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Author(s)	Takahashi, Shunsuke; Suchara, Ivan; Sucharova, Julie; Okamoto, Kazumasa; Umegaki, Kikuo; Fujiyoshi, Ryoko
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Corresponding Author:	Ryoko Fujiyoshi, Ph.D Hokkaido University Sapporo, JAPAN
Corresponding Author Secondary Information:	
Corresponding Author's Institution:	Hokkaido University
Corresponding Author's Secondary Institution:	
First Author:	Shunsuke Takahashi, Master
First Author Secondary Information:	
Order of Authors:	Shunsuke Takahashi, Master
	Ivan Suchara
	Julie Sucharova
	Kazumasa Okamoto
	Kikuo Umegaki
	Ryoko Fujiyoshi
Order of Authors Secondary Information:	
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Abstract:	Abstract Purpose This study examined the current distribution and downward migration of 137Cs in the humus horizon under temperate coniferous stands in the Czech Republic. Depth distribution profiles of lithogenic alkali metals (K, Rb and Cs) were also traced to find any indication regarding the 137Cs dynamics within the humus horizon. Materials and methods Soil (bulk soil) samples were collected manually from the uppermost down to a depth of 20 cm at three points situated 10-20 m apart in three locations diversely affected by the Chernobyl accident in 1986. Humus samples (about 6 cm in thickness) were separately collected at three points adjacent to the bulk soil sampling. The humus samples were divided into three fractions (upper, middle and lower) denoted to Hu, Hm and Hl, respectively depending on their depths. Activity concentration of 137Cs and the amount of alkali metals (K, Rb and Cs) in each humus fraction were determined with gamma spectrometry and ICP spectrometry, respectively. Some properties (pH, density, SOM and identification of clay minerals), of both bulk soils and humus horizons were also investigated. Results and discussion The highest activity concentration of 137Cs (Bq kg-1) appeared in the upper portion of the soil (mainly humus horizon) under three coniferous stands about 30 years after the Chernobyl accident. Increasing activity of 137Cs (Bq) was found in the lower humus fraction (Hl, 4~6 cm depth range) with no appreciable amounts of clay minerals like illite and smectite in the investigated sites. The findings suggest that the fallout 137Cs moves downward at a speed of 0.13~0.19 cm yr-1 with degrading organic matter within the humus horizon. Possible association of 137Cs with alkali metals (K, Rb and Cs) was suggested by depth distribution profiles of lithogenic

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Downward migration of ^{137}Cs within the humus layer under temperate coniferous stands in the Czech Republic

Shunsuke Takahashi[1] • Ivan Suchara[2] • Julie Sucharová[2] • Kazumasa Okamoto[1] • Kikuo Umegaki[1] • Ryoko Fujiyoshi[1]

[1] Faculty of Engineering, Hokkaido University, Sapporo 060-8620, Japan

[2] Silva Tarouca Research Institute for Landscape and Ornamental Gardening, Květnové náměstí 391, CZ 252 43 Pruhonice, Czech Republic

✉ Ryoko Fujiyoshi

Tel: +81 11 706 6674

Fax: +81 11 706 6675

fuji@eng.hokudai.ac.jp

Abstract

Purpose This study examined the current distribution and downward migration of ^{137}Cs in the humus horizon under temperate coniferous stands in the Czech Republic. Depth distribution profiles of lithogenic alkali metals (K, Rb and Cs) were also traced to find any indication regarding the ^{137}Cs dynamics within the humus horizon.

Materials and methods Soil (bulk soil) samples were collected manually from the uppermost down to a depth of 20 cm at three points situated 10-20 m apart in three locations diversely affected by the Chernobyl accident in 1986. Humus samples (about 6 cm in thickness) were separately collected at three points adjacent to the bulk soil sampling. The humus samples were divided into three fractions (upper, middle and lower) denoted to H_u, H_m and H_l, respectively depending on their depths. Activity concentration of ^{137}Cs and the amount of alkali metals (K, Rb and Cs) in each humus fraction were determined with gamma spectrometry and ICP spectrometry, respectively. Some properties (pH, density, SOM and identification of clay minerals), of both bulk soils and humus horizons were also investigated.

Results and discussion The highest activity concentration of ^{137}Cs (Bq kg^{-1}) appeared in the upper portion of the soil (mainly humus horizon) under three coniferous stands about 30 years after the Chernobyl accident. Increasing activity of ^{137}Cs (Bq) was found in the lower humus fraction (H_l, 4~6 cm depth range) with no appreciable amounts of clay minerals like illite and smectite in the investigated sites. The findings suggest that the fallout ^{137}Cs moves downward at a speed of 0.13~0.19 cm yr^{-1} with degrading organic matter within the humus horizon. Possible association of ^{137}Cs with alkali metals (K, Rb and Cs) was suggested by depth distribution profiles of lithogenic alkali metals (K, Rb and Cs) in humus.

