

# Vertical distribution and temporal changes of $^{137}\text{Cs}$ in soil profiles under various land uses after the Fukushima Dai-ichi Nuclear Power Plant Accident

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## Abstract

We monitored the vertical distribution of  $^{137}\text{Cs}$  in soil profiles under eight different land uses for the 2 y after the Fukushima Dai-ichi Nuclear Power Plant accident, and discussed the temporal changes in the early-stage of the migration and the determinants of the initial distribution. The soil samples were collected for four surveys using a scraper plate at each study site, which consisted of three forests (mixed forest, mature cedar, and young cedar), two grasslands (pasture and meadow) and three abandoned agricultural fields (farm land, tobacco field, and paddy field). The land use patterns have a large influence on some soil properties and the migration processes of  $^{137}\text{Cs}$  above ground, resulting in different distribution of  $^{137}\text{Cs}$  in those soil profiles. Specifically, the secondary deposition of  $^{137}\text{Cs}$  from the coniferous canopy, retention of  $^{137}\text{Cs}$  by litter layer, and the homogenization of  $^{137}\text{Cs}$  concentrations in surface soil by natural soil mixing such as the disturbance by cattle grazing, roots growing and the formation of needle ice were important to cause redistribution of the deposited  $^{137}\text{Cs}$ . Only in the paddy field, the  $^{137}\text{Cs}$  inventory in subsurface soils (5-10 cm) gradually increased and comprised 26% of the total  $^{137}\text{Cs}$  in 2 y, showing the downward migration of  $^{137}\text{Cs}$  to subsurface soil. In the other sites, it was considered that  $^{137}\text{Cs}$  were strongly adsorbed by soil particles and rarely migrated downward as soluble form. Vertical distributions during the first survey were able to be used as the initial distributions and were well fitted to the exponential equation. The distribution parameters  $\alpha$  (relaxation depth) and  $\beta$  (relaxation mass depth), calculated by the exponential equation were correlated with RIP ( $r = -0.806$ ,  $p < 0.05$ ), macro pore ( $r = 0.651$ ,  $p = 0.11$ ), and dispersible fine particle content ( $r = 0.856$ ,  $p < 0.05$ ). It indicated that the initial distribution would be influenced by the Cs fixation ability of soil, and the penetration process of water and particles in soils.

## 1. Introduction

The accident at the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) in March 2011 led to the enormous release of radionuclides into the environment. Since then, many basic monitoring data such as the radioactivities of air, water, soil and food has been collected and soil decontamination work proceeds. Now it is strongly required to predict migration of deposited radionuclides in order to establish strategies for management and recovery of the contaminated land while reducing the health risk from contaminated water and food.

Of all the radionuclides released by the FDNPP accident,  $^{137}\text{Cs}$  is the most important long-term contributor to the soil contamination because of its high release rate, estimated at  $1.3 \times 10^{16}$  Bq (Chino et al., 2011), and long half-life (30.1 y). In addition, it is well known that the cesium ion is selectively adsorbed on frayed-edges sites (FES) of 2:1 phyllosilicates in soils (Sawhney, 1972), hence remains in the soil surface for a long time (e.g. Schuller et al., 2004). In fact, after the FDNPP accident, several studies have shown that most of  $^{137}\text{Cs}$  are retained in the litter layer and soil surface (e.g. Kato et al., 2012a; Shiozawa, 2013; Koarashi et al., 2012 and Matsunaga et al., 2013). Therefore, the behavior of  $^{137}\text{Cs}$  in soils is the key factor in determining the fate of deposited  $^{137}\text{Cs}$  on terrestrial ecosystems.

After various nuclear weapons tests and the Chernobyl accident, numerous findings on the fate of  $^{137}\text{Cs}$  in soils have already been obtained. For example, the rate of downward migration of  $^{137}\text{Cs}$

decreases with time and varies with soil type, texture, or water condition due to the fixation of  $^{137}\text{Cs}$  to soil particles (e.g. Rosén et al., 1999; Arapis et al., 1997). While, Forsberg and Strandmark (2001) showed the presence of available  $^{137}\text{Cs}$  in deeper layers of 36 y old contaminated soils, indicating a small possibility of continuing downward migration to deeper soil layers even after decades. According to Tyler et al. (2001), the processes of bioturbation such as soil mixing and reconstitution by earthworm movement, ingestion, and excretion are important factors in the long-term downward migration in some upland soils. However, little is known about how the deposited  $^{137}\text{Cs}$  is distributed and migrates in the soil profile at the very early stage. Schimmack et al. (1989) reported that the vertical distribution of  $^{137}\text{Cs}$  in soil at May 7, 1986 when the first rainy day after the wet deposition of the radionuclide from Chernobyl accident, showed similar patterns to those observed four months later. It indicated that very early precipitation determines the initial distribution of  $^{137}\text{Cs}$ , however few other details, such as the relationship with certain soil properties, have been revealed.

These findings of  $^{137}\text{Cs}$  migration in soils are obtained based on the investigation of vertical distribution in soil profiles. The method of soil sampling used can affect findings because small amounts of contamination have a large influence on the analysis of the  $^{137}\text{Cs}$  concentration. According to Isaksson (1997), there are three main methods to collect the vertically sectioned soil samples. These are the bore core method, the trench method and the

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template method. The bore core method is easy and rapid but has a high risk of cross contamination and soil compression compared to other methods. The trench method has the lowest risk of cross contamination, but it is almost impossible to take the soil samples by fine depth increments. The template method is very time consuming, but has relatively low risk of cross contamination and has the ability to obtain large amounts of soil by fine depth increments. In particular, the template method using the scraper plate proposed by Campbell et al. (1988) is possible to take soil samples by 5 mm increments near the soil surface. Therefore, this is described as the optimum method in providing accurate and detailed information on the vertical distribution of <sup>137</sup>Cs, as the concentration exponentially declines with depth (Loughran et al., 2003). Although this method has now been widely adopted in estimating soil erosion and sediment accumulation rates, there is little information about the vertical distribution of <sup>137</sup>Cs and its temporal changes as measured using the scraper plate within the first few years of deposition, including the case of FDNPP.

Therefore, in the present study, we monitored the detailed vertical distributions of <sup>137</sup>Cs in soil profiles using a scraper plate under eight different land uses four times in 2 y after the FDNPP accident. Characteristics of the very early-stage temporal changes and the determinants of the initial distribution in soils were sought. In particular, we focused on the difference in land use patterns because some soil properties, the migration processes of those soils, and the necessary decontamination involved, vary with differing land use patterns.

## 2. Materials and methods

### 2.1 Study sites

We selected the eight study sites under various land uses (three

forests, two grasslands and three abandoned agricultural fields) in Kawamata Town, Yamakiya District, in the northern part of the Fukushima Prefecture (Fig. 1). This region is located roughly 40 km north-west of the FDNPP and has been highly contaminated by the deposited radiocesium (about 0.3-3.0 MBq m<sup>-2</sup>; MEXT, 2011). The Yamakiya District was included in the evacuation zone until August 2013.

Information of study sites are summarized in Table 1. The mixed forest (MF) site was dominated by *Pinus densiflora* and *Quercus* sp., among the typical secondary forest found in the warm-temperate zone of Japan. *Cryptomeria Japonica*, the most common plantation species in Japan, dominated the mature cedar (MC) and young cedar (YC) sites. Stem densities of the mature (33 y old) and young (17 y old) cedar stands were 1250 trees ha<sup>-1</sup> and 2600 trees ha<sup>-1</sup>, respectively. The pasture land (PL) site was used for cattle grazing before the FDNPP accident, but the cattle were moved with the evacuation of the residents. The meadow land (ML) site was used for meadow gathering. The farm land (FL), tobacco field (TF), and paddy field (PF) sites were uncultivated after the FDNPP accident. Therefore, various weeds such as *Artemisia indica* var. *maximowiczii*, *Solidago altissim*, and *Juncus effusus* var. *decipens* cover them throughout the sampling periods, although these sites were almost bare land when the accident occurred.

