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The Distribution and Migration of ^{137}Cs in Oak (*Quercus serrata*) and Cedar (*Cryptomeria japonica*) Forest Organic Fractions

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Abstract: To analyse the ^{137}Cs distribution and migration under various fractions of organic matter layers, this study investigated easily recognizable, originally shaped organic L-fractions, and not easily recognizable, early fermented and fragmented organic F-fractions, of both oak (*Quercus serrata*) and cedar (*Cryptomeria japonica*) sampled from Osawa watershed sites at Nihonmatsu City, Fukushima Prefecture, Japan. The organic materials were put on top of soil columns from Field Museum (FM) Tamakyuryo in Hachioji City, Tokyo. The ^{137}Cs vertical distribution in forest soil profiles was analyzed using the relaxation mass depth, ho (kg m^{-2}). Soil columns with both L and F- organic layer fractions of both oak and cedar, labelled as Oak-L, Oak-F, Cedar-L and Cedar-F with four replications (n = 16), were set up by the laboratory column-based method and kept under five months' incubation period. Soil columns after incubation were sampled at depths of 0–1 cm, 1–2 cm, 2–5 cm and 5–10 cm. Results of ^{137}Cs inventory in the organic fractions showed that 86% (oak and cedar) of the total organic layer fractions ^{137}Cs inventory accumulated within the F-layer, indicating that the transformation of litter is a huge source for potentially mobile ^{137}Cs, especially the oak F-layer (67% ^{137}Cs inventory) and further continuous transfer into the forest soil mineral layers. A higher ho in L treated soils (Oak-L and Cedar-L) compared to the F treatments implied that the low ^{137}Cs amounts penetrated faster and deeper due to their water-soluble nature. Furthermore, Cedar-F showed a higher ho of 24.3 kg m^{-2} than Oak-F of ho, 14.0 kg m^{-2}, and a significant positive relationship between ^{137}Cs retention and total carbon (TC) ($p < 0.05$) suggested the influence of soil organic matter on ^{137}Cs penetration and retention. The C/N (carbon nitrogen ratio) results revealed that organic matter fractions of high C/N including ^{137}Cs, as observed in Cedar-F, in which decomposition does not advance, penetrates soil depths while the organic matter fraction of low C/N, observed in Oak-F, showed that decomposition advanced to release ^{137}Cs which was held by adsorption unto the RIP (radiocesium interception potential) of soil surface. In addition, infiltration by water as a transportation process was suggested to largely influence the downward migration and retention of ^{137}Cs at lower depths of Cedar-F.

Keywords: forest litter; radiocesium; laboratory column-based method; soil depth distribution; relaxation mass depth; ^{137}Cs retention

1. Introduction

The Fukushima Dai-ichi Nuclear Power Plant (FDNPP) explosion, due to a large earthquake coupled with a tsunami which hit Northeastern Japan on 11 March 2011, resulted in radionuclide contamination of a large amount of the environment within Fukushima and neighbouring prefectures [1]. Major radionuclides released during the accident are ^{131}I, ^{134}Cs and ^{137}Cs, of which ^{137}Cs is of global concern due to its biological availability and relatively longer half- life ($t_{1/2}$ = 30.2 years) [2]. Of the major contaminated environments, the forest, which constitutes about 70 % of Japan's archipelago, is a major recipient of the terrestrial ^{137}Cs fallout [3,4]. Prolonged contamination of the forest is of serious concern due to the detrimental effect on timber production and various additional problems such as the contamination of edible forest products like wild mushrooms, plants and animals [5,6]. The monitoring of ^{137}Cs distribution within the forest ecosystem has therefore become necessary to provide countermeasures or remedies.

Forests, unlike other land uses, have fewer soil disturbances and a more preserved mineral horizon [7]. On such undisturbed sites, the highest radiocesium concentration is in the uppermost soil layer; it decreases exponentially with the depth, in which important characteristic features of forest soils, such as the presence of litter and organic horizons, are considered to be important for ^{137}Cs availability [8–10]. Of an approximate total of 5.5 kBq m^{-2} Fukushima-derived radiocesium, litterfall is known to contribute 45% of deposited 137Cs [11], revealing that radioactive materials primarily remain in the aboveground tree biomass, litter, and the thin top layer of soil (0–0.05 m) in forests. Of the three components, radioactive Cs concentrations (total of ^{134}Cs and ^{137}Cs) in litter ranged from 24.1 to 319 kBq kg^{-1}, making up 22–66% of the total radioactive Cs in the forest ecosystems at this survey period conducted in August and September 2011 [12]. The removal of the aboveground tree biomass and litter removal at the earlier stage of deposition can decontaminate approximately 80% of the radioactive component [13]. Furthermore, sequential extraction of ^{137}Cs by Mensah et al. [14] in Fukushima forest soils revealed that 9–44% were in the mobile organic-matter bound fraction, while the results of the pot experiment study by Choi et al. [15] showed considerably high ^{137}Cs concentrations (252 Bq kg^{-1} dry matter) in leaves of oak seedlings (*Q. serrata*) transferred from contaminated forest soil. The forest-floor organic layers are, therefore, known to potentially retain as well as release largest amounts of the deposited ^{137}Cs for a long time, and play a crucial role in the long-lasting cycling of ^{137}Cs via tree root uptake and redeposition onto the forest floor as litter fall [16], with the potency of influencing ^{137}Cs distribution and migration in forest soils.

Forest-floor organic layers within the forest ecosystem are a dynamic component composed of numerous litter materials of different precursor species and degrees of degradation [17]. In deciduous broadleaved forests, deciduous leaf fall occurs annually and the decomposition of broadleaves in such forests has been shown to be rapid [18,19]. In contrast, the mean leaf longevity of coniferous trees, in evergreen coniferous forests, is longer than a year and the decomposition of needle-like leaves in the organic layers tends to be much slower compared to broadleaves (unified with others) [20–22]. There is therefore the possibility that the ^{137}Cs retention behavior in organic layers differs depending on the forest type. This is of relevance given that the major forest types in the heavily contaminated areas after the FDNPP accident are deciduous broadleaf forests, which are more distributed in the northern part, and evergreen needleleaf forests, which are distributed in the southern part of Fukushima and its neighboring prefectures [13]. Several studies have investigated ^{137}Cs accumulation and retention behavior in organic fractions or layers of these two types of forests under specific conditions in Japan [16,17,23]. However, the contribution of both deciduous broadleaf and coniferous needleleaf organic layer fractions to ^{137}Cs vertical distribution and migration in organic layers (and further into mineral layers) of forest soils has not been thoroughly studied.

Thus, the objective of this study was to analyze the role of relatively undecomposed L- and partially decomposed F- from both deciduous oak and coniferous cedar organic

fractions to ^{137}Cs vertical distribution and migration in forest soils via a laboratory column-based method.

