Vertical Migration of ^{137}Cs in Japanese Orchards after the Fukushima Daiichi Nuclear Power Plant Accident

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We investigated the vertical ^{137}Cs distribution in soil among five sod culture orchards with different soil textures over six years after 2011, when ^{137}Cs fallout was released by the Fukushima Daiichi Nuclear Power Plant accident, to confirm the long-term ^{137}Cs downward-migration into soil. At each orchard, soil cores were collected to a depth of 30 cm and subdivided into intervals of 3 to 9 cm. The ^{137}Cs within the 3 cm of topsoil decreased from 84–94% during the first 7 months after deposition in 2011 to 41–75% in 2017. From 2012 onward, the vertical ^{137}Cs profiles in the soils were explained by a two-component negative exponential model composed of a rapid and a slow component with a change of slope at a depth of 6 to 9 cm. It took 4 years after deposition to show a significant difference in the value of the average ^{137}Cs migration distance (M_d) among the orchards. The speed of ^{137}Cs migration in the orchards during the 6 years after the accident year was 0.44 to 0.97 cm year⁻¹ based on the M_d . There was a significant positive correlation between M_d and fine sand content in the 3 cm of topsoil between M_d and the ratio of the total carbon content (TC) at a depth of 3 to 6 cm to that in the top 3 cm of soil. Furthermore, the percentage of exchangeable ^{137}Cs (ex- ^{137}Cs) to ^{137}Cs at 3 to 6 cm depth increased significantly in proportion to the ratio of TC at 3 to 6 cm depth to TC at 0 to 3 cm depth in soil collected in 2013. These findings indicate that one of the mobile forms of ^{137}Cs was ex- ^{137}Cs combined with TC and that the fine sand content and TC influenced the ^{137}Cs downward-migration in the 3 cm of topsoil in the orchard in which organic matter accumulated by sod culture.

Key Words: average ^{137}Cs migration distance, exchangeable ^{137}Cs activity concentration, Fukushima Daiichi Nuclear Power Plant accident, long-term ^{137}Cs downward-migration, orchard.

Introduction

On 11 March 2011, the Tohoku earthquake caused a huge tsunami that resulted in a partial core meltdown in units 1, 2, and 3 of the Fukushima Daiichi Nuclear Power Plant (FDNPP), followed by a series of hydrogen explosions in units 1, 3, and 4. Large amounts of radionuclides were released into the environment from 12 to 14 March 2011 after the FDNPP accident (International Atomic Energy Agency, 2015). Radioactive deposition was caused primarily by rainfall and

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snowfall on 15 March 2011 (Chino et al., 2011; Imanaka, 2012). This fallout contaminated all fruit production orchards in Fukushima Prefecture, one of the main areas of deciduous fruit production in Japan (Ministry of Agriculture, Forestry and Fisheries, 2017).

Since sod culture is conducted as a soil management method in orchards, they are characterized by being covered with plentiful grass. At least until autumn 2011, more than 90% of the radiocesium (RCs) in the soil in orchards remained in the top 3 cm (Sato, 2014), where there is an abundance of grass roots that grow under the fruit trees in most orchards in Fukushima Prefecture. Since roots of fruit trees grow below 3 cm, there was concern that if RCs migrated further down in the soil profile, they may eventually enter the rooting zone of the trees, resulting in enhancement of their RCs uptake from the soil. Studies after the Chernobyl accident revealed that the rate of downward RCs migration in soil varies with soil type (Arapis et al., 1997; Rosén et al., 1999; Schimmack et al., 1989, 1997; Shand et al., 2013), texture and physicochemical properties (Forsberg and Strandmark, 2001; International Atomic Energy Agency, 2010; Niesiobędzka, 2000; Strebl et al., 1996; Zygmunt et al., 1998), and with rainfall (Schimmack et al., 1994; Shiozawa, 2013). In recent years, both mineral clay (Cremers et al., 1988; Tsukada et al., 2008) and organic matter (Niesiobędzka, 2000; Shand et al., 2013) have been identified as factors that affect the accumulation and migration of RCs in soil. Interestingly, Shiraishi (1973) pointed out that the effective half-life of the temporal ^{137}Cs concentration (^{137}Cs) in the fruits of Satsuma orange trees growing in soils contaminated by global fallout very closely agreed with that of the exchangeable ^{137}Cs concentration ($[\text{ex-}^{137}\text{Cs}]$) in the soil.

In the first 2 or 3 years after the FDNPP accident there were numerous short-term studies of the vertical soil profiles of RCs in forest, grassland, and agricultural fields (Fujii et al., 2014; Kato et al., 2012; Koarashi et al., 2012a, b; Kusaba et al., 2015; Matsunaga et al., 2013; Nakanishi et al., 2014; Ogura et al., 2014; Ohno et al., 2012; Shiozawa, 2013; Takahashi et al., 2015; Tanaka et al., 2012; Teramaga et al., 2014). However, of these previous studies, only three examined orchard soils (Koarashi et al., 2012a, b; Kusaba et al., 2015; Matsunaga et al., 2013), and only Kusaba et al. (2015) mentioned orchards, whereas Koarashi et al. (2012a) and Matsunaga et al. (2013) categorized orchards as croplands along with herbaceous crops such as rice (paddies), onion, and buckwheat in their study (Koarashi et al., 2012a). Therefore, there is little information about RCs profiles in orchard soils after both the accident at Chernobyl (Carini and Lombi, 1997) and the FDNPP accident. Furthermore, to our knowledge, there has been no long-term study of RCs profiles in orchard soils in the years following the FDNPP accident.

Prediction of long-term changes in the vertical distri-

bution of RCs is important for assessing the risk of RCs transfer into fruit and for developing decontamination programs focused on enhancing RCs uptake from orchard soils. Of all the RCs, ^{137}Cs is the most important contributor to soil contamination because it was one of the dominant radionuclides released following the FDNPP accident and has a long radiological half-life (30.1 years). ^{137}Cs was therefore selected as the target for our examination. A model of the migration of ^{137}Cs downward in soil is needed to predict the vertical distribution of RCs. Several models based on a one-component negative exponential function (Strebl et al., 1996; Velasco et al., 1997), a soil–soil solution distribution coefficient (International Atomic Energy Agency, 2010; Unno et al., 2017), or other factors (Mamikhin, 1995) have been proposed. Most models in studies after the FDNPP accident applied the one-component negative exponential function. However, information on the ^{137}Cs vertical profile model for orchard soil is lacking. Meanwhile, Shiraishi and Okabayashi (1967) found that vertical distributions of ^{137}Cs in topsoil down to a depth of 30 cm in a Satsuma orange orchard contaminated by global fallout showed two-component negative exponential functions, which consist of two “mobility factors”, a large one in the 2-cm topsoil and a small one below 2 cm. To our knowledge, no study has validated the model of vertical ^{137}Cs migration using two-component negative exponential functions in either orchards or in other terrestrial land. Furthermore, Shiraishi and Okabayashi (1967) pointed out that 20 to 30 cm of sampling depth was needed to investigate the vertical ^{137}Cs distribution in soil in Satsuma orange orchards. As mentioned above, there are no studies on the long-term ^{137}Cs downward-migration into soil by examining to a depth of 30 cm at the same site after the FDNPP accident.