According to Comprehensive Soil Classification System of Japan First Approximation (Obara et al., 2011), the three forest soils, the PF soil, and all other soils were classified as Andosol, Brown Lowland soil, and Reformed soil, respectively. Most soil in this region was affected by deposition of volcanic ash from Mt. Adatara and the surrounding volcanoes. Andosols (volcanic ash soils) should be more prevalent, but due to massive development,

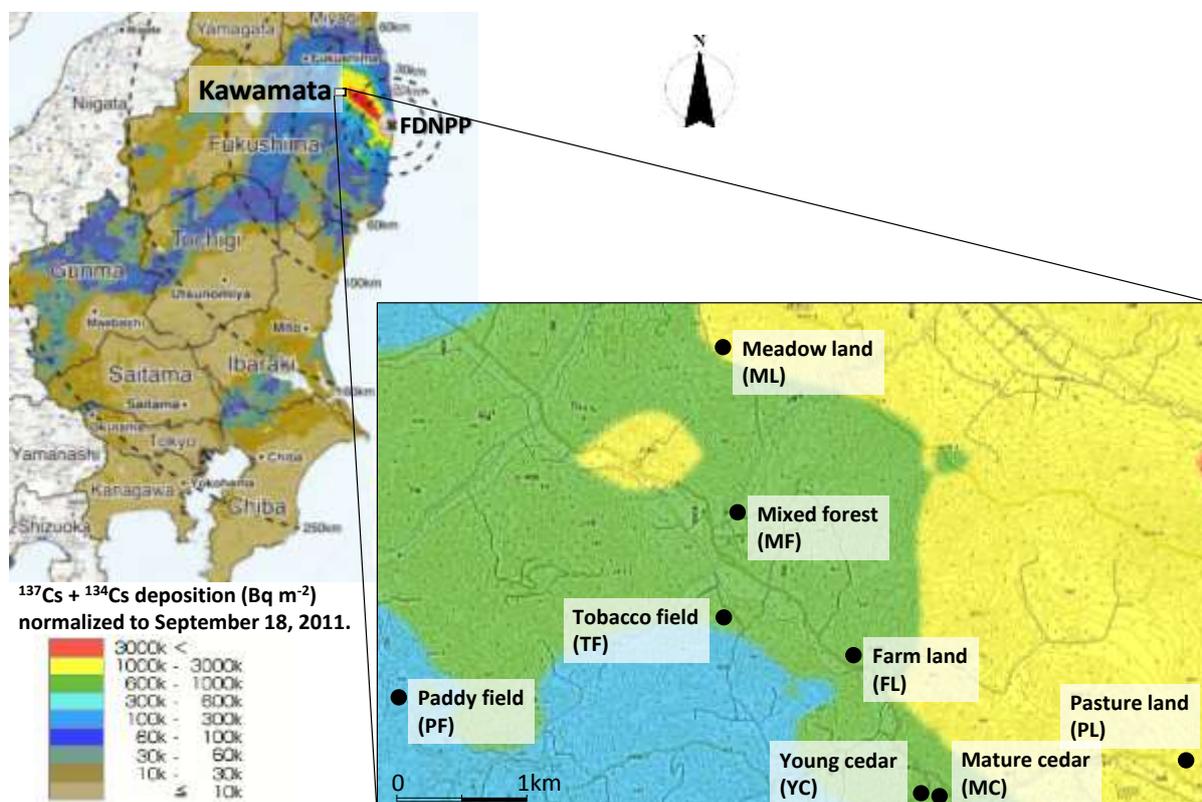


Fig. 1 Location of study sites. The <sup>137</sup>Cs deposition map was based on the third airborne monitoring survey of radioactivity by MEXT (2011).

**Table 1** Study site information.

Site	Land uses	Lat / Long	Altitude (m)	Soil Group <sup>a</sup>	Present vegetation <sup>b</sup>
<b>Mixed forest (MF)</b>	Secondary forest	N 37.6026 E 140.6773	580 m	Andosol	<i>Pinus densiflora</i> <i>Quercus</i> sp.
<b>Mature cedar (MC)</b>	Plantation	N 37.5852 E 140.6906	552 m	Andosol	<i>Cryptomeria japonica</i>
<b>Young cedar (YC)</b>	Plantation	N 37.5861 E 140.6905	556 m	Andosol	<i>Cryptomeria japonica</i>
<b>Pasture land (PL)</b>	Cattle grazing	N 37.5860 E 140.7174	582 m	Reformed soil	<i>Poa pratensis</i>
<b>Meadow land (ML)</b>	Meadow gathering	N 37.6176 E 140.6742	679 m	Reformed soil	<i>Poaceae</i> sp.
<b>Farm land (FL)</b>	Vegetable farming	N 37.5986 E 140.6804	528 m	Reformed soil	<i>Artemisia indica</i> var. <i>maximowiczii</i> <i>Solidago altissima</i>
<b>Tobacco field (TF)</b>	Cash cropping	N 37.5979 E 140.6741	542 m	Reformed soil	<i>Artemisia indica</i> var. <i>maximowiczii</i> <i>Solidago altissima</i>
<b>Paddy field (PF)</b>	Wet-paddy rice cropping	N 37.5923 E 140.6437	588 m	Brown Lowland soil	<i>Juncus effusus</i> var. <i>decipens</i> <i>Solidago altissima</i>

a: According to Comprehensive Soil Classification System of Japan First Approximation (Obara et al., 2011).

b: There were no vegetation, and little or no litters in TF and PF sites but a few withered crop residue in FL site as at the FDNPP accident.

many Andosols were buried under subsurface soil, and making Reformed soils (Fukushima Prefecture, 1988). The PF site is located in a valley plain, with a G horizon under 70 cm in depth due to the effects of groundwater.

2.2 Method of soil sampling using the scraper plate

In order to study the temporal changes in the vertical distribution of <sup>137</sup>Cs, four soil surveys were conducted from summer 2011 to winter 2012. Each sampling date is shown in Table 2. An appropriate sampling plot with an area of 10 m<sup>2</sup> at each site was located far away from the road, tree trunk and canopy gap, and separated by a rope fence in order to prevent disturbance by wild animals. One sampling was conducted within this plot for each survey.

The vertically sectioned soil samples were collected using a scraper plate. This device has two components: a metal frame with a sampling area of 450 cm<sup>2</sup> (15 cm × 30 cm) that is placed on the ground, and an adjustable metal plate that can scrape or remove fixed increments of soil within the frame. Any litters or living vegetation contained within the frame was collected separately. In forest sites, the frame was placed on the litter layers with no living vegetation. The litters were cut along the inside of the frame with scissors and knife, and collected with the hands little by little. It was easily possible to distinguish the border between litter layer and soil surface because there is no Oa horizon that is well-decomposed organic matter in this temperate area. After the litters within the frame were completely collected, the frame was reset on the soil surface with removal of surrounding litters. In grassland and agricultural field sites, litters was collected in the similar way to forest site and living vegetation was also collected by cutting from ground level without plucking. In addition, if the soil surface was uneven and protruded above the datum of the frame, it was collected as most surface soil (0-5 mm). The soil samples were taken in 5 mm increments between 0 and 5 cm and 1.0 cm increments between 5 and 10 cm. Roots were cut and collected with soil. Gravel was carefully excavated and collected with the closest corresponding soil layer. We only conducted one sampling for each survey. According to Khomutinin et al. (2004), who reported the relationship between variability of <sup>137</sup>Cs inventory and sampling area, it is possible to consider a soil sample obtained with sampler of large area as several samples next to each other

from the same area obtained with sampler of smaller area. In addition, they clarified that the standard deviation of logarithm of <sup>137</sup>Cs inventory become smaller with increasing sampling area, and does not depend on its amount of inventory, type of fallout and land uses for the sampling areas > 50 cm<sup>2</sup>. Moreover, we selected the sampling plots that are far away from the road, tree trunk and canopy gap in order to avoid the spatial singularity of <sup>137</sup>Cs deposition at the macro level. The sampler of large area also can avoid the singularity at the micro level caused by soil animals or roots. Therefore, we recognized one sampling using the scraper plate with a large area of 450 cm<sup>2</sup> as the representative of each survey.