Conclusion The humus horizon under temperate coniferous stands plays an important role in retaining fallout ^{137}Cs for a long time. Comparing the depth distributions of the fallout radionuclide ^{137}Cs with the depth distributions of lithogenic alkali metals (K, Rb and Cs) gives valuable information for clarifying mechanism of ^{137}Cs movement in humus. Further investigation is needed to elucidate mechanism of ^{137}Cs migration within humus horizon by tracing ^{137}Cs speciation and decomposing soil organic matter simultaneously.

Keywords Cesium-137 • Forest soil • Humus • Vertical migration rate, Alkali metals

1 Introduction

Many highly-contaminated forest areas have been abandoned without any effective remediation in the thirty years since the Chernobyl accident (Beresford et al. 2016). Considerable amounts of ^{137}Cs still remain in the surface portion of forest soils (Winkelbauer et al. 2012; Szabó et al. 2012; Vahabi-Moghaddam and Khoshbinfar 2012; Petrovič et al. 2016). Transportation of deposited ^{137}Cs on the forest floor are affected by various environmental factors including surface runoff, microbial activity, root uptake and water movement in soil. Among the many studies that have focused on the dynamics of ^{137}Cs in forest ecosystems, clay mineralogy was found to be an important factor controlling the rather low mobility of ^{137}Cs caused by sorption and fixation by certain clay minerals (Sawhney 1972; Cornell 1993, Dołhańczuk-Śródka et al. 2006, Szabó et al. 2012). Recently, Fujii et al. (2016) investigated vertical migration of radiocesium (^{134}Cs and ^{137}Cs) deposited on several forest sites due to the accident of Fukushima Daiichi Nuclear Power Plant in Japan in 2011. They concluded that both organic components and clay minerals had enough capacities for retaining radiocesium in the surface soils during the initial stage of contamination.

In our previous paper on ^{137}Cs retention with soil components under selected forest sites in the Czech Republic (Takahashi et al. 2017), the highest proportion of ^{137}Cs in soil was found in the organic horizons (O_f and O_h) under temperate coniferous stands diversely affected by the atmospheric fallout from the Chernobyl accident. We obtained relatively low apparent burial rate of ^{137}Cs (0.04-0.08 cm yr^{-1}) in the upper soil layer (< 15 cm depth), which contained no significant amounts of clay minerals, suggesting that the ^{137}Cs may be associated with humic macromolecules in the upper soil layer down to a depth of about 15 cm. Humus has a much longer life span than any other organic materials among the various forest components. The "environmental memory" of an area can be traced back for at least the last 60-100 years (Sucharova et al. 2011). In a study of the interactions of radiocesium with humus in forest soils, Strebl et al. (1995) investigated distribution of ^{137}Cs in surface soils and found that the transfer rate of ^{137}Cs from forest soils to plants was much more strongly influenced by the quality of humus components rather than by their amounts. Kruse-Irmer and Gian (2003) reported that radiocesium would be involved in a shortcut element cycle in the system of humus layer-plant uptake-litter, considering the high bioavailability and low migration of radiocesium in the humus layers. High bioavailability and low migration rate of ^{137}Cs in humus obtained in their study suggested that ^{137}Cs may be rather strongly associated with humic components with appreciable amounts of ionic and/or exchangeable ^{137}Cs. Therefore, evaluating the distribution and mobility of ^{137}Cs in the humus layer in considerable detail is necessary for long-term fate of ^{137}Cs deposited in forest floors.

It is also important to consider the distribution of alkali metals (K, Rb and Cs) as well as ^{137}Cs in humus, because i) all belong to the alkali metal group with similar physicochemical properties, ii) stable alkali metals are from lithogenic (natural) source, whereas ^{137}Cs is originated from the atmosphere by accidental release (anthropogenic source), and iii) little is known about the distribution of the alkali metals as well as ^{137}Cs in forest humus. There are several papers describing analytical results of K, Rb and Cs in spruce needles (*Picea abies* Karst.) and in the associated soils in Switzerland (Wyttenbach et al. 1995). Mean concentrations of these elements in soils were found in the order K >> Rb > Cs, which were consistent with the average composition of the upper continental crust. They concluded that the change of the concentration with soil depth was completely explained by the dilution of the mineral soil with organic matter in near surface. Vinichuk et al. (2010) carried out a comparative study of accumulation of alkali metals (K, Rb, Cs) and ^{137}Cs in soil and fungi in a Swedish forest. They found a similar mechanism of Cs uptake by fungi to that of Rb, different with K uptake. These studies were aimed at understanding bioavailability of radionuclides to prevent ultimate exposure to living organisms.