The aim of this study was to (i) confirm the temporal changes in ^{137}Cs among five orchards with different soil textures in the 6 years after the FDNPP accident; (ii) to examine the validity of fitting the vertical distributions of ^{137}Cs in orchard soil to a two-component negative exponential decay function model; and (iii) to identify the factors that affect the vertical migration of ^{137}Cs in orchards.

Materials and Methods

Study sites

Five orchards approximately 60 to 65 km northwest of the FDNPP were selected for investigation. Three of them belonged to the Fruit Tree Research Center (FTRC), Fukushima Agricultural Technology Center in the city of Fukushima: a peach orchard [*Prunus persica* (L.) Batsch] ‘Akatsuki’ (AO), an apple orchard (*Malus pumila* Mill.) ‘Kougyoku’ (‘Jonathan’) (KO), and a ‘Mishima Fuji’ apple orchard (MO). The soil at MO was imported from the Yamakiya area, where soil is a granite type, approximately 40 years before the FDNPP

accident to investigate fertilization. The other two were a ‘Hatsuhime’ peach orchard (HO) in the town of Kohri and a ‘Fuji’ apple orchard (FO) in the city of Fukushima. According to the results of an airborne monitoring survey conducted by the Ministry of Education, Culture, Sports, Science and Technology (2011), on 26 May 2011 the deposition densities or inventories of ^{137}Cs were 100 to 300 $\text{kBq}\cdot\text{m}^{-2}$ at FTRC, < 100 $\text{kBq}\cdot\text{m}^{-2}$ at HO, and 300 to 600 $\text{kBq}\cdot\text{m}^{-2}$ at FO. The observation area in each orchard was approximately 500 to 1500 m^2 . The soil at HO was classified as a brown lowland soil; those in the other orchards were classified as brown forest soils (Obara et al., 2011). Sod culture was practiced in every orchard we studied. The topsoil was not cultivated and was left undisturbed at all of the survey sites. The main grass species were *Poa pratensis* L. (Kentucky bluegrass, KB) and *Trifolium repens* L. (white clover, WC) at AO, KO, MO, and FO, and *Capsella bursa-pastoris* (L.) Medik (shepherd’s-purse) and *Digitaria ciliaris* (Retz.) Koeler at HO. Total precipitation in the period from April to November in each year from 2011 to 2017 in FTRC was 969 mm, 722.5 mm, 909 mm, 1011.5 mm, 809 mm, 799 mm and 971 mm, respectively. The precipitation intensity reached 87 mm per day particularly on 10 September 2015, which is recorded as “Heisei 27 September Kanto-Tohoku heavy rain”.

Soil sampling to examine temporal changes in the depth profile of ^{137}Cs

Topsoil down to a depth of 30 cm was collected from each orchard with a stainless-steel core sampler (83 mm inside diameter), which was made in-house by the Research Center for Electron Photon Science at Tohoku University. Khomutinin et al. (2004) recommended

sampling areas of > 50 cm^2 to avoid any dependence on the variability of the ^{137}Cs inventory on the sampling scale. The core sampler that we used had a cross-sectional area of 54 cm^2 and thus satisfied this criterion. To prevent contamination, the inner surface of the core sampler was wiped with a paper towel moistened with deionized water before each sampling. Table 1 shows the sampling days and locations; note that replicates were not taken at each sampling in 2011. The above-ground parts of the grass at the sampling location were removed with pruning scissors before sampling. Each core sample was subdivided into 6 to 10 portions of 3 to 9 cm thickness by using a kitchen knife and a semicircular plate with the same diameter as the core sampler on one side to examine the vertical distribution of ^{137}Cs . Each sample was mixed well after the removal of rocks over 2-mm in diameter and then placed in a plastic counting vessel (U-8 vessel; 5 cm in diameter and 5 cm high, AS ONE Corporation, Japan). In the first year, the soil samples were not dried before measurements were made; this protocol was therefore continued in subsequent years.

Soil sampling to examine in high resolution the vertical profile of ^{137}Cs in 2016

The vertical profile of ^{137}Cs in high resolution in soil was examined to confirm the depth of the highest [^{137}Cs], and the relationship between subdivision intervals and the shape of the distribution of [^{137}Cs]. On 19 December 2016, 15-cm topsoil cores were collected from three locations at sites AO and MO by using the core sampler described in the previous section. The cores were wrapped and stored in a freezer (MDF-293AT; Sanyo, Japan) at -85°C . Each core sample was subdivided into 12 portions 0.5 to 3 cm thick by

Table 1. Sampling dates at each survey site to examine temporal changes in the depth profile of ^{137}Cs .

Survey site ^z	Latitude Longitude	Deposition density ^y ($\text{kBq}\cdot\text{m}^{-2}$)	Sampling date and number of replicates							
			Year							
			2011	2012	2013	2014	2015	2016	2017	
‘Hatsuhime’ orchard (HO)	37°51’03” N 140°33’34” E	< 100	11 May	26 Oct.	24 May	10 July	15 July	29 July	20 July	20 July
			1	1	2	3	3	3	4	3
‘Fuji’ orchard (FO)	37°44’26” N 140°28’54” E	300 to 600	11 May	26 Oct.	25 Apr.	30 Apr.	15 July	12 Aug.	22 July	13 Sept.
			1	1	2	3	3	3	3	4
‘Kougyoku’ orchard (KO)	37°48’49” N 140°26’21” E	100 to 300	15 Apr.	25 Oct.	24 May	22 May	16 July	17 July	13 July	13 Sept.
			1	1	2	2	3	3	3	3
‘Akatsuki’ orchard (AO)	37°48’48” N 140°26’38” E	100 to 300	26 Apr.	25 Oct.	25 Dec.	1 July	16 July	27 July	13 July	13 Sept.
			1	1	1	3	3	3	3	3
‘Mishima Fuji’ orchard (MO)	37°48’50” N 140°26’37” E	100 to 300	26 Apr.	25 Oct.	24 May	22 May	16 July	17 July	13 July	13 July
			1	1	3	3	3	3	2	2

^z The five orchards were located approximately 60 to 65 km northwest of the Fukushima Daiichi Nuclear Power Plant. Sites KO, MO and AO belonged to the Fruit Tree Research Center of the Fukushima Agricultural Technology Center in Fukushima City in Fukushima City. Sites FO and HO were commercial orchards in Fukushima City and the town of Kohri, respectively.

^y According to the results of an airborne monitoring survey conducted by the Ministry of Education, Culture, Sports, Science, and Technology (2011), on 26 May 2011.

using a pruning saw. Each subsample was then immediately freeze-dried and placed in a U-8 vessel for measurement of [^{137}Cs].