2.3 Measurement of radiocesium in samples

All samples taken by the scraper plate were partially dried at 110°C for 24 h. Litter samples were fragmented after drying. The samples were passed through a 2-mm sieve and placed in a plastic container (U8 container: diameter = 47 mm, height = 60 mm). The radioactivity of <sup>137</sup>Cs in the samples was determined by gamma-ray spectroscopy. Gamma-ray emissions at 662 keV was measured using a high purity n-type germanium coaxial gamma-ray detector (EGC25-195-R, Canberra-Eurisys, Meriden, U.S.A.) coupled to an amplifier (PSC822, Canberra, Meriden, U.S.A.) and multichannel analyzer (DSA1000, Canberra, Meriden, U.S.A.).

**Table 2** Sampling date for each survey period.

Site	1st survey	2nd survey	3rd survey	4th survey
<b>Mixed forest (MF)</b>	July 1, 2011	Jan. 10, 2012	Aug. 27, 2012	Dec. 11, 2012
<b>Mature cedar (MC)</b>	June 30, 2011	Jan. 10, 2012	Aug. 28, 2012	Dec. 13, 2012
<b>Young cedar (YC)</b>	July 2, 2011	Jan. 17, 2012	Aug. 28, 2012	Dec. 13, 2012
<b>Pasture land (PL)</b>	June 21, 2011	Jan. 18, 2012	Sep. 22, 2012	Dec. 12, 2012
<b>Meadow land (ML)</b>	June 21, 2011	Jan. 18, 2012	Aug. 29, 2012	Dec. 11, 2012
<b>Farm land (FL)</b>	June 27, 2011	Jan. 25, 2012	Aug. 29, 2012	Dec. 12, 2012
<b>Tobacco field (TF)</b>	June 27, 2011	Jan. 25, 2012	Aug. 29, 2012	Dec. 18, 2012
<b>Paddy field (PF)</b>	June 28, 2011	Dec. 8, 2011	Aug. 29, 2012	Dec. 19, 2012

The measurement system was calibrated using standard gamma sources with different sample heights, and the self-absorption of gamma-rays by the sample were corrected (Cutshall et al., 1983). Details of the standard gamma sources were described by Kato et al. (2012a). Single sample in U8 container for each layer was measured one time and its counting error was less than 10%. The minimum detectable amount (MDA value) calculated from the background counts for April 2011 to November 2012 was  $8 \times 10^{-3}$  Bq.

#### 2.4 Measurements of chemical and physical properties of soil samples

In order to determine some chemical and physical properties of soil samples, undisturbed soil samples of surface horizon (0-5 cm) were collected using a 100 mL core sampler during the first survey. Volumes of the solid and liquid phases were determined using a digital actual volumeter (DIK-1150, Daiki, Tokyo, Japan). The gaseous phase was calculated as the remainder. After gravitative drainage, the weight of the soil sample with capillary water was measured and the saturated hydraulic conductivity was tested with a falling head permeameter (DIK-4050, Daiki, Tokyo, Japan). The samples were then oven-dried at 105°C for 24 h or more. The moisture was then determined in order to calculate micro pore (capillary pore space), macro pore (gravitational pore space), and bulk density.

Other core samples were air-dried and passed through a 2 mm or 0.5 mm sieve before chemical analysis. Soil pH was measured in H<sub>2</sub>O (soil: solution, 1: 2.5) by the glass electrode method. Total carbon (TC) was determined by the dry combustion method using an NC analyzer (Sumigraph NC-900; Sumika Chemical Analysis Service, Ltd., Osaka, Japan). Cation exchange capacity (CEC) was estimated by the ammonium acetate saturation method at pH 7.0. Radiocesium interception potential (RIP) proposed by Cremers et al. (1988) was determined as a quantitative index of the FES calculated from the solid/liquid distribution coefficient of Cs and the K concentration in solution. According to Wauters et al. (1996), the RIP is described as

$$\text{RIP (mol kg}^{-1}\text{)} = K_D^{\text{Cs}} \times \text{mK} = K_c^{\text{FES}}(^{137}\text{Cs/K}) \times [\text{FES}]$$

where  $K_D^{\text{Cs}}$  is the solid/liquid distribution coefficient of Cs (L kg<sup>-1</sup>), mK is the K concentration in solution (mol L<sup>-1</sup>),  $K_c^{\text{FES}}(^{137}\text{Cs/K})$  is the selectivity coefficient of Cs against K in the FES, which is constant and estimated to be 1000, and [FES] is the concentration of FES in the soil (mol kg<sup>-1</sup>). Based on the method of Wauters et al. (1996),  $K_D^{\text{Cs}}$  was determined as follows. A 1 g portion of each soil sample was weighed into a dialysis bag (cellulose dialysis membrane, Wako Chemicals, VA, USA) with 5 mL of Ca-K solution (i.e. 0.1 mol L<sup>-1</sup> CaCl<sub>2</sub> and 0.5 mmol L<sup>-1</sup> KCl). The dialysis bag was transferred to a plastic bottle with 100 mL of Ca-K solution, and then shaken for 2 h twice a day for 5 d. The outer solution was renewed each time before shaking. The dialysis bag was transferred to a plastic bottle with 94 mL of Ca-K solution and 1 mL of <sup>137</sup>Cs solution (i.e. 5 kBq mL<sup>-1</sup> of carrier-free <sup>137</sup>Cs). After continuous shaking for 5 d, the activity concentration of <sup>137</sup>Cs in the outer solution was measured by a liquid scintillation counter (AccuFLEX γ7001, Hitachi Aloka Medical, Ltd., Tokyo, Japan) to determine  $K_D^{\text{Cs}}$ . The RIP (i.e.  $K_D^{\text{Cs}} \times \text{mK}$ ) was calculated assuming that the K concentration in the solution was constant (0.5 mmol L<sup>-1</sup>) throughout the experiment.

We also measured the particle size distributions in the upper most surface soils (0-0.5 cm) sampled by scraper plate. The particle size distributions were analyzed by sieve (>450 μm) and a laser diffraction particle size analyzer (SALD-3100, Shimadzu Co., Ltd., Kyoto, Japan) without the decomposition treatment of soil organic matters. Therefore, these finer particles represented the amount of dispersible particles, rather than clay minerals, in a natural state.