2. Materials and Methods

2.1. Soil Sampling and Preparation

2.1.1. Study Sites

Study sites for organic horizon sampling were selected at Osawa watershed in Nihonmatsu City, Fukushima Prefecture (37°36′11.2″ N, 140°36′40.4″ E) due to the presence of various forest vegetation (Figure 1).
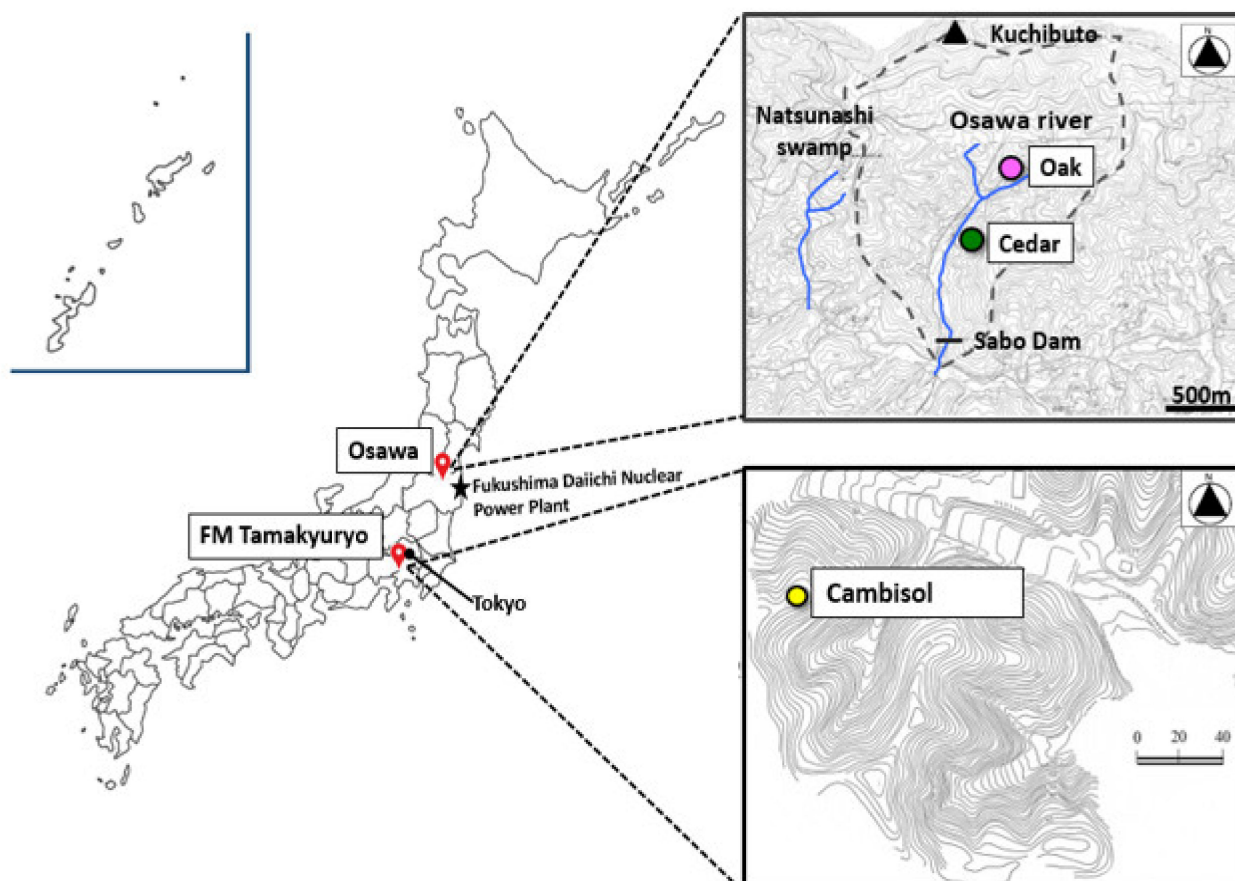

Figure 1. Map showing forest litters and forest soil sampling sites. Location of forest litter site is at Osawa and the forest soil site is at FM Tamakyuryo.

Nihonmatsu City is located approximately 70 km northwest from the FDNPP in Japan and covers an area of 344.65 km^2. The average annual precipitation is 1215 mm with a mean temperature approximated at 12.0 °C from 2008–2017 [23]. The sampling sites selected for this study were Osawa oak and Osawa cedar forest sites. The Osawa cedar site is dominated by *C. japonica* while the dominant tree species at the Osawa oak site is *Q. serrata*. The initial ^{137}Cs deposition density, measured in June 2020, were approximately 250 kBq m^{-2} and 300 kBq m^{-2} for the cedar site and oak site, respectively. In addition, the air dosage rates measured in the same year were1.24 ± 0.14 μSv/h and 1.72 ± 0.17 μSv/h for cedar and oak sites, respectively. The estimated tree density in 2013 were 675 trees per ha and 850 trees per ha for oak and cedar sites, respectively. In addition, measured tree height (m) and diameter at breast height, DBH (cm), in the same year were 14 ± 1.8 m and 19 ± 5.4 cm respectively for the oak site, while that of the cedar site was 17 ± 1.5 m and 28 ± 7.1 cm, respectively.

The study site for forest soil sampling was selected from Field Museum (FM) Tamakyuryo of Tokyo University of Agriculture and Technology in Hachioji City, Tokyo in Japan (35°38′18.5″ N 139°22′41.7″ E) (Figure 1). The basis for selecting the study site was due to no ^{137}Cs contamination or minimal contamination beyond detection. The average amount of annual precipitation in Hachioji city is 1642 mm with the mean temperature approximated at 14.8 °C from 2008–2017 [23]. The soil types at FM Tamakyuryo are classified as Andosol and Cambisol [24].