Texture and chemical properties of soils

After measurement of [^{137}Cs], samples collected in 2013 were air-dried, passed through a 2-mm stainless-steel sieve and then assayed for soil texture (particle size distribution), cation exchange capacity (CEC), and total carbon content (TC). Soil texture in each 3-cm layer to a depth of 30 cm was determined by the sieving method (sand content) and by the pipette method (silt and clay contents). The particle size distributions of all the samples from every core were classified as coarse sand (2 to 0.2 mm), fine sand (0.2 to 0.02 mm), silt (0.02 to 0.002 mm), and clay (< 0.002 mm). Soil texture was classified in accordance with the International Society of Soil Science classification scheme (United States Department of Agriculture, 2014). TC in each 3-cm layer to a depth of 6 cm was determined by the dry combustion method using an NC (nitrogen–carbon) analyzer (Sumigraph NC-220F; Sumika Chemical Analysis Service, Osaka, Japan). CEC at depths of 0 to 3 and 3 to 6 cm was estimated by using the $\text{CH}_3\text{COONH}_4$ saturation method at pH 7.0.

Exchangeable ^{137}Cs

The exchangeable ^{137}Cs activity concentration [$\text{ex-}^{137}\text{Cs}$] at depths of 0 to 3 and 3 to 6 cm in samples collected in 2013 was determined by using the following procedure. (i) Weigh out 5 g of air-dried soil and add 25 mL of 1 mol·L $^{-1}$ $\text{CH}_3\text{COONH}_4$ solution. (ii) Shake for 30 min on a reciprocating shaker. (iii) Filter the supernatant into a 100-mL volumetric flask after centrifugation. (iv) Add 25 mL of 1 mol·L $^{-1}$ $\text{CH}_3\text{COONH}_4$ solution to the residue and conduct the same procedure from (i) to (iii) 4 times. (v) Top up the volume of each extract to 100 mL by adding 1 mol·L $^{-1}$ $\text{CH}_3\text{COONH}_4$ to the 100-mL volumetric flask. Place the aliquot of 70 to 80 mL of each extract into a U-8 container to determine [$\text{ex-}^{137}\text{Cs}$] in the solution by using a germanium (Ge) semiconductor detector. [$\text{ex-}^{137}\text{Cs}$] (Bq·kg $^{-1}$ DW) in each soil layer was calculated by using the following equations:

$$[\text{ex-}^{137}\text{Cs}] = 1.014 \times (100 + f_w) f_d f_m C / 100 \quad (1)$$

$$f_m = 100/5 \text{ (mL} \cdot \text{g}^{-1}) \quad (2)$$

where f_d represents the decay coefficient (1.05) corrected on the basis of the sampling date, f_m is the dilution ratio of the 5-g sample, 1.014 is the specific gravity of the solution extracted with 1 mol·L $^{-1}$ $\text{CH}_3\text{COONH}_4$, C is a measured activity concentration (Bq·kg $^{-1}$), and f_w is the moisture coefficient calculated from the water content. The soil water content was determined by oven-drying each air-dried sample at 105°C for 24 h or more.

Radiocesium measurements

[^{137}Cs] in the 30-cm topsoil core samples was analyzed by using the Ge semiconductor detector systems at the RCEPS at Tohoku University in 2011 and 2012; at the Foundation for the Promotion of Material Science and Technology of Japan in 2013 and 2014; at the Graduate School of Agricultural and Life Sciences, The University of Tokyo, in 2015 and 2016; and at the Foundation for Promotion of Material Science and Technology of Japan in 2017. [$\text{ex-}^{137}\text{Cs}$] was measured with a Ge semiconductor detector at the Graduate School of Symbolic Systems Science and Technology Fukushima University. The gamma-ray peak used in the measurements was 662 keV for ^{137}Cs . The counting time for each sample ranged from 1 to 24 h, depending on the activity level. Activities were decay-corrected to the sampling date. The [^{137}Cs] in soil was converted to a dry weight (DW) basis using the water content after oven-drying of the residue from each sample at 105°C for at least 24 h.

Data analysis

In 2015, the bulk densities of dry soil (hereafter, bulk density, g·cm $^{-3}$) in each core sample were estimated to determine the ratio of the DW (g) of each layer to the volume (cm 3) of a cylinder with a height of 3 cm and a diameter of 8.3 cm, representing one intercept of the core sample. The vertical distribution of the ^{137}Cs inventory from 2011 to 2014 within the top 30 cm of soil was expressed for a distribution percentage (DP) which was calculated as a function of depth with the following equations:

$$\text{DP} = 100 \times f_i w_i C_i T_c^{-1} \quad (3)$$

$$w_i = h_i \cdot h_{\min}^{-1} \quad (4)$$

$$T_c = \sum f_i w_i C_i \quad (5)$$

where the subscript i represents the sample interval number, f_i is the soil density coefficient, w_i is a weighting coefficient based on the thickness of the soil interval, h_i is the thickness of each interval, $h_{\min}$ is the thickness of the smallest interval, and C_i is the [^{137}Cs] of the i th soil interval. The value of f_i in the top 3 cm of soil was equated to the ratio of the bulk density in the top 3 cm of soil to the bulk density below 3 cm (Table 2); the bulk density at depths of 3 to 30 cm was assumed to be 1.0 because no differences in this parameter were found within each of the five orchards at depths below 3 cm (data not shown). After 2015, DP was calculated by the measured value of bulk density instead of f_i .

The average ^{137}Cs migration distance from 2011 to 2014 and that from 2015 to 2017 were calculated by altering the formula of Shiozawa (2013) as follows:

$$M_d = \sum s_i f_i C_i \Delta s_i (\sum f_i C_i \Delta s_i)^{-1} \quad (6)$$

and

$$M_d = \sum s_i q_i C_i \Delta s_i (\sum q_i C_i \Delta s_i)^{-1} \quad (7)$$

Table 2. Bulk densities and soil density coefficients (top 3 cm of soil as a ratio of the soil at 3 to 30 cm depth) in 2015 for each survey orchard.

Survey site	Bulk density ($\text{g}\cdot\text{cm}^{-3}$)						Soil density coefficient (f_i ; a/b)		
	Depth (cm)								
	0–3 (a)			3–30 (b)					
	Mean	SD ^z		Mean	SD		Mean	SD	
HO	0.81	0.18	ab ^y	1.34	0.17	d	0.71	0.08	h
FO	0.84	0.03	ab	1.34	0.14	d	0.64	0.00	h
KO	0.66	0.06	a	1.47	0.13	e	0.47	0.02	g
AO	1.03	0.12	b	1.11	0.18	c	0.98	0.01	i
MO	0.96	0.11	b	1.61	0.19	f	0.65	0.03	h

^z SD represents standard deviation.^y Different letters indicate significant differences among the mean value of each survey site at $P \leq 0.05$ by Tukey's test.

where the subscript i represents the sample interval number, M_d is the average ^{137}Cs migration distance, f_i is the soil density coefficient, C_i is the measured ^{137}Cs at depth s_i , Δs_i is the measured depth interval, and q_i is the bulk density at i th layer.