#### 2.5 Estimation of vertical distribution parameters

In the case of fresh fallout, the vertical distribution of <sup>137</sup>Cs in undisturbed soil is expected to exhibit an exponential decline with depth. The radionuclide concentration at depth  $\chi$  can be approximated with the following exponential function (Beck, 1966):

$$C(\chi) = C(0) \cdot e^{-\frac{\chi}{\alpha}} \quad (1)$$

where  $C(\chi)$  and  $C(0)$  are the activity concentration of the radionuclide (Bq kg<sup>-1</sup>) at depth  $\chi$  (cm) and at  $\chi$  equal to zero, respectively, and  $\alpha$  (cm) is the coefficient representing the characteristics of the distribution. According to Isaksson and Erlandsson (1998), the concept of mass depth (g cm<sup>-2</sup>) should be introduced because the parameter  $\alpha$  is determined with assuming a uniform density for the whole soil profile. Therefore, we also calculated for the mass depth in equation (1) and determined the distribution parameter as  $\beta$  (g cm<sup>-2</sup>) as follows:

$$C(\chi') = C(0) \cdot e^{-\frac{\chi'}{\alpha \cdot D}} = C(0) \cdot e^{-\frac{\chi'}{\beta}} \quad (2)$$

where  $C(\chi')$  and  $C(0)$  are the activity concentration of the radionuclide (Bq kg<sup>-1</sup>) at mass depth  $\chi'$  (g cm<sup>-2</sup>) and at  $\chi$  equal to zero, respectively, and  $D$  is bulk density (g cm<sup>-3</sup>). In this study,  $D$  was calculated from each soil weight per unit volume sampled using a scraper plate. The method of least squares was used to determine the parameters  $\alpha$  and  $\beta$ , not including the litter layer. The parameters  $\alpha$  and  $\beta$  correspond to the relaxation depth and relaxation mass depth, respectively, i.e. the necessary distance or weight which  $C(0)$  decreases to  $1/e$  ( $\approx 1/2.7$ ).

### 3. Results and discussion

#### 3.1 The characteristics of soil chemical and physical properties

Tables 3 and 4 summarize physical and chemical properties of surface soils (0-5 cm) under eight study sites. As previously mentioned, the soil classification of the three forest sites, the PF site, and every other site were Andosol, Brown Lowland soil, and Reformed soil, respectively. In particular, the physical properties of surface soils in grassland and agricultural field sites were strongly impacted by the properties of the mounded soils, although several decades have passed since massive land development. The three forest Andosols without land development showed low bulk density (0.30-0.33 g cm<sup>-3</sup>), high saturated hydraulic conductivity ( $1.03 \times 10^{-1}$ - $1.38 \times 10^1$  cm s<sup>-1</sup>), high percentages of macro pore (26.4-41.9%) and gaseous phase (57.0-63.2%), indicating very high water permeability. The other soils, including PF, showed high bulk density (0.71-1.39 g cm<sup>-3</sup>), low saturated hydraulic conductivity ( $1.50 \times 10^{-4}$ - $1.06 \times 10^{-2}$  cm s<sup>-1</sup>) and low percentages of macro pore (1.3-8.5%), suggesting that these soils were hard and dense. The percentages of micro pore were similar in all soils and ranged from 47.0 to 65.4%.

**Table 3** Physical properties of surface soils under eight different land uses.

Site	Three phases distribution (%)			Micro pore (%)	Macro pore (%)	Bulk density (g cm <sup>-3</sup> )	K <sub>30</sub> <sup>a</sup> (cm sec <sup>-1</sup> )	Particle size distribution (%) <sup>b</sup>			Field Texture
	Solid	Liquid	Gaseous					< 5 μm	5-74 μm	>74 μm	
Mixed forest (MF)	11.1	25.7	63.2	47.0	41.9	0.31	1.05×10 <sup>-1</sup>	12.3	63.0	24.7	SiL
Mature cedar (MC)	8.1	28.8	63.1	57.1	34.5	0.30	1.38×10 <sup>-1</sup>	13.8	79.6	6.7	LiC
Young cedar (YC)	10.0	33.0	57.0	63.6	26.4	0.33	1.03×10 <sup>-1</sup>	6.0	70.9	23.1	LiC
Pasture land (PL)	27.4	20.4	52.2	65.4	7.5	0.71	2.95×10 <sup>-3</sup>	15.1	62.0	22.9	CL
Meadow land (ML)	31.7	23.6	44.7	63.2	6.5	0.82	1.06×10 <sup>-2</sup>	8.9	56.2	34.9	LiC
Farm land (FL)	52.9	44.4	2.7	47.5	1.3	1.39	1.50×10 <sup>-4</sup>	12.3	71.5	16.2	CL
Tobacco field (TF)	37.0	28.7	34.4	54.6	8.5	0.98	2.55×10 <sup>-3</sup>	16.3	75.6	8.2	LiC
Paddy field (PF)	38.1	48.2	13.7	59.3	3.1	1.00	1.26×10 <sup>-3</sup>	18.6	71.8	9.6	L

a: Saturated hydraulic conductivity converted at 20°C.

b: Measured by a laser diffraction particle size analyzer using surface soils sampled by a scraper plate (0-0.5 cm) without decomposition of the soil organic matters.

Soil chemical properties, such as soil pH and TC, were affected by land use patterns. The concentrations of TC in forest soils (87.0-165.2 g kg<sup>-1</sup>) and grassland soils (106.1-115.5 g kg<sup>-1</sup>) were higher than that of agricultural soils (10.9-32.3 g kg<sup>-1</sup>). The CEC values ranged from 19.7 to 58.1 cmol<sub>c</sub> kg<sup>-1</sup>, and were closely related to TC contents. On the other hand, the RIP values covered a wide range (0.93-9.57 mol kg<sup>-1</sup>). Nakao et al. (2014) reported that lower RIP values were found in soil clays classified as amorphous (< 4 mol kg<sup>-1</sup>) than those in soil clays belonging to the micaceous, vermiculitic and chloritic groups (> 10 mol kg<sup>-1</sup>). According to Vandebroek et al. (2012), Andosols with rich amorphous minerals were clearly characterized by low RIP (0.09-1.6 mol kg<sup>-1</sup>), but in the present study, the three forest Andosols showed relatively high RIP values (1.35-3.47 mol kg<sup>-1</sup>). This indicated that these Andosols contained phyllosilicate such as micaceous minerals in varying degrees. The lowest RIP was found in the PF site with a coarser soil texture and the highest RIP was found in the ML site.

### 3.2 The variability of the <sup>137</sup>Cs inventory

All data about dry weight of soil samples, mass depth, <sup>137</sup>Cs concentration, and the inventory and distribution ratios of <sup>137</sup>Cs in each soil layers are summarized in the supplementary material. The total inventory of <sup>137</sup>Cs was calculated as the total amount of <sup>137</sup>Cs contained within the top 10 cm of soils including the litter layer, using the <sup>137</sup>Cs concentration at each layer and soil weight per unit area.

The total inventory of <sup>137</sup>Cs at the first survey ranged from 0.25 to 1.0 MBq m<sup>-2</sup>. The lowest was found in the PF site while the highest was found in the PL site. As shown in Fig. 1, these inventories showed a similar tendency to the total Cs deposition reported by MEXT (2011), hence the variability among the sites was due to the spatial difference of the radiocesium deposition. However, there was high variability within single sites throughout the survey period (Fig. 2), showing that the coefficient of variations (CV) of the <sup>137</sup>Cs inventory ranged from 13 to 38%. As shown in Fig. 2, <sup>137</sup>Cs neither increased nor decreased with time. Schimmack and Schultz (2006) investigated the small-scale spatial variability of the radiocesium inventory at a flat grassland site by sampling five plots within 10 m<sup>2</sup> and found that the inventory of <sup>137</sup>Cs varied because of the spatial variability of the deposited radiocesium with CV of about 10%. The freezing and expansion of soil water in surface soils were observed in the second survey at the MC, YC, and TF sites, hence these soil

sampling weights reduced almost by half compared to those in other surveys. Moreover, as will be discussed later, secondary deposition of intercepted <sup>137</sup>Cs by the coniferous canopy or grass root growth may contribute to an increased variability in total inventory. Meanwhile, the annual rate of runoff in <sup>137</sup>Cs caused by soil erosion were lower than 3.2% in some of these study sites (Yoshimura et al., this issue).