The present study has especially focused on the current distribution and downward migration of ^{137}Cs in the humus horizon under the same stands as those previously investigated to elucidate site-specific and common properties of ^{137}Cs retention within the humus horizons. Depth distribution profiles of a lithogenic stable cesium (^{133}Cs) and other alkali metals (K and Rb) were also investigated to compare those of ^{137}Cs, and to find any relationships among them within the humus horizon 30 years after the Chernobyl accident.

2 Material and methods

2.1 Location, site description, and sample collection

The sites for investigation in this study were the same as those described in our previous paper (Takahashi et al. 2017), i.e. three temperate coniferous (*Picea abies*) stands with a mean stand age of 60-120 years in the Czech Republic. These sites were Přemyslov (50.101ºN; 17.078ºE), Opatovice (49.962ºN; 15.196ºE) and Prühonice (49.990ºN; 14.543ºE), as denoted sites I, II and III, respectively.

Soil (bulk soil) samples were manually collected every 2 cm from the uppermost down to a depth of 12 cm (6 subsamples), and one subsample each from 12 to 15 cm, and 15 to 20 cm in depth with a stainless frame (10 × 10 × 10 cm) at three points situated 10-20 m apart in each location in September 2014. Humus samples were separately collected at three points adjacent to bulk soil sampling. Mean thickness of the humus horizon was 6 cm, which was divided into three fractions of equal thickness (2 cm). The separated humus fractions were denoted by the upper (H_u) middle (H_m) and lower (H_l) depending on their depth.

2.2 Analytical procedures

All samples were air-dried and sieved with a 2-mm mesh filter in the laboratory. Soil pH (H_2O) was measured directly on each soil sample with a pH meter (HI-99121, Hanna Instruments, Ltd., Woonsocket, RI, USA). The amount of soil organic matter (SOM) was estimated by ignition weight loss (%) of each sample before and after heating at 500 ºC for 2 hrs. Powder X-ray diffraction analyses (RINT-2000m, Rigaku, Tokyo, Japan) of all the samples investigated in this study were carried out under identical experimental conditions (X-ray source; CuKα, applied voltage; 40 kV, maximum current; 30 mA, diffraction angle 2θ; 3-40º).

The activity concentration of ^{137}Cs in each soil component was determined by gamma spectrometry with a HPGe detector (GMX20P4-70; EG&G ORTEC, Oak Ridge, TN, USA). Acquisition time was set to be 144000 s for all measurements including a background check. Certified reference materials purchased from the International Atomic Energy Agency (IAEA-372) and from the Japan Society for Analytical Chemistry (JSAC0471, JSAC0472 and JSAC0473) to evaluate the ^{137}Cs activity concentration of individual samples from count data obtained with the same geometry under identical operating conditions. The lowest limit of detection was found to be 0.34 Bq kg^{-1} with a confidence level of 95 % for ^{137}Cs activity concentration under the experimental conditions in this study.

Inductively coupled plasma (ICP) analyses were carried out for all the samples to evaluate distribution of some alkali metals (K, Rb and Cs) in humus and in bulk soils. Both Rb and Cs were determined with ICP-mass spectrometry (ELAN 6000; Perkin Elmer Inc., Waltham, MA, USA), and K was analyzed with ICP-atomic emission spectrometry (OPTIMA 5000; Perkin Elmer Inc., Waltham, MA, USA). Details of the analytical procedures were described in our previous paper (Sucharová and Suchara 2006).

2.3 Amount of alkali metals (K, Rb and Cs) and activity of ^{137}Cs in humus fraction

Determination of the alkali metal in humus gives its concentration (C_X; mg kg^{-1}). The amount (X_{hum}; mg) of an alkali metal (K, Rb or Cs) contained in humus fraction of a definite thickness (d; cm) and dry density (ρ; g cm^{-3}) could be calculated from the equation (1).

$$X_{hum} = C_X \times \rho \times S \times d \times 10^{-3} \tag{1}$$

where S is the surface area of the humus fraction defined to be 100 cm^2 in this study.