Vertical ^{137}Cs profiles collected during the period from 2011 to 2017 were fitted to a one- or two-component negative exponential model by using KaleidaGraph 4.5J (HULINKS Inc., Tokyo, Japan), as follows:

$$C = K_1 \exp(-d/\alpha_1) + K_2 \exp(-d/\alpha_2) \quad (8)$$

where C is the measured ^{137}Cs at depth d , K_i is the maximum of each component, and α_i is the relaxation depth (Takahashi et al., 2015) of the i th component. The relaxation depth is the depth at which K_i has decreased to e^{-1} of its value at the surface. The data used for fitting were equated to the average values in each orchard in every year.

Akaike's information criterion (AIC: Akaike, 1973) was used to select the better component-negative exponential model of vertical ^{137}Cs profiles. The AIC—the smaller the value, the better the fit—is a popular tool for comparing the adequacy of models.

The data from 2013 were used to conduct a factor analysis based on the normal varimax criterion to identify factors associated with the downward-migration of ^{137}Cs . The data used in the factor analysis included measurements made at depths of both 0 to 3 and 3 to 6 cm in 2013. The value of M_d was calculated from ^{137}Cs at a depth of 0 to 21 cm as an index of the downward-migration of ^{137}Cs . Because the sampling interval at 21 to 30 cm was three times those at 0 to 3, 3 to 6, and 6 to 9 cm, the values measured at depths of 21 to 30 cm were excluded to avoid bias in the calculation of M_d . Regression analysis was applied to the measured data that were found to be associated with M_d on the basis of the factor analysis. STATISTICA 3.0J (StatSoft Japan, Tokyo, Japan) was used for the statistical analysis.

Results

Vertical distribution of ^{137}Cs in soil

Table 2 shows the bulk densities and the f_i of the top 3 cm of soil for each survey orchard. An analysis of variance (ANOVA) revealed no significant differences among the bulk densities of each depth interval below a depth of 3 cm within each of the five orchards (data not shown). The bulk density of the top 3 cm of soil was significantly lower than that at each depth interval below 3 cm ($P = 2 \times 10^{-5}$ by Tukey's test) within each orchard. The bulk density of the top 3 cm of soil at KO ($0.66 \text{ g}\cdot\text{cm}^{-3}$) was significantly lower than at MO ($0.96 \text{ g}\cdot\text{cm}^{-3}$) and AO ($1.03 \text{ g}\cdot\text{cm}^{-3}$) ($P = 0.048$ and 0.014 , respectively, by Tukey's test). The bulk density below 3 cm at AO ($1.11 \text{ g}\cdot\text{cm}^{-3}$) was significantly lower than at the other orchards. The minimum and maximum f_i coefficients were 0.47 (at KO) and 0.98 (at AO), respectively.

We plotted the vertical profiles of ^{137}Cs in samples collected during both spring (15 April, 26 April, or 11 May) and autumn (25 or 26 October) in 2011 (Table 1; Fig. 1). ^{137}Cs values were up to an order of magnitude higher in the top 3 cm of soil than in the next-deeper layer during both collection periods. ^{137}Cs in the layers at 21 cm and deeper were variable (Fig. 1A, B). The DP of ^{137}Cs in the top 3 cm of soil in each orchard ranged from 90% at HO to 97% at MO in spring, and from 69% at MO to 94% at AO in autumn. Only at MO was the DP of ^{137}Cs in the top 3 cm more than 20% lower in autumn than in spring. The ^{137}Cs within the top 3 cm of soil clearly decreased three years after the accident and DP of ^{137}Cs in the top 3 cm of soil ranged from 41% at AO to 75% at MO in 2017 (Table 3). Because the measured values of ^{137}Cs in the top 3 cm of soil collected in 2011 on 15 April, 26 April, 11 May, or 25 or 26 October diverged considerably from the values in the layer below, the one- or two-component negative exponential model could not explain the vertical ^{137}Cs profile in the soil.

We then examined the vertical profiles of ^{137}Cs obtained by fitting to the one- or two-component negative

exponential model during the period from 2012 to 2017 (Fig. 2; Table 4). The one-component negative exponential model gave smaller AICs than those with the two-component model in only two vertical profiles. The vertical profiles of ^{137}Cs during the period from 2012 to 2017 consisted of two decreasing components, one of which described rapid downward-migration and the other slow downward-migration. The one-component model gradually diverged from the measured values at depths greater than 6 cm. We plotted the relaxation

depths of the rapid and slow components (Fig. 3). The ranges of the relaxation depths of the rapid and slow components (α_1 and α_2) were approximately 1 to 4 cm and 4 to 10 cm, respectively. The rapid relaxation depths in the peach orchards tended to increase after 2015, whereas those of the apple orchards remained. There were no temporal trends in the slow relaxation depth among the orchards.

We plotted high-resolution vertical profiles of ^{137}Cs at AO and MO in 2016 (Fig. 4). There was a change in the slope of the trend of decreasing ^{137}Cs at a depth of approximately 3 cm. The maximum [^{137}Cs] between 0 and 15 cm occurred at a depth interval of 0.5 to 1 cm in all samples, with the exception of one high value in the depth interval of 0 to 0.5 cm at AO. Another change of slope occurred at approximately 6 to 9 cm depth—a pattern similar to that in Figure 2. Below a depth of 9 cm at AO, [^{137}Cs] was highly variable (i.e., dependent on the sampling location), in contrast to the pattern at MO.

Temporal changes in the downward-migration of ^{137}Cs in soil

We then examined the temporal changes in M_d (Fig. 5). M_d increased slowly until 2013 (777 to 848 days after the FDNPP accident) but began to accelerate after 2013. There were no significant differences among the orchards until 2014 (1219 days), 3 years after the FDNPP accident. After 2014 (1218 days), the M_d values of AO showed a higher trend compared with those of the other orchards. In 2015, (1585 to 1611 days) the M_d of AO was significantly higher than those of the other orchards except that of HO (ANOVA, $P = 0.017$). In 2017, (2319 to 2374 days) the M_d value at AO was 5.68 ± 1.11 cm (mean $\pm$ standard deviation, SD) and significantly higher than those of two apple orchards, which were 3.00 ± 1.60 cm (KO) and 2.64 ± 0.58 cm (MO). Although HO showed the next highest trend to AO after 2015, no significant differences to KO and

Fig. 1. Vertical profiles of ^{137}Cs in soil collected in (A) spring and (B) autumn in 2011. [^{137}Cs] is the ^{137}Cs activity concentration. A soil core was sampled at one location in each of the five orchards.

Table 3. ^{137}Cs concentrations and distribution percentages (DPs) in the top 3 cm of soil in each survey orchard from 2011 to 2017.

Survey year	Survey site and DP (%) ^z									
	HO		FO		KO		AO		MO	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
2011 (Spring) ^y	90	— ^x	91	—	97	—	93	—	97	—
2011 (Autumn)	88	—	84	—	91	—	94	—	69	—
2012	86	4.9	79	7.2	85	5.1	93	—	94	0.8
2013	75	6.5	86	5.3	76	14	83	7.7	86	7.0
2014	72	11	68	2.7	69	12	58	15	85	7.1
2015	54	10	86	7.6	81	12	55	7.7	78	6.9
2016	54	8.0	70	7.2	78	14	41	8.3	73	8.8
2017	42	7.0	60	16	74	25	41	18	75	14

^z DP calculated by soil density coefficient (f_i) until 2014 and by the measured value of bulk density instead of f_i after 2015.

