.7°C and 1,910 mm, respectively[38]. The soils of this area are heterogeneous, exhibiting a mosaic-style pattern of distribution of Cambisols and Andosols[39]. The parent materials are metamorphic rock and Late Quaternary volcanic ash[39]. The site was located approximately 70 km southwest of the Fukushima Daiichi NPP, and affected by radioactive fallout from the Fukushima NPP accident at a level of 10–60 kBq m^{-2} of ^{137}Cs deposition according to an airborne monitoring survey[40].

Litter sample collection and fractionation. In August 2013 (before newly emerged leaves began to fall), litter samples were collected from litter layers on the surface of the soil at three slope positions: bottom, and 8 and 12 m above the bottom of the hillslope. Litter samples were collected from three replicate plots (each having a 30 cm × 30 cm square) at each slope position. The litter samples were immediately transported to our laboratory with special care to avoid artificial fragmentation, and then gently spread on wide trays to dry to a constant weight at room temperature.

To investigate the distribution of Fukushima-derived ^{137}Cs among litter materials of different degrees of degradation, the litter samples were physically separated by hand into the following four fractions (Fig. 2). The first fraction (F1) consisted of leaves showing no visible signs of degradation. The second (F2) and third (F3) fractions both consisted of leaves that are more or less chipped or degraded, and also included "skeleton leaves" in which the leaf parenchyma has been largely decomposed, but the midrib and veins (leaf tissues more resistant to microbial decomposition) persist. These two fractions were differentiated by leaf size: >3 cm × 3 cm in size for the F2 fraction and <3 cm × 3 cm for the F3 fraction. The last fraction (F4) consisted mainly of fine leaf fragments (<1 cm × 1 cm in size), including petioles detached from leaves and macroscopically unrecognizable materials. Here we assumed that the degree of degradation of litter materials is closely related to their sizes and therefore increases in the order of F1 < F2 < F3 < F4 in the fractionation method. Note that coarse woody debris (fallen branches and twigs) in the collected samples were removed before fractionation.

The amount of litter materials per unit area (or litter inventory; in kg m^{-2}) in a given fraction was estimated as the mass of litter fraction (kg dw) divided by the area (m^2) where the litter sample was collected (i.e., 900 cm^2).

Fresh leaf sample collection. Fresh leaves were collected from a single beech tree growing around the hillslope in October 2013. In May 2014, newly emerged leaves were collected from a single beech tree at each slope position on the hillslope. The leaf samples collected in May 2014 were divided into two in our laboratory: one was dried without pretreatment; and the other was washed with water to remove adhering soil particles before drying. We did not collect leaf samples at the hillslope site in 2011, but had a sample (newly emerged beech leaves) collected ~180 m away from the hillslope within the Ogawa Forest Reserve in May 2011 (two months after the Fukushima NPP accident). This sample was used to estimate the radiocesium contamination level in fresh leaves in 2011.

There was also no sample collection at the site before the Fukushima NPP accident. Fortunately, we had an archived litter sample that was collected in January 2007 at the point where the 2011 leaf sample was collected. This archived litter sample was used to estimate the pre-accident level of ^{137}Cs concentration in beech leaves at the site. All the leaf samples were dried to a constant weight at room temperature before the following analyses.

Radiocesium analysis. The activity concentrations of ^{137}Cs and ^{134}Cs in the litter fractions (F1 to F4) and fresh leaf samples were determined using gamma ray spectrometry, and their values were expressed in activity per unit dry weight (Bq kg^{-1} dw). Samples (dried) were finely chopped using a mixer, sealed in plastic tubes (5 cm diameter, 7 cm height), and analyzed for ^{137}Cs and ^{134}Cs using a high-purity coaxial germanium detector (model GEM25P4-70, ORTEC, USA) at the Nuclear Science and Engineering Center of the Japan Atomic Energy Agency. The detector was calibrated with standard gamma sources (each with a relative uncertainty of ~5% for ^{137}Cs) with different sample heights. The measurement times were normally 2,000–4,000 s for litter samples and 5,000–50,000 s for fresh leaf samples, which allowed us to obtain both ^{137}Cs and ^{134}Cs concentration values with relative errors <10% (with some exceptions for fresh leaf samples having low ^{134}Cs concentration). The activity concentrations were corrected for radioactive decay to the sampling date.

Radiocesium inventory (Bq m^{-2}) in a given litter fraction was estimated by multiplying the radiocesium activity concentration (Bq kg^{-1} dw) of the litter fraction by the inventory of the litter fraction (kg m^{-2}).

C and N analysis and mean age estimation. The litter fractions were further analyzed for their total C and N content using an elemental analyzer (vario PYRO cube, Elementar). A three-year litterbag experiment[14] previously conducted in the Ogawa Forest Reserve showed that the C/N ratio of decomposing leaf litter decreased exponentially with time (and thus with decreasing litter mass) for two dominant species (beech and oak). The data were used to derive a relationship between the C/N ratio (R) and the field incubation period (or the mean age of litter materials since the deposition: Y in years) (see Fig. S1 in Supplementary information):

$$Y = -0.81 \times \ln((R - 19.31)/25.85), \quad (1)$$

and then the mean ages for the litter fractions obtained in the present study were estimated from their C/N ratios through this relationship.

Data analysis. The inventories of litter materials and ^{137}Cs at different slope positions were statistically analyzed using analysis of variance. Means that exhibited significant differences were subjected to Tukey's honestly significant difference test to find which means were significantly different from one another at a 5% significance level.

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Author contributions

J.K. designed the study; J.K., M.A.A. and S.N. participated in the field work; J.K. performed the laboratory experiments; J.K. and E.T. conducted the radioactivity measurements; J.K. wrote the manuscript. All the authors contributed to discussions about this study and reviewed the manuscript.

Additional information

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