### 3.3 Vertical distribution and temporal changes of <sup>137</sup>Cs in soil profiles

The vertical distribution of the <sup>137</sup>Cs concentration under the three forest sites is described in Fig. 3. In all forest sites, <sup>137</sup>Cs concentrations exponentially decreased with depth. The <sup>137</sup>Cs inventories in the 0-5 cm depth including litter layers accounted for 88-100% of the total, comprising more than 40% in the litter layers. Such a high proportion of the <sup>137</sup>Cs inventory in litter layers is a common characteristic in forest sites (Koarashi et al., 2012). However, the temporal changes in the <sup>137</sup>Cs proportion in litter layers were different among the three forest sites observed. In the MF site, the <sup>137</sup>Cs proportions in litter layers drastically decreased from 91% at the first survey to 41% at the fourth survey (Fig. 2a). Those in the MC site, however, maintained at about 40% over 2 y (Fig. 2b). The decrease ratio of the YC site was slower than that of the MF site because the weight of sampled litter at the fourth survey (31.4 g) was significantly less than the previous surveys (92.1-229.0 g) (Fig. 2c). This was because a large part of the released <sup>137</sup>Cs immediately fell down to the litter layer in the MF site since the leaf development of broad leaf trees had not begun when the FDNPP accident occurred, while, most of the released

**Table 4** Chemical properties of surface soils (0 - 5 cm) under eight different land uses.

Site	pH(H <sub>2</sub> O)	TC (g kg <sup>-1</sup> )	CEC (cmol <sub>c</sub> kg <sup>-1</sup> )	RIP (mol kg <sup>-1</sup> )
Mixed forest (MF)	4.10	115.5	44.3	2.22
Mature cedar (MC)	5.22	165.2	58.1	1.35
Young cedar (YC)	4.67	87.0	48.9	3.47
Pasture land (PL)	5.62	106.1	44.8	4.25
Meadow land (ML)	4.25	115.5	36.4	9.57
Farm land (FL)	5.80	10.9	25.1	3.55
Tobacco field (TF)	6.03	32.3	32.5	3.67
Paddy field (PF)	5.59	15.8	19.7	0.93

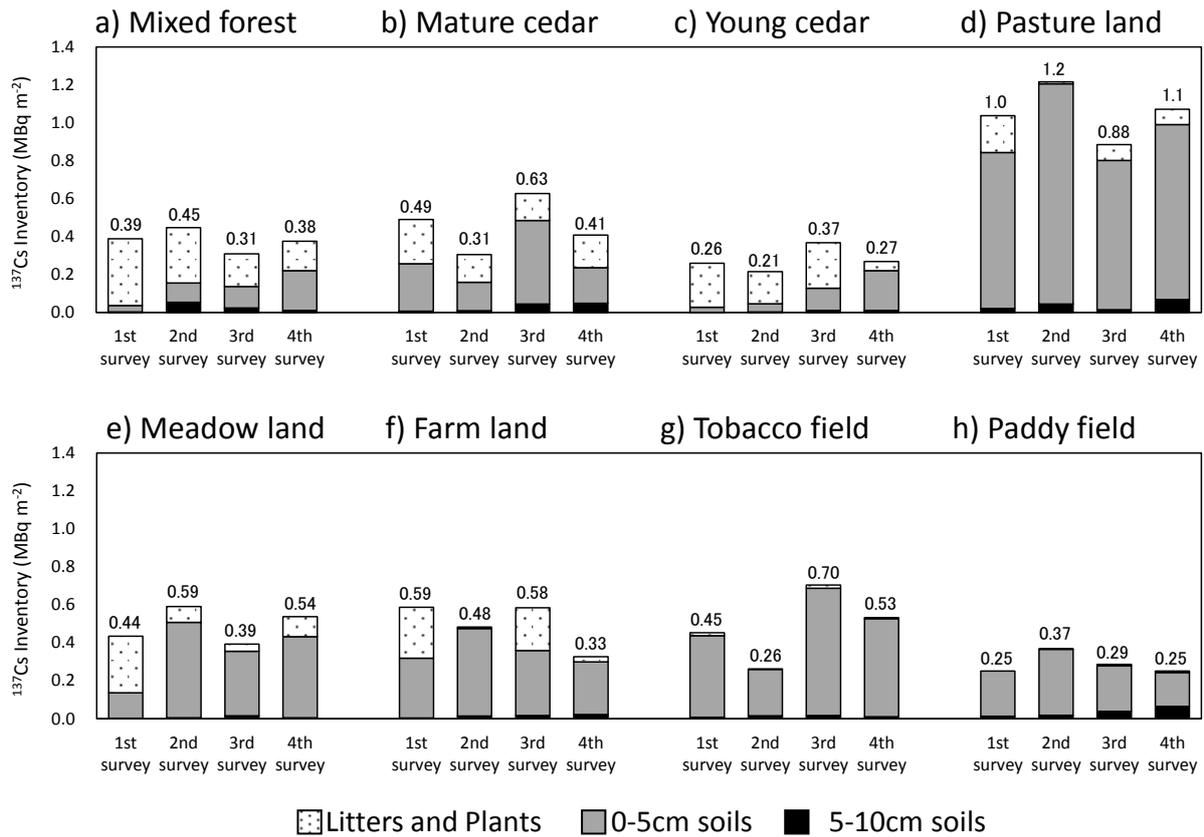


Fig. 2 Distributions of the <sup>137</sup>Cs inventories between litters and soils collected by four surveys in 2 y under different land uses.

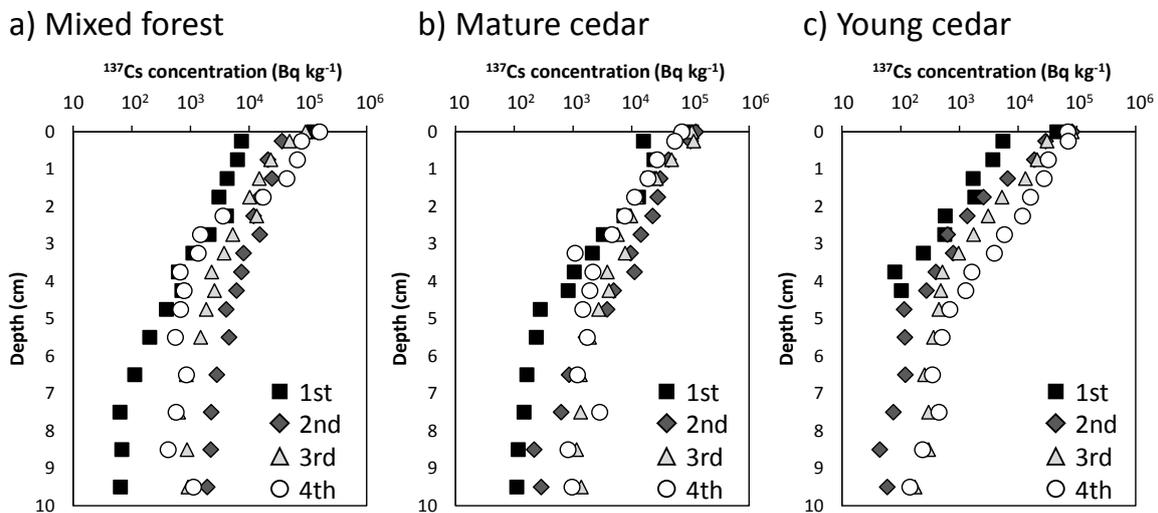


Fig. 3 Temporal changes in the vertical distributions of <sup>137</sup>Cs concentrations in the three forest sites.

<sup>137</sup>Cs was deposited on the cedar canopies in the MC and YC sites. In these three sites, Kato et al. (this issue) investigated the redistribution processes of the deposited <sup>137</sup>Cs on the canopies to the forest floor via throughfall, stemflow and litter-fall. They reported that the greatest deposition from the canopy was observed via throughfall in the MC site, while a relatively small deposition was found in the YC site, where young cedars with high closed canopy densely stand. Therefore, the <sup>137</sup>Cs inventory of the MC site was higher than that of the YC site although both sites are

nearby. In addition, it was considered that the secondary deposition from canopy by weathering and litter-fall sustained the high <sup>137</sup>Cs proportion in the litter layer in the MC site and increased the spatial variability of <sup>137</sup>Cs due to heterogeneous throughfall and litter-fall.