^y DP of HO and FO were measured at the top 5 cm of soil only in the spring of 2011.

^x — represents no data.

Fig. 2. Vertical profiles of ^{137}Cs in soil collected during the period 2012 to 2017. ^{137}Cs is the ^{137}Cs activity concentration. Soil cores were collected at two to four locations in each survey orchard. Vertical bars represent standard deviations.

MO were found except for 2016 because of the large deviation in the valued data within the same orchard. Finally, the average M_d values from 2011 to 2017 at AO and HO were 3.83 ± 1.57 cm and 3.52 ± 0.93 cm, significantly higher than those of KO and MO, which were 2.70 ± 0.81 cm and 2.41 ± 0.57 cm by Tukey's test at $P \leq 0.05$.

Relationship among texture, chemical properties of soils, and vertical migration of ^{137}Cs and $[\text{ex-}^{137}\text{Cs}]$

Table 5 lists the soil textures at depths of 0 to 3 and 3 to 6 cm in the five orchards in 2013. The proportions of sand and clay varied among the five orchards from FO-type heavy clay (HC) to MO-type sandy loam (SL). The MO soil consisted of 56.8% to 66.6% coarse sand, but only 5.7% to 7.7% clay, whereas the FO soil consisted of 37.9% to 47.7% clay and 6.3% to 13.1% coarse sand. In contrast, fine sand was relatively abundant at HO, where it accounted for up to 43.2% of the soil. In 2013, we determined the values of ^{137}Cs , $[\text{ex-}^{137}\text{Cs}]$, chemical properties, and soil texture in the top 0 to 3 and 3 to 6 cm of soil, and of M_d in the top 21 cm (Table 6). These data were selected for factor analysis. M_d was the variable used to quantify the vertical migration of ^{137}Cs in the soil. Table 7 shows the factor loadings that resulted from the factor analysis. Factor analysis was performed with 13 variables. Because the changes in the eigenvalues were 4.88, 3.70, 2.00, and 1.18, a three-factor structure was considered reasonable. The cumulative percentage of the variance accounted for by these three factors exceeded 80%. The first factor consisted of five variables: M_d , the ratio of TC at 3 to 6 cm depth to TC in the top 3 cm of soil (TC ratio), the CEC ratio,

the fine sand content in the top 3 cm of soil, and ^{137}Cs in the top 3 cm of soil. The second factor consisted of four variables: the clay content of the top 3 cm of soil, the coarse sand content of the top 3 cm of soil, the CEC in the top 3 cm of soil, and $[\text{ex-}^{137}\text{Cs}]$ in the top 3 cm of soil. The third factor consisted of four variables: ^{137}Cs at a depth of 3 to 6 cm, $[\text{ex-}^{137}\text{Cs}]$ at a depth of 3 to 6 cm, TC in the top 3 cm of soil, and TC at a depth of 3 to 6 cm. The first factor indicated that there was a close relationship between M_d and the TC ratio, or between M_d and the CEC ratio, or between M_d and the fine sand content in the top 3 cm of soil. In fact, there was a significant positive correlation between M_d and the TC ratio and between M_d and the fine sand content of the top 3 cm of soil (Fig. 6). Furthermore, there was a significant positive correlation between the percentage of $[\text{ex-}^{137}\text{Cs}]$ to ^{137}Cs at 3 to 6 cm depth and the TC ratio (Fig. 7).

Discussion

Temporal changes in vertical ^{137}Cs migration in orchard soil

More than 84% of the total ^{137}Cs down to a depth of 30 cm was localized in the top 3 cm of soil, much greater than the proportion of ^{137}Cs in the next layer, at least 7 months after the FDNPP accident. The greater decrease at MO by autumn 2011 compared with spring was probably due to the fact that the core was collected at a location disturbed by field management, judging by the temporal trend in ^{137}Cs distribution at MO during the following sampling year (Fig. 2). Koarashi et al. (2012a) mentioned an exponential decline in the values measured in cores collected at 15 locations in south-

Table 4. Coefficients (Coe.) of a one- or two-component negative exponential model of vertical ^{137}Cs profiles in orchards (Orc.), together with Akaike's information criterion values.

Orc.	Coe.	Days elapsed since 15 March 2011											
		436 (2012)		848 (2013)		1218 (2014)		1597 (2015)		1954 (2016)		2319 (2017)	
		One	Two	One	Two	One	Two	One	Two	One	Two	One	Two
HO	K_1	37.0	135	13.2	15.3	16.3	18.2	9.4	9.3	3.6	2.8	3.6	2.8
	α_1	1.1	0.5	1.6	1.3	1.6	1.3	2.5	1.9	4.6	4.6	4.6	4.6
	K_2		1.3		0.8		0.5		1.2		0.8		0.8
	α_2		4.9		6.0		7.8		6.2		4.6		4.6
	AIC	-5.9	-33.0	-11.8	-28.7	-11.5	-28.8	10.4	-19.0	10.4	-19.0	23.1	-8.4
Orc.	Coe.	Days elapsed since 15 March 2011											
		407 (2012)		777 (2013)		1218 (2014)		1611 (2015)		1956 (2016)		2374 (2017)	
		One	Two	One	Two	One	Two	One	Two	One	Two	One	Two
FO	K_1	46.2	135	93.0	99.9	62.6	95.9	62.8	203.7	7.0	37.1	31.3	32.6
	α_1	1.2	0.6	1.2	1.1	2.2	0.1	0.9	0.5	1.8	0.5	1.9	1.7
	K_2		2.8		0.5		62.6		0.8		1.0		0.8
	α_2		3.9		9.5		2.2		5.8		6.3		8.8
	AIC	-2.9	-25.5	-5.9	-27.1	14.7	14.7	-8.2	-33.8	-6.4	-29.3	-4.0	-15.3
Orc.	Coe.	Days elapsed since 15 March 2011											
		436 (2012)		799 (2013)		1219 (2014)		1585 (2015)		1947 (2016)		2374 (2017)	
		One	Two	One	Two	One	Two	One	Two	One	Two	One	Two
KO	K_1	67.1	81.1	46.7	50.0	37.3	42.9	22.2	24.5	29.6	40.5	25.4	30.8
	α_1	1.0	0.9	1.4	1.3	1.4	1.2	1.2	1.1	1.4	1.0	1.3	1.1
	K_2		0.4		0.6		1.2		0.1		2.0		0.9
	α_2		7.1		7.7		5.3		10.0		4.3		5.0
	AIC	-13.1	-22.1	-7.5	-21.9	0.6	-2.1	-20.1	-29.7	-6.5	-23.6	-12.1	-27.7
Orc.	Coe.	Days elapsed since 15 March 2011											
		651 (2012)		839 (2013)		1219 (2014)		1595 (2015)		1947 (2016)		2374 (2017)	
		One	Two	One	Two	One	Two	One	Two	One	Two	One	Two
AO	K_1	26.2	149	38.8	38.3	5.8	5.0	10.1	10.5	7.4	5.9	12.9	12.8
	α_1	1.2	0.5	1.8	1.8	4.9	4.8	3.0	2.1	3.7	3.7	3.2	2.8
	K_2		1.4		0.7		0.8		1.2		1.5		0.7
	α_2		5.0		5.6		5.0		11.1		3.7		15.0
	AIC	-2.9	-20.8	-9.9	-6.3	-5.0	-4.9	15.4	-18.0	5.2	-8.4	1.2	-1.2
Orc.	Coe.	Days elapsed since 15 March 2011											
		436 (2012)		799 (2013)		1219 (2014)		1585 (2015)		1947 (2016)		2374 (2017)	
		One	Two	One	Two	One	Two	One	Two	One	Two	One	Two
MO	K_1	45.8	56.0	59.6	67.9	24.3	75.3	20.7	70.7	15.4	16.4	15.1	16.3
	α_1	0.9	0.8	1.1	1.0	1.1	0.6	1.2	0.6	1.7	1.5	1.5	1.3
	K_2		0.2		0.8		0.8		1.0		0.3		0.5
	α_2		5.3		4.7		4.6		5.1		10.2		5.3
	AIC	-30.2	-56.7	-13.3	-49.6	-13.0	-36.4	-9.3	-30.9	-13.2	-24.7	-20.0	-36.8