2.1.2. Sampling Methods

Two organic layer fractions, L- and F-, on the soil surface were collected from the study sites on 8–9 December 2014 by hand (Figure 2).
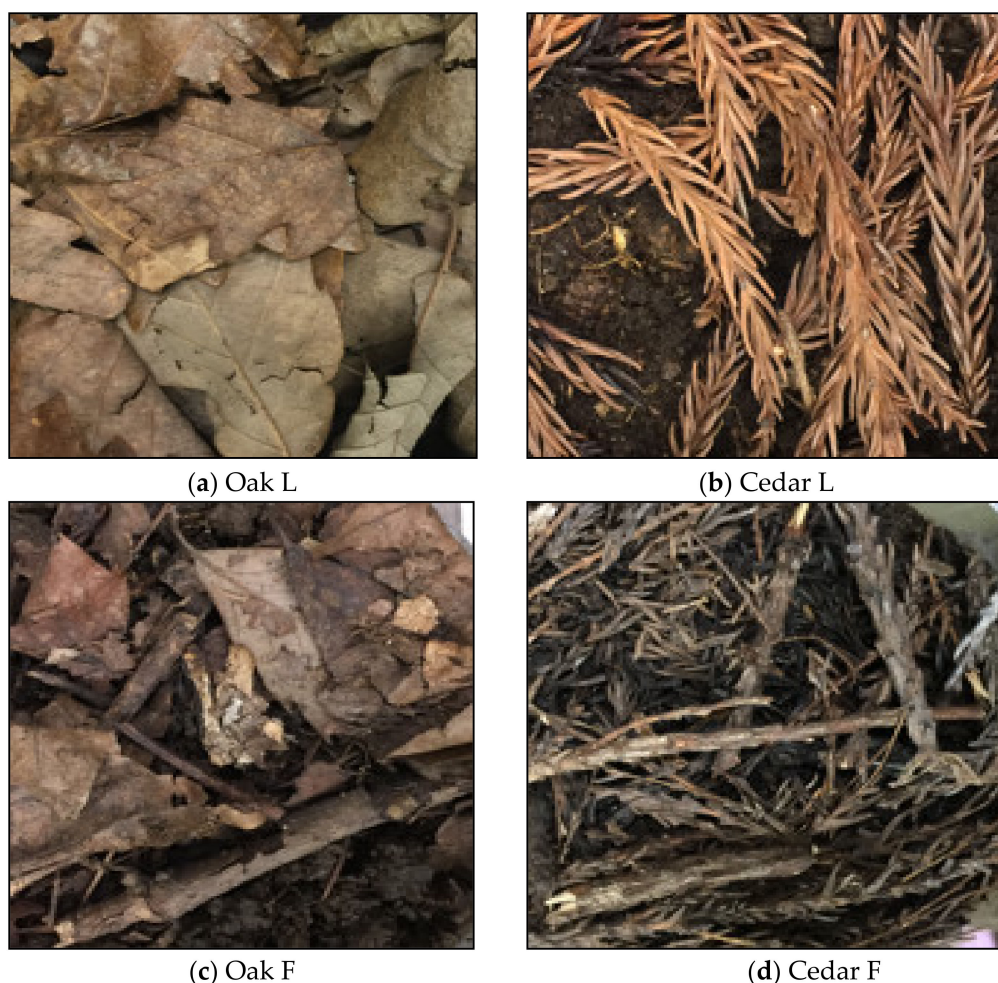

(**a**) Oak L   (**b**) Cedar L

(**c**) Oak F   (**d**) Cedar F

Figure 2. Separated (**a**) Oak L—litter organic layer, (**b**) Cedar L—litter organic layer, (**c**) Oak F—litter organic, (**d**) Cedar F—litter organic layer fractions. The L-fraction consists of easily recognizable, originally shaped organic components made up of periodically falling raw litter while the F- fraction is composed of an early fermented and fragmented litter component in which the original shapes of the litter were not easily identified.

The basis for sampling was similar to that described by Koarashi & Atarashi-Andoh [16]. The L-layer fraction consisted of easily recognizable, originally shaped organic components made up of periodically falling fresh litter. The F-layer fraction, located beneath the L-layer fraction, was composed of an early fermented and fragmented litter component in which the original shapes of the litter were not easily identified. All samples were transported carefully to maintain their natural fragmentation and treated by spreading the samples on a tray and drying at 60 °C in an oven for 72 h to a constant weight at the laboratory.

The organic layer samples were later cooled and stored in well-labelled plastic bags based on their fractionation.

Sixteen columns of Cambisol were sampled from FM Tamakyuryo on 28 July, 2015 to the depth of 10 cm (0–10 cm) of the A-horizon, using a PVC pipe column of area 87 cm^2 (diameter = 10.5 cm) and height 10 cm. Sampling was done such that the soil columns were structurally undistributed as a basis of natural prediction for ^{137}Cs distribution, while avoiding interference from fine roots and rock fragments. The soil columns were transferred into already-prepared transparent PVC pipes of the same column area and marked at 1 cm intervals. Attached to the base of each column was a non-biodegradable plastic material with uniform holes and the inside lined with fine white cloth which will allow water other than soil to pass through. The point of attachment between PVC pipes and the base cover was sealed with a strong adhesive to prevent water loss by seepage. Soil columns in pipes were covered with aluminum foils, and carefully transported and stored under low temperature to maintain soil structure and weight. Additional soil columns were sampled for physicochemical analyses.

2.2. Experimental Setup

The experimental setup was based on 2×2 factorial design with four replications each and a modification of the laboratory column-based method by [25] (Figure 3a).

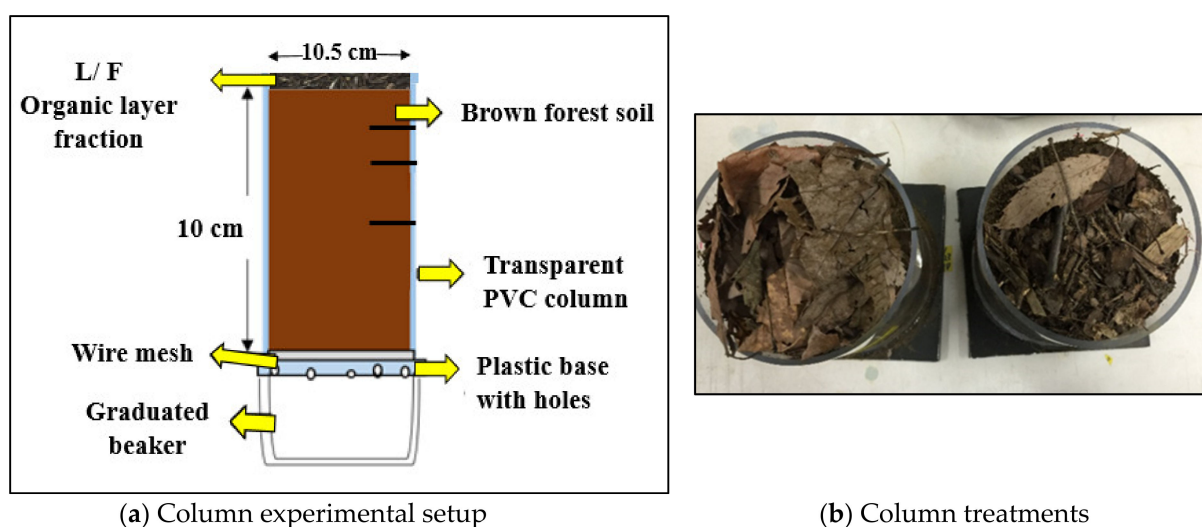(**a**) Column experimental setup　　　　　　　　　(**b**) Column treatments

Figure 3. (**a**) Column-based experimental setup. Black lines on the column indicate sampling intervals of 0–1 cm, 1–2 cm, 2–5 cm and 5–10 cm respectively after incubation. (**b**) Soil columns with forest litter fractions for incubation.