Activity of ^{137}Cs (Bq) in humus fraction is also obtained from its activity concentration (Bq kg^{-1}) through the equation (1).

The amount of an alkali metal (K, Rb Cs or ^{137}Cs) in the mineral component (X_{min}) of a humus fraction could be estimated from the intercept of a linear relationship between SOM (%) and X_{hum} (mg, or Bq in the case of ^{137}Cs), assuming that the forest humus is exclusively composed of organic (SOM) and mineral (min) components.

$$X_{hum} = a \cdot [SOM] + X_{min} \tag{2}$$

Where X = K, Rb and Cs, and the subscripts of X, hum and min, denote humus and soil organic component, respectively. A symbol ɑ is the slope of the linear regression in the equation (2).

2.4 Vertical migration rate of ^{137}Cs

Vertical migration of ^{137}Cs in humus was investigated based on its depth distribution profiles obtained in this study at all three sites. Equation (3) was used to estimate a vertical migration rate of ^{137}Cs (Arapis and Karandinos 2004):

$$\bar{x} = \sum_i x_i g_i, \tag{3}$$

where $\bar{x}$ is the weighted mean depth (cm) of the vertical concentration distribution, x_i is the mean depth of the layer (cm), and g_i is the fraction of ^{137}Cs in the corresponding depth x_i.

The vertical migration rate of ^{137}Cs (R, cm yr^{-1}) was estimated from the mean depth values ($\bar{x}$, cm) divided by elapsed time (t, years) after the Chernobyl accident in 1986 ($R = {}^{\bar{x}}/_{t}$).

3 Results and discussion

3.1 Soil and site properties

Table 1

Table 1 summarizes site-specific properties of soil including those (location, climate, soil type, forest type, stand age, ^{137}Cs inventory, apparent annual burial rate to mineral horizon and ecological half-life of ^{137}Cs) described in our previous paper (Takahashi et al. 2017). Among three sites investigated, soil and forest types are similar in properties, while other environmental factors are different, especially climatic conditions and stand age. The humus horizons in three sites (I, II and III) were selected to be similar thickness (~6 cm), which was further divided into three parts with equal thickness (2 cm); upper (H_u), middle (H_m) and lower (H_l). Moisture and the amount of soil organic matter (SOM) within each humus fraction decreased gradually to lower portion. Mean pH value was relatively constant ($3.4 \leq pH \leq 3.7$) independent of the sites with an exception in the site I (pH 4.5 at H_u). All the soils were classified into acidic Cambisol suggesting that the amounts of dissolved chemical species are relatively large in soil water. Gonet et al. (2008) investigated compositional and chemical changes of humus with depth in undisturbed forest soils in Slovakia. They showed that down the profile gave higher hydrophilic property of humus with higher carboxyl groups. Higher concentration of hydrophilic humic acids at deeper portion of the soil may result from increasing amount of decomposed SOM. Paul et al. (2015) investigated acidic and metal complexing properties of humic acids extracted from tea garden soils of different depths (surface, 15 and 30 cm in depth) in India. They obtained different properties of the humic acids with depth, e.g. decreasing moisture content and increasing pKa values with depth. The pH value of each humus fraction was relatively low (pH; 3.4~4.5), which may result from pH buffer effects of hydrophilic humic acids with carboxylic and hydroxylic functional groups (Paul et al. 2015). Such low values are generally observed in humus layers of non-calcareous sandy soils summarized by De Vries and Leeters (2001) in forest stands in the Netherlands.

These factors may facilitate dissociation of $^{137}Cs^+$ with anionic humus components at low pH in the lower humus horizon in our investigated sites.

3.2 Depth distribution profiles of alkali metals (K, Rb and Cs) in bulk soil and in humus horizon

Uptake of nutrients from soil is an important process in biological cycling in forest environment, in which radiocesium may be indiscriminately taken up by plant roots together with other alkaline elements such as rubidium (Rb) and stable cesium (Cs) as well as an essential nutrient potassium (K).