western Fukushima city, approximately 70 km north-west of the FDNPP, from 18 to 20 June 2011. They measured [^{137}Cs] in the top 5 cm of soil by subdividing the core at intervals of 1 to 2 cm. Since the vertical ^{137}Cs profile in two orchards in their study were explained by one-negative exponential models, the non-conformity in fitting the negative exponential model in our study may have been due to the choice of subdivi-

sion intervals for measurement of [^{137}Cs]. From another viewpoint, this result indicated that ^{137}Cs downward-migration below 3 cm was negligible until 7 months after the fallout from the FDNPP accident.

Takahashi et al. (2015) investigated the vertical ^{137}Cs profiles at eight survey sites in the Yamakiya area of Fukushima Prefecture in 2011 and 2012 by fitting a one-component negative exponential function. How-

Fig. 3. Temporal changes in relaxation depth of the rapid and the slow components from 2012 to 2017. α_1 and α_2 are the relaxation depths calculated by using the following two-component negative exponential model: $C = K_1 \exp(-d/\alpha_1) + K_2 \exp(-d/\alpha_2)$, where C is the measured ^{137}Cs at a depth of d , K_i is the maximum of each component, and α_i is the relaxation depth of the i th component. The relaxation depth is the depth at which K_i has decreased to e^{-1} of its value at the surface.

ever, a logarithmic plot of the vertical distribution of ^{137}Cs in the top 0 to 10 cm of soil at almost all locations revealed a change in the trend at about 3 cm depth. If the vertical profile of ^{137}Cs were to fit a negative exponential function, the logarithmic graph of the ^{137}Cs plot would be linear. However, the graphs of vertical profiles of ^{137}Cs in the soil at the study sites of Takahashi et al. (2015) were not strictly linear. In particular, the vertical ^{137}Cs profile in forests showed the two trends turned at a depth of 3 to 6 cm in a clearly similar pattern to the high-resolution vertical profiles of ^{137}Cs in our study (Fig. 4). These results differed from the findings of Koarashi et al. (2012a), which explained the vertical ^{137}Cs distribution in forest soil by a one-component negative exponential function. Although no studies have applied a two-component negative exponential model to the investigation of forests, it was considered reasonable to describe the vertical ^{137}Cs distribution in orchard soil by using a two-component negative exponential model, at least for a long-term investigation. In our study, the trend in the rapid relaxation depth from 2012 to 2017 largely agreed with that of M_d (Figs. 3 and 5), implying that the rapid compo-

Fig. 4. High-resolution vertical profiles of ^{137}Cs in soil collected at (A) AO and (B) MO in 2016. ^{137}Cs is the ^{137}Cs activity concentration. Soil cores were sampled at three locations in each survey orchard.

Fig. 5. Temporal changes in average ^{137}Cs migration distance (M_d) after 15 March 2011 (the day ^{137}Cs was deposited). Vertical bars represent standard deviations. Different letters indicate a significant difference at $P \leq 0.05$ by Tukey's test.

nent directly controlled the speed of downward-migration. Figure 3 suggests that the slow component probably reflected the downward-migration of ^{137}Cs below 6 to 9 cm, where the upper rhizosphere of a fruit tree is found (data not shown). In other words, the slow component was indicative of the amount of ^{137}Cs potentially transferred to the fruit tree via the roots. In 2017, the initial ^{137}Cs of the slow component was estimated at 0.2 to 0.9 $\text{kBq} \cdot \text{kg}^{-1} \text{DW}$ (Table 4). Furthermore, the relaxation depth of the rapid component in AO and HO accelerated after 2014, and that in HO was more than

Table 5. Particle size distributions in soils collected in 2013 from the survey sites.

Survey site	Rep. no. ^z	Sampling depth and particle size distribution							
		0–3 (cm)				3–6 (cm)			
		Coarse sand ^y (%)	Fine sand (%)	Silt (%)	Clay (%)	Coarse sand (%)	Fine sand (%)	Silt (%)	Clay (%)
HO	1	47.6	12.6	22.3	17.5	37.8	32.0	17.1	13.1
HO	2	28.9	37.2	19.3	14.6	32.6	35.4	18.1	13.9
HO	3	15.5	41.7	25.0	17.7	13.5	43.2	26.0	17.3
FO	1	7.8	15.1	29.4	47.7	7.4	19.2	29.2	44.3
FO	2	7.0	18.4	28.1	46.5	6.3	18.4	29.7	45.7
FO	3	13.1	18.8	29.9	38.3	10.9	19.9	31.3	37.9
KO	1	27.4	31.7	20.6	20.3	27.5	32.7	21.4	18.4
KO	2	34.5	20.7	22.4	22.3	22.7	35.9	21.9	19.4
AO	1	31.1	27.6	21.7	19.7	33.5	26.6	21.3	18.6
AO	2	34.6	25.0	21.0	19.4	31.3	31.0	19.4	18.4
AO	3	28.8	29.6	20.9	20.7	29.6	30.6	20.5	19.3
MO	1	56.8	26.8	9.8	6.6	56.8	27.2	9.7	6.3
MO	2	59.5	21.8	11.0	7.7	66.6	18.5	9.2	5.7
MO	3	59.8	22.5	10.2	7.5	58.8	25.2	9.9	6.1

^z Rep. no. represents the replicate number.^y Coarse sand, 2 to 0.2 mm; fine sand, 0.2 to 0.02 mm; silt, 0.02 to 0.002 mm; clay, <0.002 mm.**Table 6.** Measured values for the top 0 to 3 cm or 3 to 6 cm of soil collected in 2013. Values were applied to a factor analysis.