The first factor consisted of the two organic layer types: oak and cedar. The second factor consisted of the organic layer fractions, namely the L- and F- fractions, of each organic layer type. Initial weights of soil columns were determined by the difference of the total weight of soil column and PVC pipe and the weight of the PVC pipe. Dry weights of 3 g L- and 6 g F- organic layer fraction of each organic layer type, per column area, were added to the soil columns labelled as Oak-L, Oak-F, Cedar-L and Cedar-F depending on the organic layer type and fraction added to the soil columns (Figure 3b). The added amounts were based on average amounts of each organic layer fraction, approximately 350 g m^{-2} (3.5 Mg ha^{-1}) and 700 g m^{-2} (7.0 Mg ha^{-1}), respectively, estimated from O-horizon measurements at Osawa watershed in Nihonmatsu City, Fukushima Prefecture (Sasaki, unpublished data). The column treatments were incubated at a temperature of 25 °C for five months from August 2015 to January 2016. The weekly addition of deionized water was performed during the incubation period to maintain soil moisture content. Prior to this, the initial soil moisture content was determined by the difference between the soil fresh weight and dry weight from the additional soil columns sampled. At the

end of the incubation period, the remaining organic layer fractions were separated from soil columns and soils sampled from the pipes at depths of 0–1 cm, 1–2 cm, 2–5 cm and 5–10 cm. Sampled soils were air-dried, crushed, sieved through 2 mm mesh and placed in well-labelled plastic bags for further analyses.

2.3. Laboratory Analyses

2.3.1. Soil Physicochemical Analyses

Soil bulk density was determined by the core method [26]. The pH was measured in a 1:2.5 (*w/v*) ratio of 10 g soil to 25 mL water supernatant suspension, after shaking for 1 h, using a pH meter (PKG-260, Beckman Coulter, Carlsbad, CA, USA) [27]. Total carbon (TC) and total nitrogen (TN) were determined after incubation by combustion. 300 to 500 mg of weighed soil were completely burned using a CN corder (MT-700, Yanaco, Kyoto, Japan). Both TC and TN values were expressed in kg m^{-2}. Bulk density (BD) ranged from 450 to 550 kg m^{-3} and soil pH was very strongly acidic to moderately acidic measuring from 4.9 to 5.6 (Table 1).

Table 1. Basic soil physiochemical properties.

Soil Group (WRB, 2006)	Depth (cm)	Bulk Density (kg m^{-3})	pH (H$_2$O)
Cambisol	0–1	450	4.9
	1–2	450	5.1
	2–5	450	5.5
	5–10	550	5.6

TC and TN measured at each soil depth after the incubation ranged from 2.61 to 1.23 kg m^{-2} and 0.21 to 0.10 kg m^{-2}, respectively, for all treatments (Table 2).

Table 2. Depth measurement of total carbon, total nitrogen and ^{137}Cs in soil columns.

Soil Column Treatment	Depth (cm)	TC [a] (kg m^{-2})	TN [b] (kg m^{-2})	C/N [c]	^{137}Cs (Bq kg^{-1}) [d]	^{137}Cs (Bq m^{-2}) [e]	^{137}Cs Retention [f] (%)
Oak-L	0–1	2.20	0.16	13.8	355 a	1596 a	56
	1–2	1.78	0.14	12.7	168 b	756 b	26
	2–5	1.47	0.12	12.3	67 b	302 b	11
	5–10	1.23	0.11	11.2	38 b	207 b	7
Oak-F	0–1	2.61	0.21	12.4	3050 a	13713 a	91
	1–2	1.89	0.16	11.8	211 b	947 b	6
	2–5	1.40	0.12	11.7	50 b	225 b	2
	5–10	1.75	0.14	12.5	24 b	131 b	1
Cedar-L	0–1	2.01	0.16	12.6	409 a	1842 a	55
	1–2	1.99	0.16	12.4	212 b	953 b	29
	2–5	1.28	0.10	12.8	66 c	295 c	9
	5–10	1.25	0.10	12.5	40 c	221 c	7
Cedar-F	0–1	2.24	0.17	13.2	1523 a	6855 a	71
	1–2	1.87	0.14	13.4	300 b	1349 b	14
	2–5	1.83	0.14	13.1	202 b	908 b	10
	5–10	1.64	0.13	12.6	81 b	446 b	5

[a] Total carbon (TC) content in soil columns [b] Total nitrogen (TN) in soil columns. [c] Carbon-Nitrogen ratio (C/N), [d] ^{137}Cs concentration in soil columns. [e] ^{137}Cs inventory in soil columns. [f] Cs-137 retention is defined as the ratio of the ^{137}Cs inventory in each soil depth layer to the total ^{137}Cs inventory in the soil for each column.

2.3.2. Measurement of ^{137}Cs Activity

The easily recognizable, originally shaped L- and the early fermented and fragmented F- organic layer fractions, as well as the <2 mm soil fractions from each depth sampled after incubation, were placed in U-8 plastic containers and sealed for analysis. The radioactivity of ^{137}Cs in the samples was determined by gamma-ray spectroscopy with gamma-ray

emissions at 662 keV measured using a high purity *n*-type germanium coaxial gamma ray detector (EGC 25-195-R, Canberra-Eurysis, Meriden, CT, USA) connected to an amplifier (PSC822, Canberra, Meriden, CT, USA) and a multichannel analyzer (DSA1000, Canberra, Meriden, CT, USA). The absolute counting efficiency of the detector was calibrated using various weights of IAEA-2006-03 standard soil samples with background correction. Radioactivity of samples was then determined by gamma-ray spectrometry. Each organic layer fraction sample in the U-8 container was measured for 10,800 s, while that of the soil samples was measured for 18,000 to 36,000 s. The counting error ranged between 5–10% for all samples.

2.4. Estimation of Soil Depth Distribution Parameters

The depth distribution of ^{137}Cs in undisturbed soils, including forest soils, was expected to exhibit an exponential decline with depth [28]. Thus the radionuclide concentration at depth χ can be approximated by following the exponential function [29]

$$C(\chi) = C(0).\, e^{-\alpha x} \tag{1}$$

where $C(\chi)$ and $C(0)$ are the activity concentration of the radionuclide (Bq kg^{-1}) at depth (cm) and at χ equal to zero, respectively, and α (cm^{-1}) is the coefficient representing the characteristics of the distribution.