The vertical distribution of the <sup>137</sup>Cs concentration under two grassland sites is described in Fig. 4. Both grasslands were fully covered by dried perennial grass when the FDNPP accident occurred. In the ML site, the concentration of <sup>137</sup>Cs exponentially

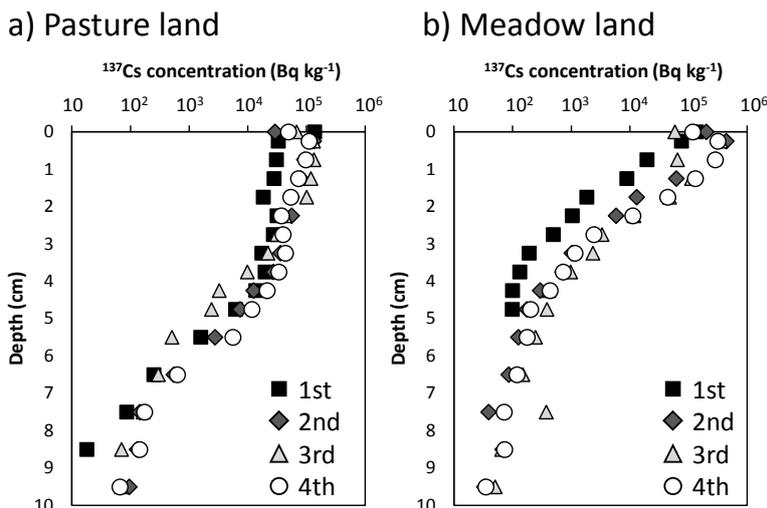


Fig. 4 Temporal changes in the vertical distributions of <sup>137</sup>Cs concentrations in the two grassland sites.

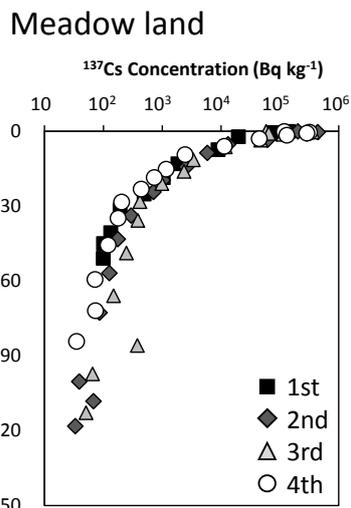


Fig. 5 Temporal changes in the mass depth distributions of <sup>137</sup>Cs concentrations in the ML site.

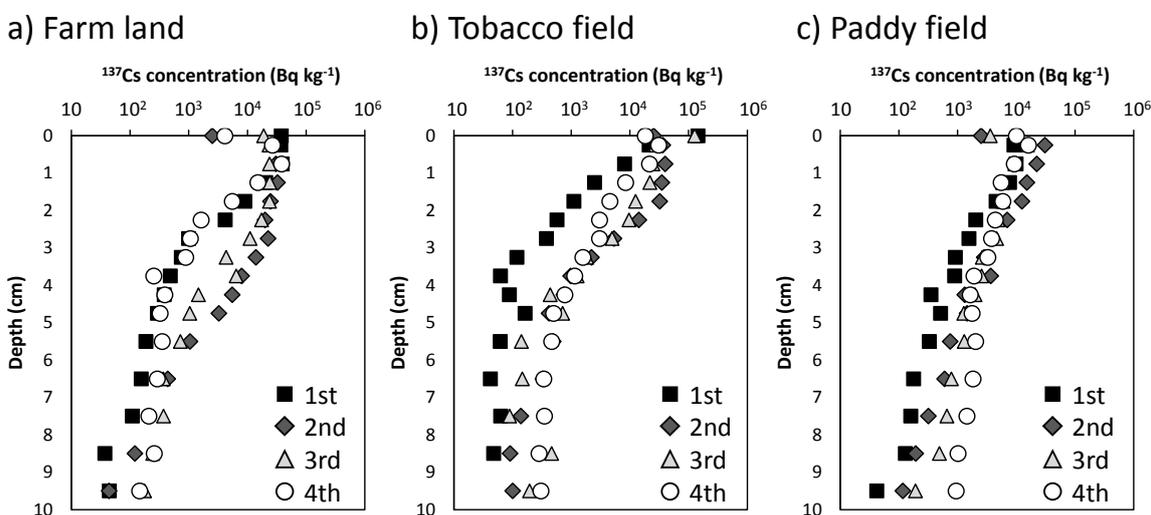


Fig. 6 Temporal changes in the vertical distributions of <sup>137</sup>Cs concentrations in the three abandoned agricultural field sites.

decreased and was not detected under 5 cm in depth at the first survey (Fig. 4b). The proportion of the <sup>137</sup>Cs inventory in the litter layer, including living grass, comprised 69% of the total, which was higher than the two cedar sites. However, that of the second survey drastically decreased to 14% (Fig. 2e). This indicates that the <sup>137</sup>Cs transfer to the soil surface occurred more rapidly here than the forest sites because there was no secondary deposition and the decomposition rate of herbaceous plant litter is commonly higher than that of deciduous tree litter (Haibara et al., 1983). Kirita et al. (2002) investigated the change of the weight of pasture litter over a fifteen-month period, from March to June, and showed that a 20% reduction was found by June, rapid decomposition (60% reduction) was observed from July to October, and then the rest slowly decomposed (70% reduction by June of the following year). This result approximately corresponds to our findings, which indicate a decrease in the proportion of the <sup>137</sup>Cs inventory in the litter layer of the ML site. The concentration of <sup>137</sup>Cs in the litter layer is influenced by the mix rate of decomposing and living grass. On the other hand, in the PL site, the <sup>137</sup>Cs from the surface

to 5 cm showed a similar concentration since the first survey (Fig. 4a). In addition, the proportion of <sup>137</sup>Cs inventory in the litter layer was 18.7% at the first survey and much less than that in the ML site (Fig. 2d). The reason for this is believed to be due to surface disturbance by cattle, such as the mixing of surface soils and the breakup of litters, because cattle had been grazing for a while after the FDNPP accident.

The unique feature of grassland soil profiles is the formation of root mat. In these sites, remarkable root mat growth was observed because cattle grazing or meadow gathering had been discontinued. For example, the weights gradually decreased in 0-3 cm soil samples from the ML site, indicating that surface soil particles with high <sup>137</sup>Cs concentration would be slid down by the roots (Fig. 4b). Therefore, the vertical distribution of the <sup>137</sup>Cs concentration against mass depth (kg m<sup>-2</sup>) showed little change over time (Fig. 5). These results indicate that <sup>137</sup>Cs in the ML site have been strongly adsorbed by soil particles and rarely migrate downward as soluble form.

The vertical distribution of the <sup>137</sup>Cs concentration under the

Table 5 Temporal changes in the vertical distribution parameters  $\alpha$  and  $\beta$ .