Survey site	Rep. no. ^z	Measurement item and sampling depth											
		¹³⁷ Cs (kBq·kg ⁻¹)		[ex. ¹³⁷ Cs] (kBq·kg ⁻¹)		CEC	CEC ratio	TC	TC	TC ratio	Coarse sand	Fine sand	Clay
		0–3 cm	3–6 cm	0–3 cm	3–6 cm	0–3 cm (mol _c ·kg ⁻¹)	3–6 cm/ 0–3 cm	0–3 cm (g·kg ⁻¹)	3–6 cm (g·kg ⁻¹)	3–6 cm/ 0–3 cm	0–3 cm (g·kg ⁻¹)	0–3 cm (g·kg ⁻¹)	0–3 cm (g·kg ⁻¹)
HO	1	6.51	1.18	0.72	0.10	20.9	0.90	33.1	18.1	0.55	636	168	234
HO	2	3.79	0.35	0.37	0.02	21.8	0.97	27.4	18.4	0.67	289	372	146
HO	3	5.69	0.93	0.54	0.11	25.1	1.03	26.2	22.2	0.85	155	417	177
FO	1	44.8	2.90	3.45	0.15	41.4	0.76	73.7	15.4	0.21	78.1	151	477
FO	2	11.3	0.45	0.44	0.02	36.2	0.75	42.3	12.5	0.29	70.4	184	465
FO	3	22.5	2.81	1.59	0.19	32.3	0.80	51.9	19.7	0.38	131	188	383
KO	1	14.9	2.73	0.52	0.12	24.7	0.71	66.8	33.7	0.50	274	317	203
KO	2	17.1	0.94	0.17	0.02	30.6	0.58	119	31.9	0.27	345	207	223
AO	1	11.0	0.77	0.27	0.07	24.5	0.84	48.5	28.3	0.58	311	276	197
AO	2	23.1	6.22	0.99	0.43	28.6	0.77	75.5	41.9	0.55	346	250	194
AO	3	16.3	2.55	0.92	0.32	25.5	0.85	59.6	35.9	0.60	288	296	207
MO	1	5.86	0.58	0.11	0.02	11.1	0.87	22.6	5.4	0.24	568	268	66
MO	2	32.2	1.94	0.49	0.05	17.8	0.54	39.4	11.1	0.28	595	218	77
MO	3	8.18	0.50	0.15	0.02	13.1	0.70	31.1	4.5	0.14	598	225	76

^z Rep.no. represents the replicate number.

4 cm in 2016. The speed of the increase in the relaxation depth of the rapid component at HO (0.76 cm·y⁻¹) was more than 3 times higher than that at MO (0.22 cm·y⁻¹). From a long-term viewpoint, there is no doubt that a further survey of the rapid component is needed. Because the transfer factor to fruit (Bq·kg⁻¹ fresh weight) from soil (Bq·kg⁻¹ DW) is in the range of 10⁻⁴ to 10⁻² (Carini, 2001), continued monitoring of ¹³⁷Cs in the fruits of shallow-rooted fruit and nut trees may be necessary for a while, at least in areas more

heavily contaminated with ¹³⁷Cs than in our study area.

In our study, it took 4 years after the FDNPP accident to show a significant difference in the M_d values between AO and other orchards. No studies have examined the temporal changes in the M_d after fallout, so it is difficult to explain the reason for the above time-dependence. Considering the physicochemical characteristics of AO and HO, which showed higher M_d values, the precipitation as a factor to control seepage and the soil temperature as a factor to increase the dis-

Table 7. Factor analysis of ^{137}Cs migration in the top 0 to 3 cm or 3 to 6 cm of soil.

Variable ^z (Measurement item)	Depth (cm)	Factor loading			Commonality
		Factor 1	Factor 2	Factor 3	
[^{137}Cs]	0–3	–0.59	0.48	0.41	0.76
[^{137}Cs]	3–6	–0.08	0.18	0.89	0.82
[ex. ^{137}Cs]	0–3	–0.21	0.78	0.24	0.70
[ex. ^{137}Cs]	3–6	0.19	0.17	0.86	0.81
CEC	0–3	–0.12	0.92	0.28	0.93
CEC ratio	3–6/0–3	0.89	0.14	–0.21	0.86
TC	0–3	–0.43	0.25	0.63	0.64
TC	3–6	0.28	0.07	0.86	0.82
TC ratio	3–6/0–3	0.90	–0.01	0.26	0.87
Coarse sand content	0–3	–0.26	–0.86	–0.12	0.82
Fine sand content	0–3	0.82	–0.30	0.03	0.76
Clay content	0–3	–0.15	0.95	0.00	0.93
M_d	0–21	0.89	–0.03	0.20	0.84
Variance accounted for		3.87	3.56	3.13	
Cumulative contribution ratio (%)		29.8	57.1	81.2	

^z [^{137}Cs], total ^{137}Cs activity concentration; [ex- ^{137}Cs], exchangeable ^{137}Cs activity concentration; CEC, cation exchange capacity; TC, total carbon content; M_d , average ^{137}Cs migration distance. CEC ratio, ratio of CEC at 3 to 6 cm soil depth to that at 0 to 3 cm depth; TC ratio, ratio of TC at 3 to 6 cm depth to that at 0 to 3 cm depth.

Fig. 6. Regressions between average ^{137}Cs migration distance (M_d) and (A) fine sand content and (B) the TC ratio. The TC (total carbon) ratio is the ratio of TC at 3 to 6 cm depth to that at 0 to 3 cm depth. Soil samples were collected in 2013.

solved carbon (Tegen et al., 1991) are related. On the other hand, to assess the effectiveness of long-term measures against ^{137}Cs deposition in orchard soils it is

Fig. 7. Regression of the ratio of exchangeable ^{137}Cs activity concentration [ex- ^{137}Cs] to total ^{137}Cs activity concentration [^{137}Cs] at 3 to 6 cm soil depth against the total carbon (TC) ratio. The TC ratio is the ratio of TC at 3 to 6 cm depth to TC at 0 to 3 cm depth. Soil samples were collected in 2013.

important to evaluate the speed of ^{137}Cs migration. Arapis et al. (1997) used a quasi-diffusion model and the migration center of the vertical distribution of radionuclides to evaluate the speed of ^{137}Cs migration in three regions of Greece (Antonopoulos-Domis et al., 1991) 7 years after the Chernobyl accident. Their results indicated a speed of 0.71 to 1.54 $\text{cm}\cdot\text{year}^{-1}$ for the rate of ^{137}Cs migration. Rosén et al. (1999) showed that migration rates from 1987–1995 in Sweden were 0.5 to 1.0 $\text{cm}\cdot\text{year}^{-1}$ for the first year; thereafter, they were 0.2 to 0.6 $\text{cm}\cdot\text{year}^{-1}$ (based on median depths). We found here, on the basis of M_d , that the speed of ^{137}Cs migration in orchards in the 6 years after the FDNPP accident was 0.44 to 0.97 $\text{cm}\cdot\text{year}^{-1}$; this speed is consistent with the range of 8.2 to 17.4 cm for the mean migration

depth of ^{137}Cs in organic soil in Scotland 22 years after the Chernobyl accident (Shand et al., 2013). Each migration center, median depth and the mean migration depth was calculated as the centroid, which represents the center of the amount or activity concentration of ^{137}Cs using the same algorithm as M_d .