The parameter α depends on the characteristics of the radionuclides, the soil type and the physicochemical characteristics, land use type, time elapsed after deposits and climatic conditions [30]. The value of α corresponds to the inverse ($1/\alpha$) of the relaxation length of ^{137}Cs in the vertical profile representing the shape of the tail of the depth distribution. The depth can also be expressed in mass depth (kg m^{-2}) as the relaxation mass depth, *ho*, describing its penetration strength into soil mass [11,31]. Hence the *ho* value can be estimated by fitting the following exponential function [32] to the empirical data of the reference site as:

$$C(x') = C(0)e^{-x'/ho} \tag{2}$$

where x' is the mass depth from soil surface (kg m^{-2}), $C(x')$ is the concentration of radiocesium at depth x' (Bq kg^{-1}) and $C(0)$ is the radionuclide concentration (Bq kg^{-1}) of the surface soil. The *ho* value was determined using the linearized least square regression method.

2.5. Statistical Analysis

Statistical analysis was performed using the Minitab software program (18th version for Windows 10). The data were subjected to an analysis of variance (ANOVA) test to determine significant differences among the organic layer fractions as well as the soil column treatments at sampled depths. The mean activity concentrations and inventories were separated using the Tukey's Least Significant Difference (LSD) test. The differences between organic layer fractions and organic layer types were compared using Student's *t*-test. A Pearson correlation was performed to show the relationship between soil physicochemical properties and ^{137}Cs retention rate, defined by the % Cs inventory, as the ratio of ^{137}Cs inventory at each soil depth to the total inventory in the column expressed as a percentage. All analyses were done at 5% (and 1% where necessary) significant difference.

3. Results and Discussion

3.1. Cs Retention in Forest Litter Organic Layer Fractions

Within the litter fractions, both activity concentrations and inventory of ^{137}Cs were in the order: oak F > cedar F > oak L, cedar L (F = 884.77, *p* = 0.000) (Figure 4).
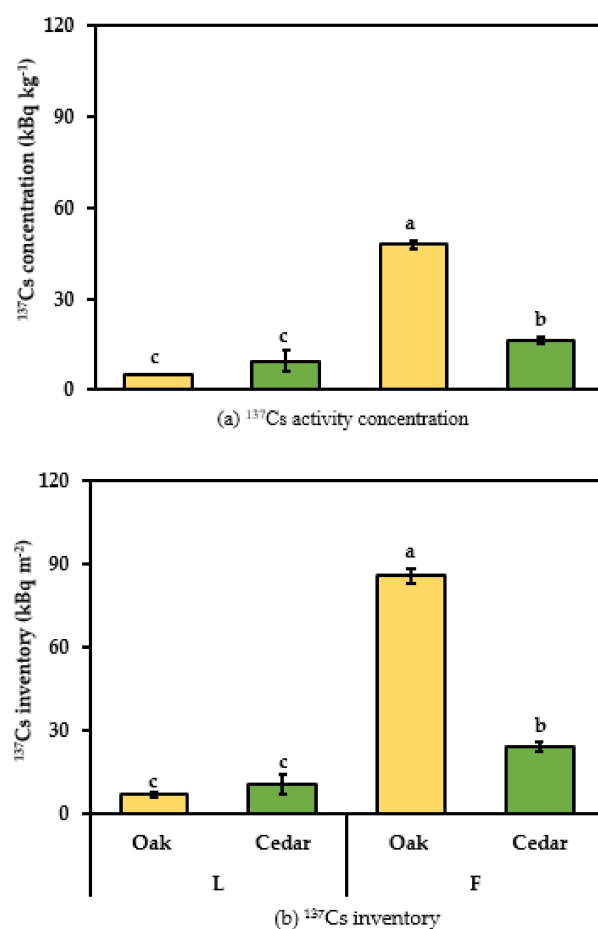
Figure 4. (**a**) ^{137}Cs activity concentration (kBq kg^{-1}) and (**b**) inventory (kBq m^{-2}) in the litter organic layer fractions. Different letters indicate significant difference ($p < 0.05$) within each figure.

The oak F layer fraction accumulated the highest activity concentration and inventory of 47.9 ± 1.3 kBq kg^{-1} and 85.9 ± 2.5 kBq m^{-2} respectively, and was significantly higher (t = 33.89, $p < 0.05$) than the cedar F fraction of 16.4 ± 0.9 kBq kg^{-1} and 24.4 ± 1.6 kBq m^{-2}, whereas both oak and cedar L layer fractions showed no significant differences in both ^{137}Cs concentrations and inventories (Figure 3). Reports from two independent, preliminary surveys conducted by the Forestry and Forest Products Research Institute (FFPRI) and Forestry Agency of Japan (FAJ) and by the Ministry of Education, Culture, Sports, Science and Technology (MEXT), in August and September 2011, revealed that the contribution of litter components to total cesium proportions was low for Japanese cedar forests (*C. japonica*; evergreen needleleaf forests) and high for deciduous oak forests (*Q. serrata*; deciduous broadleaf forests [13]. In this study, a similar trend was observed, and was suggested that due to no leafing of the trees in deciduous oak forests, the large proportion of ^{137}Cs was by direct disposition on the forest floor [33]. In addition, trapped ^{137}Cs in the litter component of evergreen needleleaf forests is not currently as high as that in the deciduous broadleaf forests, as nearly 40 % of the total cesium is retained in the leaves attached to the trees in the forests [12,34].

More than 80 % (55.1 ± 33.7 kBq m^{-2}) accumulation of ^{137}Cs implies that there is higher retention in F layer fractions compared to that of the L organic layer fraction (8.8 ± 3.0 kBq m^{-2}) (Figure 4b) which is most possibly due to transfer from the L layer fraction to the F layer fraction via mechanical and biological breakdown [35].This was in agreement with results obtained by Koarashi and Atarashi-Andoh [16] for litter fractions sampled in the same year as this study. According to Koarashi and Atarashi-Andoh [16], the F fractions acted as a dominant pool of ^{137}Cs in the organic layers in July 2015 due to

leaching from the organic layers by rain and litter decomposition over the years (2011–2015). This is also due to the F layer fraction having more residual litterfall with high ^{137}Cs amounts at the time of deposition. Furthermore, study results from Koarashi et al. [36] showed a significant positive correlation between the ^{137}Cs inventory and the litter material inventory in the L and F litter organic layers at both deciduous broadleaf forest (DBF) and cedar-dominated coniferous forest (CF) sites; the ^{137}Cs inventory tended to increase linearly with increasing litter-material inventory irrespective of the litter layer type at the CF site while the ^{137}Cs inventories for the L and F layers were not observed to be approximated by a single linear relationship at the DBF sites. It was indicated, from the observations made from the study, that the larger accumulation of ^{137}Cs in the organic layer at the CF site could simply be described by the larger accumulation of litter materials on the forest floor, whereas that at the DBF site depended more on the larger accumulation of highly ^{137}Cs-contaminated materials in the F layer [36]. These explanations were suggested to be the reason for the significantly higher amounts (t = 4.02; p = 0.02) retained in the F litter organic layer fraction compared to the L litter organic layer fraction in this study. Thus, litter transformation contributes to a major potential long-term source of particularly mobile ^{137}Cs and the continuously decomposing litter is continuously transferred into the soil component [1,13].