Site	$\alpha$ (cm)				$\beta$ (g cm <sup>-2</sup> )			
	1st	2nd	3rd	4th	1st	2nd	3rd	4th
Mixed forest (MF)	1.69	2.71	1.64	1.29	0.60	0.87	0.58	0.41
Mature cedar (MC)	1.40	1.43	1.43	1.75	1.02	0.22	0.73	0.95
Young cedar (YC)	1.02	0.84	1.19	1.12	0.46	0.30	0.78	0.44
Pasture land (PL)	4.02	1.92	1.42	2.30	3.08	1.91	0.58	1.16
Meadow land (ML)	0.54	0.56	0.71	0.77	0.60	0.38	0.53	0.30
Farm land (FL)	0.76	1.42	1.44	1.28	1.10	1.23	1.70	1.49
Tobacco field (TF)	0.95	2.45	1.96	1.12	1.06	0.95	2.08	1.00
Paddy field (PF)	1.46	1.69	2.18	2.89	2.02	1.33	2.26	2.60

three agricultural sites is described in Fig. 6. Immediately following the FDNPP accident, there was no vegetation, and little or no litter in the TF and PF sites, and only a few withered crop residue in the FL site. Therefore, it was presumed that most of the released <sup>137</sup>Cs deposited directly onto the soil surface. Although several weeds rapidly invaded these abandoned agricultural fields, the proportion of the <sup>137</sup>Cs inventory in the litter layer, including living weeds, was less than 3% in the TF and PF sites throughout every survey period (Fig. 2g and 2h), indicating only a minute <sup>137</sup>Cs uptake by the weeds. That of the FL site accounted for 46% and 39% of the total <sup>137</sup>Cs inventory at the first and third surveys (Fig. 2f), respectively, because some withered crop residue was contained in the litter layers during only these two surveys. This meant that the decomposition of the withered plant residue might be slower than fallen litter, and leaching by precipitation is small for the deposited <sup>137</sup>Cs on plant residue. Matsunaga et al. (2013) reported that the vertical distributions of <sup>137</sup>Cs, including the litter layer, were almost unchanged by rainfall events during the rainy season. Passeck et al. (1995) reported that water extractable <sup>137</sup>Cs in organic horizons of Histosol and Podzol were 1-7% of the total <sup>137</sup>Cs and the mobility of <sup>137</sup>Cs tended to increase as the decay of litter was progressed because dissolve organic substances might carry a considerable amount of <sup>137</sup>Cs. Thus, it is considered that litter decomposition and the dissolved organic substances are necessary to <sup>137</sup>Cs transfer, rather than the precipitation.

In the TF site, <sup>137</sup>Cs concentration in 0-3 cm soil was homogenized as with the second survey, slightly changing the vertical distributions of <sup>137</sup>Cs (Fig. 6b). This downward migration of <sup>137</sup>Cs is considered to be caused by the repeated formation and melting of needle ice in the surface soil during winter. Bare and cultivated agricultural lands, such as the TF site, are subject to the formation of needle ice. In the FL site, the same tendency of homogenized <sup>137</sup>Cs concentration in surface soil was observed, except in the fourth survey (Fig. 6a), likely indicating a partial formation of needle ice. As shown in Fig. 6c, the PF soil showed the smallest gradient of <sup>137</sup>Cs concentration and a pronounced increase in <sup>137</sup>Cs concentration over time under 5 cm. Moreover, the <sup>137</sup>Cs inventory in 5-10 cm soils gradually increased and comprised 26% of the total at the fourth survey (Fig. 2h). Although the downward migration of <sup>137</sup>Cs obviously occurred in the PF soil profile, the mechanism of this migration is not clear. According to Shiozawa (2013), the convective velocity of radiocesium in Fukushima paddy soil was two or three orders greater than the expected velocity which was calculated from laboratory determined distribution coefficients during the first 3-4 months

after the accident. However, the vertical distribution of the radiocesium concentration changed slightly during the next 8-9 months due to strong fixation by soil. On the other hand, Eguchi et al. (2010) reported that mobile soil particles with high radioactivities (i.e. surface soil particles) discharged after a rainfall event from a tile drain installed at a depth of 65 cm, indicating that soil particles moved quickly through some flow paths such as the large cracks which are frequently observed in smectitic heavy clay soil. However, the large cracks were not observed in this PF soil. Thus, further study is required to determine the concentration of <sup>137</sup>Cs under 10 cm depth and to clarify the mechanism and form of transport of the downward migration.

### 3.4 The relationships between the vertical distribution parameters and some soil properties

As previously mentioned, the migration patterns of <sup>137</sup>Cs from litter layer into soil surface or from surface to subsurface soil were highly influenced by land use patterns. Specifically, the existence of coniferous canopy caused the secondary contamination of <sup>137</sup>Cs, litter type affected the holding time of <sup>137</sup>Cs above ground, and the homogenization of <sup>137</sup>Cs concentrations in surface soil resulted from natural soil mixing such as the disturbance by cattle grazing, roots growing and the formation of needle ice. These functions contributed to the redistribution processes of deposited <sup>137</sup>Cs, but would not have no influence on the initial distribution of <sup>137</sup>Cs after its arrival in soil. Consequently, the vertical distribution of the first survey can be regarded as the initial distribution of deposited <sup>137</sup>Cs. Thus, we analyzed the effect of some soil properties on the initial distribution using the vertical distribution parameters  $\alpha$  and  $\beta$ , whose values are listed in Table 5.

The estimated concentrations of <sup>137</sup>Cs, found by substituting  $\alpha$  or  $\beta$  into eq (1) or (2), were relatively well fitted to the measured concentrations except the PL site which was influenced by cattle grazing. The determination coefficients calculated from the first survey tended to be higher ( $R^2=0.70-0.96$ ) than those from other survey periods ( $R^2=0.58-0.98$ ). It indicated that some distribution such as the PF site have been changing from the exponential distribution due to the effects of the downward migration or the secondary contamination. However, a weak relationship was obtained between  $\alpha$  and  $\beta$  ( $n=32, R^2=0.49, p<0.001$ ), because the three forest Andosols showed low values for  $\beta$  relative to  $\alpha$  due to low bulk density compared to other soils.

Table 6 summarizes the correlation coefficients obtained between some soil properties and the parameters  $\alpha$  and  $\beta$ , calculated from the first survey, excepting the PL site. As a result,

**Table 6** Correlation coefficients (*r*) obtained between the vertical distribution parameters and some soil properties, excluding the PL site (n=7).

	$\alpha$	$\beta$	pH	TC	CEC	RIP	Micro pore	Macro pore	Bulk density	K <sub>25</sub>	Particle of size < 5 $\mu$ m
$\alpha$	1										
$\beta$	0.279	1									
pH	0.017	-0.296	1								
TC	0.232	-0.081	-0.537	1							
CEC	0.251	-0.647	-0.255	0.894**	1						
RIP	-0.806*	-0.507	-0.422	0.178	-0.031	1					
Micro pore	-0.314	-0.044	-0.280	0.222	0.173	0.381	1				
Macro pore	0.651	-0.511	-0.260	0.762*	0.845*	-0.331	-0.159	1			
Bulk density	-0.528	0.515	0.366	-0.830*	0.881**	0.167	-0.241	-0.918**	1		
K <sub>25</sub>	0.552	-0.471	-0.290	0.801*	0.922**	-0.338	0.0021	0.943**	-0.919**	1	
Particle of size < 5 $\mu$ m	0.426	0.856*	0.554	-0.416	-0.492	-0.537	-0.322	-0.284	0.390	-0.365	1

\* and \*\*, Significant at the 0.05 and 0.01 probability levels, respectively.

a significant negative correlation was obtained between  $\alpha$  and RIP ( $r = -0.806, p < 0.05$ ), indicating that the penetration distance of <sup>137</sup>Cs in the very early stages decreased with an increasing RIP of the topsoil (0-0.5 cm). In addition, there was a positive correlation with macro pore ( $r = 0.651, p = 0.11$ ), suggesting that the penetration process of <sup>137</sup>Cs likely relates to gravitational water. These results also indicated that <sup>137</sup>Cs in less mobile water retained

by micro pore or soil structures could be gradually adsorbed or fixed with time. The parameter  $\beta$ , however, significantly correlated with the rate of fine particle (particle size < 5  $\mu$ m) content ( $r = 0.856, p < 0.05$ ). These fine particles were measured without the decomposition treatment of organic matter, so represent the amount of dispersible particles rather than clay minerals in a natural state. Therefore, a higher  $\beta$  was observed in