Factors affecting vertical migration of ^{137}Cs in orchard soils

Investigations at FTRC in 2014 revealed that the root weight in the top 3 cm of soil accounted for up to 73.8% of the total root weight at a depth of 0 to 15 cm (Sato et al., 2019). Dominance of grass roots was likely the reason why we found here that the bulk density at all sites was lower at 0 to 3 cm than at 3 to 30 cm. Fukushima Prefectural Government recommends the use of sod culture, a soil management method intended to enrich the nutritional status of orchard soil through the cultivation of herbaceous plants or grasses (Sato et al., 1978). In sod culture, the swards are mown every time they grow to more than approximately 20 cm high. It is commonly known that an aggregate structure, which is characterized by abundant pores and much organic C, develops in the topsoil of grassland soil (Kobayashi and Yamane, 1976). In fact, Matsunaga et al. (2013) verified that the organic carbon contained in the top 3 cm of soil in an orchard was more than twice the level found in herbaceous farmland. Sod culture retains this characteristic aggregate structure of orchard topsoil. KB is well known to develop a good root mat (Sugiura et al., 1988). WC does not develop a root mat, but it spreads large numbers of stolons (runners) from which fibrous roots are generated (Devkota et al., 1997). Weeds are generally inferior to KB and WC in terms of root development. During the 2014 investigations at FTRC (Sato et al., 2019), the weight of the roots in the top 3 cm of soil at KO, which was well covered with KB and WC, was $0.40 \text{ kg} \cdot \text{m}^{-2}$, whereas in the peach orchard near AO, which was invaded by weeds, the corresponding weight was $0.21 \text{ kg} \cdot \text{m}^{-2}$, indicating that the differences between the orchards in terms of bulk density in the top 3 cm of soil were associated with the presence of invading weeds. On the other hand, the ^{137}Cs amount in the roots in the top 3 cm of soil was $0.46 \text{ kBq} \cdot \text{m}^{-2}$ at KO and $0.37 \text{ kBq} \cdot \text{m}^{-2}$ in the peach orchard near AO, whereas the ^{137}Cs amount in the same layer of soil was $339 \text{ kBq} \cdot \text{m}^{-2}$ and $151 \text{ kBq} \cdot \text{m}^{-2}$. These findings indicate that the ^{137}Cs amount in the roots of grass is relatively small, so the influence of ^{137}Cs absorption by grass on ^{137}Cs kinetics in soil is likely small. Takahashi et al. (2015) showed that survey sites with high pore volume have relatively low bulk density. There was likely a large pore volume at AO, which had the lowest bulk density below 3 cm (Table 2). The difference in the bulk density due to the method of sod culture likely affects the activity of soil microorganisms in soil and the infiltration of precipitation, resulting in

differences in the supply of organic carbon and the percolation of seepage water.

Our regression of M_d against fine sand content was significant in the top 3 cm of soil. Downward-migration of ^{137}Cs into the soil may be due to the infiltration and percolation of precipitation (Shiozawa, 2013). Sato et al. (2019) found that more ^{137}Cs migrates below 3 cm in open spaces, where rainfall hits the ground directly, than at locations below the canopy. Tegen et al. (1991) confirmed that dissolved ^{137}Cs is transported by seepage water through the soil. The permeability coefficient of water is larger in fine sand than in coarse sand in the unsaturated state (Kaihotsu, 1979, 1984). It is likely that the surface soil layer is maintained in an unsaturated state because the topsoil lies directly below the air and develops an aggregate structure due to sod culture. The third factor in the factor analysis provided interesting information about the downward-migration of ^{137}Cs , the implication being that ex- ^{137}Cs migrated with TC. It is thus clear that organic matter plays an important role in the downward-migration of ^{137}Cs , at least in soil with abundant organic matter. Shand et al. (2013) concluded that “Clay alone did not have a prime role in controlling the migration of ^{137}Cs down the profile”, because in their investigation of organic soils in Scotland, they could not find a relationship between the type of clay minerals and the profile of downward-migration of ^{137}Cs . Moreover, they quoted the results of Schimmack et al. (1997) in Bavarian grassland, where there was “no correlation between the content of clay mineral in the soils ranging (between 11 and 41%)” and the profile of downward-migration of ^{137}Cs . Niesiobędzka (2000) revealed that of mobile forms represented as the total ^{137}Cs extracted by distilled water, NH_4COOH and HCl were positively correlated with organic carbon and organic matter in the top 10 cm of sandy soil. On the other hand, Koarashi et al. (2012a) found a significant correlation between ^{137}Cs retention and the ratio of organic C to clay in the top 3 cm of soil at 15 locations. According to Figure 7, one of the mobile forms of ^{137}Cs is the ex- ^{137}Cs combined with TC. Tegen et al. (1991) found that the ^{137}Cs in seepage water is bound to the dissolved organic carbon of which the supply in the surface depends on soil microorganisms. The significant positive correlation between the percentage of [ex- ^{137}Cs] to [^{137}Cs] at 3 to 6 cm depth and the TC ratio (Fig. 7) found in the present study is supported the finding by Tegen et al. (1991). Since the activity of soil microorganisms depends on the soil temperature (Tegen et al., 1991), continuous supply of organic matter by the sod culture and the vertical trend of soil temperature are the reasons why the vertical ^{137}Cs profile in orchard soil separates the two components. In our study, the highest [ex- ^{137}Cs] and TC content in the top 6 cm of soil occurred at AO (Table 6). Interestingly, AO had the lowest bulk density below 3-cm depth (Table 2), indicating that there was likely abundant pore

space below 3-cm depth. On the other hand, the bulk density below 3-cm depth at KO and MO, where the mean value of M_d over 6 years was significantly lower, was significantly higher than that of others so one reason for the difference of M_d in orchards is probably the bulk density. Moreover, retention factors, e.g. undecomposed organic matter and mica, rather than migration factors, may also be involved. Koarashi et al. (2012a) revealed that there was a significant positive correlation between the ^{137}Cs retention and the bulk density in the top 1 to 3 cm of soil, but few studies have examined on the relationship between pores and the ^{137}Cs downward-migration into soil. Further studies are needed to determine the factors controlling ^{137}Cs downward-migration into soil in orchards.

We conclude based on the first time evidence in orchards as follows: (i) the vertical ^{137}Cs profile in orchard soil consisted of two components related to the content of TC, which likely consists mainly of organic carbon, in the top 3 cm of soil supplied by sod culture; (ii) ^{137}Cs downward-migration into soil depended on the factors of the infiltration and percolation of seepage, e.g. fine sand content and pore space; (iii) exchangeable ^{137}Cs combining with TC as a carrier was one of the migration processes in orchards.

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