3.2. Radioactive ^{137}Cs in Soil Profile Layers

The activity concentration and inventory of ^{137}Cs were significantly ($p < 0.05$) highest at the 0–1 cm depth for all treatments, with no significant difference among soil depths below 0–1 cm (Table 2). An exception was observed in the ^{137}Cs mean concentration and inventory for Cedar-L, in which the 1–2 cm depth was significantly different (F = 29.49; p = 0.00) from depths 2–5 and 5–10 cm (Table 2).

For ^{137}Cs retention, about 80% of the total ^{137}Cs inventory within the soil profiles was distributed in the top 0–1 cm depth, and approximately 90% absorbed above the 2 cm soil depth; of this total, both Oak-L and Cedar-L contributed to 66% and 74% of the distributed ^{137}Cs inventory in the 0–1 cm depth and that absorbed above the 2 cm soil depth, respectively. For the Oak-F and Cedar-F soil profiles, 91% and 71% of the ^{137}Cs inventory were distributed in the upper 0–1 cm depth, with 97% and 85% absorbed in the upper 2 cm depth, respectively (Table 2). Furthermore, about 3% of the ^{137}Cs inventory within Oak-F was below the 2 cm depth while 15% of the ^{137}Cs inventory was below the same depth within Cedar-F. In comparison to the L-type treated soil profile, Oak-L and Cedar-L, 55-56% of the ^{137}Cs inventory were contained in the 0–1cm depth and 82–84% absorbed in the upper 2 cm depth; 15–16% of the distributed ^{137}Cs inventory was below the 2 cm depth of Oak-F and Cedar-F, respectively (Table 2). The results confirm a greater ^{137}Cs retention at the surface depth of F litter fraction soils, especially Oak-F, and a deeper penetration in L litter fraction soils (Oak-L and Cedar-L).

3.3. Depth Distribution of ^{137}Cs in Soil Profile Layers

The depth distribution of ^{137}Cs for all treatments are illustrated in Figure 5.

The ^{137}Cs concentrations at the surface depth (0–1 cm) were 355 Bq kg^{-1} for Oak-L, 3050 Bq kg^{-1} for Oak-F, 409 Bq kg^{-1} Cedar-L and 1523 Bq kg^{-1} for Cedar-F and were the peaks for all treatments. All treatments exhibited a general decreasing pattern with increasing depth. The profile shape created as a result of the observed distribution pattern is a typical reflection of the nature of radiocesium adsorption along the depth [10]. Both Oak-F and Cedar-F exhibited a sharp decline in ^{137}Cs distribution from the 0–1 cm depth into deeper layers. A similar trend was observed in Cedar-L, whereas Oak-L exhibited a rather slow and gradual decline with depth especially within the 1–5 cm soil depth. It was evident from the results that ^{137}Cs tended to dominate the deeper layers of Cedar-F measuring the highest concentrations among all the soil profiles of 300 Bq kg^{-1}, 202 Bq kg^{-1} and 81 Bq kg^{-1} at 1–2 cm, 2–5 cm and 5–10 cm depths.
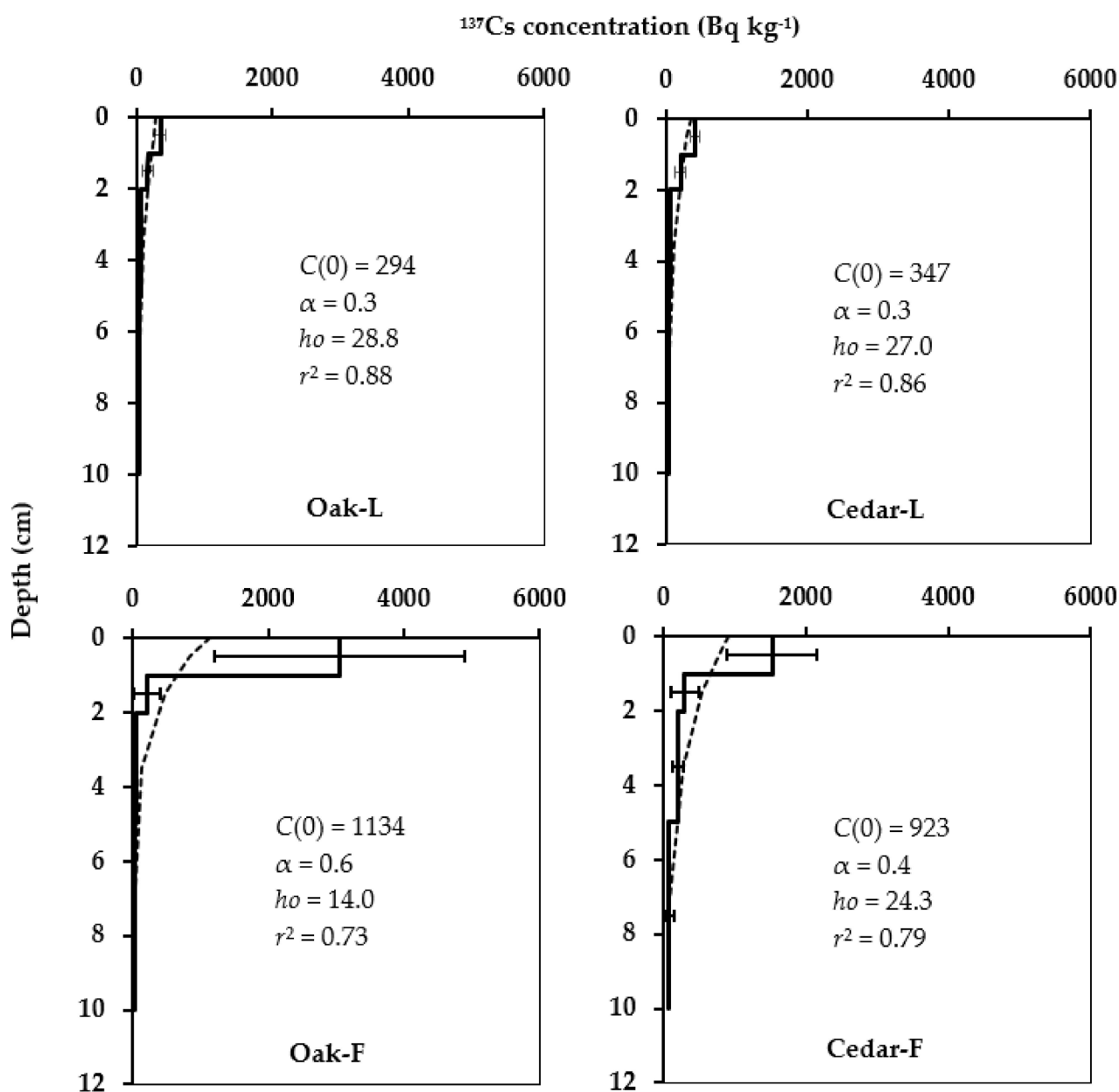
Figure 5. The depth distribution of the ^{137}Cs activity concentration derived from forest litter layer fractions in soil columns; Oak-L, Cedar-L, Oak-F and Cedar-F ^{137}Cs concentration profiles. $C(0)$ is the ^{137}Cs concentration (Bq kg^{-1}) at the surface soil (x = 0), α is the inverse of relaxation depth (cm^{-1}) and *ho* is the relaxation mass depth (kg m^{-2}). The black solid line indicated measured 137Cs concentration and the broken lines indicated the ^{137}Cs concentration fitted by Equation (1).