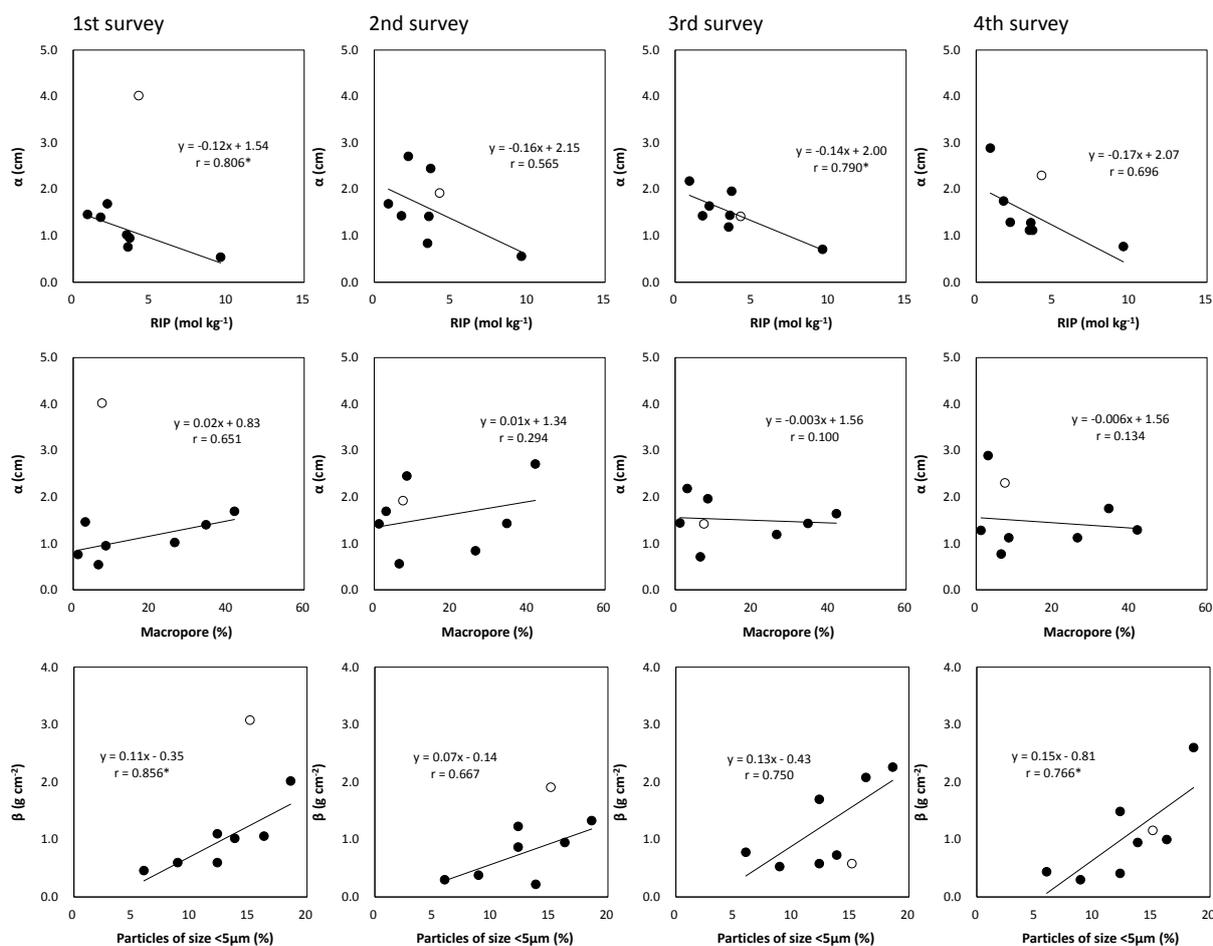


Fig. 7 Temporal changes in the relationships between some soil properties and migration parameters  $\alpha$  and  $\beta$ . The PL site (open circle) which was influenced by cattle grazing was excluded from the regression analyses. \* and \*\* are significant at the 0.05 and 0.01 probability levels, respectively.

soils containing high amounts of dispersible particles, such as the paddy field. These results indicated that the penetration of <sup>137</sup>Cs would be impacted by not only water flow but also spontaneous particle dispersion.

Only in the PF site the parameters  $\alpha$  and  $\beta$ , excepting the second survey, obviously increase over time (Table 5), showing the downward migration of <sup>137</sup>Cs. There were few changes, or a decreasing trend of these parameters in other sites, indicating that the amount of downward migrations was slight or lower than the amount supplied from litter.

Finally, we analyzed the temporal change in three soil properties as they relate to  $\alpha$  or  $\beta$  of the first survey. As shown in Fig. 7, the relationships with RIP and the ratio of fine dispersible particles were found throughout every survey period. However, there was no correlation with macropore after the second survey. These results indicated that the effects of RIP and fine particles, i.e. the fixation of <sup>137</sup>Cs by clay minerals and the dispersion of soil particles, continued for at least 2 y, whereas the effect of macropore, i.e. the penetration of <sup>137</sup>Cs by gravitational water, was slight after the initial deposition. This was because the <sup>137</sup>Cs on and in soils took on a mostly insoluble form after the second survey. In fact, although much of the released <sup>137</sup>Cs in the aftermath of the FDNPP accident existed in liquid form, such as in rainfall, throughfall and stemflow, the <sup>137</sup>Cs became less soluble after the second survey season, resulting in litter-fall becoming a major transfer pathway (Kato et al., 2012b; this issue). Some column experiments showed that the depth of initial penetration of <sup>137</sup>Cs was influenced by the intensity of the rainfall during the wet deposition (Schimmack et al., 1994), and the sequential downward migration increased with temperature because of microbial organic matter decomposition (Tegen et al., 1991). Further study is required to demonstrate these mechanisms of both initial and sequential migration of <sup>137</sup>Cs at field scale.

#### 4. Conclusions

In this study, we investigated the vertical distribution of <sup>137</sup>Cs in soil profiles using a scraper plate for 2 y after the FDNPP accident, focusing on the difference in land use patterns. As a result, characteristic patterns of the <sup>137</sup>Cs migration were observed at each land uses as follows.

Forest sites: the migration of <sup>137</sup>Cs from litter layer to soil surface was demonstrated in the mixed forest and young cedar sites, resulting in the proportion of <sup>137</sup>Cs in litter layer to the total inventory decreased from 90-91% to 18-41% for the 2 y. However, little change in the proportion in litter layer and an increase in the spatial variability of <sup>137</sup>Cs were observed in the mature cedar site due to the secondary deposition by the coniferous canopy.

Grassland sites: the migration of <sup>137</sup>Cs from litter layer to soil surface was faster than that of the forest sites, and has been almost completed. Although the <sup>137</sup>Cs concentrations in surface soil were homogenized by natural soil mixing such as the disturbance by cattle grazing and roots growing, it was considered that <sup>137</sup>Cs were strongly adsorbed by soil particles and rarely migrated downward as soluble form.

Agricultural field sites: the proportion of <sup>137</sup>Cs inventory in the litter layer, including living weeds, was less than 3.4% in tobacco field and paddy field sites, indicating little effect of <sup>137</sup>Cs uptake by invaded weeds. Only in the paddy field, the downward

migration of <sup>137</sup>Cs to subsurface soil clearly occurred, resulting in a gradual increase in the <sup>137</sup>Cs inventory in 5-10 cm soils, and comprising 25.5 % of the total <sup>137</sup>Cs in 2 y.

In addition, vertical distributions during the first survey were able to be used as the initial distributions and were well fitted to the exponential equation. The distribution parameters  $\alpha$  and  $\beta$ , calculated by the exponential equation tended to be correlated with RIP, macro pore, and dispersible fine particle content, indicating that the initial distribution would be influenced by the Cs fixation ability of soil, and the penetration process of water and particles in soils.

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