The relaxation mass depth, *ho*, obtained from distribution fitting by Equation (1), for both Oak-L and Cedar-L were almost the same of the values 28.8 kg m^{-2} and 27.0 kg m^{-2}, with Oak-F and Cedar-F having values of 14.0 kg m^{-2}, and 24.3 kg m^{-2}, respectively (Figure 4). From the results, deeper penetration of ^{137}Cs was observed in L organic fraction treated soils (Oak-L and Cedar-L) compared to the F organic fraction treated soils (Oak-F and Cedar-F). A number of column experiments, similar to that of this study, showed that the depth of initial penetration of ^{137}Cs was influenced by the intensity of the rainfall during the wet deposition [37]. Additionally, larger amounts of precipitation can increase ^{137}Cs leaching, especially in organic soils [38]. The L fractions (Oak-L and Cedar-L), comprised of freshly fallen leaves, with low ^{137}Cs penetrated first and deeply because of the water soluble nature of the ^{137}Cs. The water soluble ^{137}Cs was evident that leaching or infiltration

by the added water is the possible reason for ^{137}Cs penetration into the deeper depths of Oak-L and Cedar-L. Furthermore, ^{137}Cs deeper penetration, similar to that of the L organic fraction treated soils, was observed in Cedar-F while greater ^{137}Cs retention was observed in Oak-F. It was implied that the organic matter effect may have contributed to ^{137}Cs retention at the soil surface of Oak-F. Similar to Teramage et al. [11], the *ho* values for ^{137}Cs migration could not be compared directly to results from the Chernobyl accident mainly due to values from agricultural fields [39,40]. The depth distribution parameters were, however, compared to previous field studies of Fukushima-derived ^{137}Cs to evaluate the migration trend obtained in this study. Koarashi et al. [1] reported a relaxation length of 1.4–2.9 cm and a relaxation mass depth of 7.4–10.9 kg m^{-2} when investigating forest land uses. Additionally, a relaxation length of 0.8–2.7 cm and a relaxation mass depth of 2.2–10.2 kg m^{-2} was recorded by [41]. The α values obtained in both studies were almost consistent with that of this study. Study results by Teramage et al. [11] in coniferous forest soil revealed the same α as Oak-F in this study, of 0.6 (1.7 cm relaxation length), and an *ho* value of 11.1 kg m^{-2} for Fukushima-derived [137]Cs. Given the difference in methodology between the previous field studies [1,11,41] and this column-based experimental study, owing to the comparatively high *ho* values of the latter, a similar trend of increased or rapid migration of ^{137}Cs in the coniferous cedar forest soils was observed. Direct comparison of *ho* of radiocesium migration with those of previous studies seems difficult because the previously reported values are either from agricultural fields or are from studies that were conducted several years after the Chernobyl accident.

3.4. The Effect of Soil Properties on [137]Cs Retention in Soils

The depth distribution patterns of ^{137}Cs in the soils were considered to be influenced by soil properties. From the correlation coefficients, results between ^{137}Cs retention and the selected soil physiochemical properties summarized in Table 3, ^{137}Cs retention in Oak-L and Cedar-L showed a significant negative correlation with bulk density, mass depth and soil pH ($p < 0.05$) with no significant correlation in Oak-F and Cedar-F.

Table 3. Correlation analysis between ^{137}Cs retention and soil physiochemical properties in the soil columns ($n = 3$).

Soil Physiochemical Properties	[137]Cs Retention (%)			
	Oak-L	Oak-F	Cedar-L	Cedar-F
Bulk density (kg m^{-3})	−0.969 *	−0.801	−0.985 *	−0.834
Mass depth (kg m^{-2})	−0.990 **	−0.901	−0.979 *	−0.936 *
pH	−0.944 *	−0.788	−0.967 *	−0.823
Total carbon (kg m^{-2})	0.976 *	0.925	0.889	0.956 *
Total nitrogen (kg m^{-2})	0.987 *	0.910	0.863	0.973 *

* and ** are significant correlation at $p < 0.05$ and $p < 0.01$ probability levels.

A distinguishing observation was made in Oak-L in which ^{137}Cs retention strongly (and negatively) correlated ($p < 0.01$) with mass depth. Again, ^{137}Cs retention positively and significantly correlated with TC and TN in Oak-L and Cedar-F ($p < 0.05$), while no significant positive correlation was observed in Oak-F and Cedar-L (Table 3). A similar result to that of the correlation analysis was obtained from the regression analysis between ^{137}Cs retention and TC (Figure 6).

The obtained results indicate the possible influence of organic matter on ^{137}Cs retention. Organic matter, like clay minerals, is typically negatively charged (and thus has a high CEC), and therefore has the ability of attracting and retaining [137]Cs. However, organic matter exhibits little selectivity for ^{137}Cs for adsorption in comparison to clay minerals, and hence its ^{137}Cs adsorption is rather weak and fully reversible in competition with other cations [42–45]. The C/N of the F treatments revealed higher values for Cedar-F, particularly at the 1–3 cm depth, than that of Oak-F (Table 2). The high C/N specifies that the organic matter decomposition of the cedar L fraction does not advance. Hence, it is thought

that the organic matter of high C/N, including ^{137}Cs, penetrates deeper, as observed in Cedar-F, due to their coarse particle size.
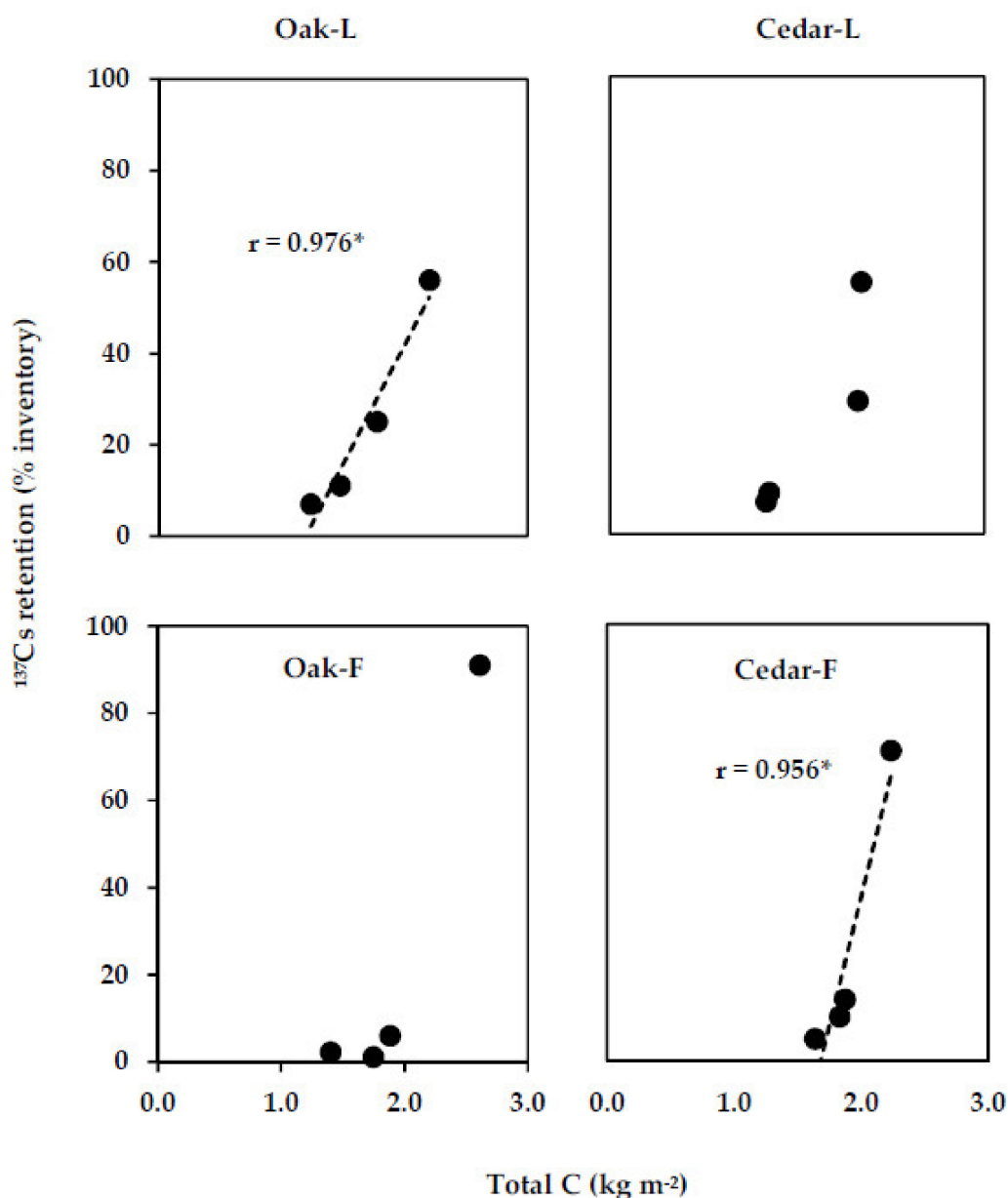

Figure 6. Regression analysis between ^{137}Cs retention (% inventory) and total carbon (TC) in soil columns. * is significant at $p < 0.05$ probability level.

On the other hand, the organic matter of the oak F fraction in which the decomposition advanced, owing to the comparatively low C/N, releases large amounts of ^{137}Cs. However, the regression results, which showed no significant relationship between ^{137}Cs retention and TC as observed in Oak-F (and Cedar-L) (Figure 6), indicates that organic matter plays a weak and indirect role in the processes of both ^{137}Cs retention and penetration, and that finer-sized soil particles tend to strongly and directly influence the retention of ^{137}Cs in soils [1]. According to Takahashi et al. [41] the distribution parameters α and β, described as *ho* in this study, calculated by the exponential equation from their study tended to be correlated with radiocesium interception potential (RIP), macro pore (gravitational pore space), and dispersible fine particle content, indicating that the initial distribution would have been influenced by ^{137}Cs fixation ability of soil and the penetration process of

water and particles in soils. Although it was unclear in this study, it was suggested that the released ^{137}Cs from the advanced decomposition of the oak F fraction were held by adsorption onto RIP of the soil surface (0–1cm). Most downward migration models consider a single phase of migration rate that assumes a gradual and slow adsorption-desorption process of radiocesium movement in the soil profile [11]. Nonetheless, the migration of some fractions of Fukushima-derived ^{137}Cs into deeper horizons of coniferous forest soils observed by Teramage et al. [11] was mainly attributed to the infiltration of radiocesium-circumscribed rainwater during the fallout before selective adsorption started, and it was inferred that an additional and quick phase of radiocesium migration occurs in forest soil. A similar phenomenon due to infiltration by water within the early months of incubation could be suggested for the faster penetration and deeper retention of released ^{137}Cs from the coarse cedar F sized organic fraction in Cedar-F, owing to the significant effect ($p < 0.05$) of TC content on ^{137}Cs retention, in contrast to the slow penetration by water of ^{137}Cs from the finer oak F sized organic fraction in Oak-F, which showed no significant effect of TC content on ^{137}Cs retention (Figure 6).

It is therefore important to consider parameters such as finer-sized soil particles, diffusion and migration rates for futuristic understanding and clarity of the influence of forest organic layer fractions on vertical distribution processes.

4. Conclusions

In this study, we investigated the influence of Fukushima-derived L and F forest litter organic layer fractions of both oak and cedar on the ^{137}Cs vertical distribution in forest soil profiles using the laboratory column-based method. The following conclusions were made from the results obtained:

The L organic fraction is a large storage for relatively longer ^{137}Cs retention, more dominantly in the coniferous cedar than in the deciduous oak, while the F organic fraction influences ^{137}Cs transfer from litter to soil minerals and its distribution within the soil profile via seepage. The slower rate of litter decomposition of the cedar L fraction allows for greater accumulation of ^{137}Cs within the litter organic layer fraction. In contrast, the faster rate of litter decomposition, resulting in transformation of the oak L to a fine particle-sized F, showed that deciduous forest organic matter releases large amounts of ^{137}Cs which remains distributed at the topsoil. Therefore, future ^{137}Cs soil distribution within deciduous oak forests will continue to be limited within the top layer soil as leaching by water decreases with time. As opposed to the deciduous forest, the coarse particle-sized F cedar organic fraction potentially aids deeper distribution of ^{137}Cs within soil mineral layers with increasing futuristic vertical distribution or migration.

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