Fig. 1

Figure 1 shows linear relationships between SOM and concentration of alkali metals (K, Rb and Cs) in bulk soil in the site III (Plühonice). The plots suggest that the concentrations of alkali metals increase linearly with increasing soil depth. The correlation coefficients between them were obtained for K ($R=0.998$, $n=8$, $p<0.01$), Rb ($R= 0.994$, $n=8$, $p<0.01$) and Cs ($R =0.622$, $n=8$, $p<0.01$). Assuming that the soil is simply composed of organic and mineral components, the results in Fig. 1 indicate that these alkali metals are of lithogenic origin. There is scare information available for comparing the amounts of alkali metals in humus. Among them Sucharova et al. (2011) conducted chemical analyses of 39 elements including alkali metals (K, Rb, Cs) in humus samples collected in coniferous forest sites across the Czech Republic. Median values in humus samples were reported as 3240, 19 and 2.2 mg Kg^{-1} for K, Rb and Cs, respectively. Using analytical data obtained on these elements, they investigated a trend analysis of the spatial distribution. They found that most anomalies of typical elements in the distribution could directly be linked to known industrial sites, mines, power plants or urbanization. Johanson et al. (2004) determined many chemical elements including alkali metals (K, Rb and Cs) in forest soils close to the Nuclear Power Plant at Forsmark in Sweden. They summarized concentrations (median values) of K, Rb and Cs in bulk soil to be 605 (370-986), 2.52 (1.48-10.1) and 0.21 (0.080-0.68) mg kg^{-1}, respectively. They also showed two to five times higher values of K and Rb concentrations in rhizosphere than those in bulk soil except for Cs which remained at low levels independent of soil compartments. Vicichuk et al. (2010) investigated the distribution of K, Rb and Cs in forest soil fractions including bulk soil and rhizosphere. Results in bulk soil (mean ± standard deviation) were 642 ± 215, 3.9 ± 2.7 and 0.3 ± 0.2 (mg kg^{-1}) for K, Rb and Cs concentrations, respectively. The similar trends appeared with higher K and Rb concentrations, and constant Cs level in rhizosphere compared with those in bulk soil. All these results may partly support our finding shown in Fig. 1, in which the alkali metal contents in soil vary at different soil compartments, probably depending on the fraction of mineral components.

To estimate the amounts of alkali metals (K, Rb and Cs) contained in the mineral components (X_{min}; mg) of a humus fraction of known thickness (d; cm), their amounts (x; mg), not concentration (mg kg^{-1}) were calculated using the equations (1) and (2) explained in the Material and methods section. Table 2 lists the results of regression analysis between SOM (%) and the amounts of alkali metals (mg) in each humus fraction in three sites (Sites I, II and III), where the symbols r, n, p, a and X_{min} in the table denote Pearson's correlation coefficient, number of data, probability, slope and intercept of the regression line, respectively. As seen in Table 2, estimated amounts of K, Rb and Cs in mineral components (X_{min}, the value at the

Table 2

intercept) decreased in the order, Site II > Site I > Site III, suggesting that the underlying minerals in the Site II contain the highest amounts of the alkali metals among others. Another finding in Table 2 is that increasing ratios (the ratio of slopes) of Rb to K (3~4 × 10^{-4}), and of Cs to K (1~3 × 10^{-4}) against SOM resulted in similar values in all the investigated sites. Although there appeared some variability for Cs to K values, degradation and/or weathering mechanisms may play a role controlling the amounts of alkali metals in humus horizon. Regarding uptake of K and Rb by plants, Simonsson et al. (2016) investigated sources of K and its relation with K/Rb ratio in soil and biomass under a Norway spruce stand (*Picea abies* Karst.). They showed quite low K/Rb ratios in the forest floor as well as interlayers of 2:1 clay minerals and biomass to conclude the importance of clay minerals as fixing and releasing K and Rb in the cycling of nutrients in forest soil environment.

The results in Table 2 offer a new finding that concentration of alkali metals in humus varies at different depths (or with elapsed time after deposition of fresh soil organic matter) suggesting that the importance of humus fractions to be considered. The properties of humus components are supposed to change gradually, and to affect downward migration of ^{137}Cs in humus horizon. To elucidate a long-term behavior of ^{137}Cs in the forest ecosystem, Karadeniz and Yaprak 2007 investigated distribution of both ^{137}Cs and stable ^{133}Cs in forest ecosystems. They attributed observed higher concentration of Cs in O2 horizon to its possible bioaccumulation in the horizon. They suggested that differences in the ^{137}Cs and stable ^{133}Cs depth profiles reflect the latter being enclosed in bedrock particles in the mineral horizons.

3.3 Depth distribution profiles of ^{137}Cs in bulk soil and in humus horizon

Figure 2 shows the depth distribution profiles of ^{137}Cs activity concentration (Bq kg^{1}) in bulk soil investigated in this study. Significant portion of ^{137}Cs was still found in the upper soil horizon (< 8 cm depth) 30 years after the Chernobyl accident in 1986, which had already been reported at different sampling points within the same sites in our previous paper (Takahashi et al. 2017). The highest activity concentration of ^{137}Cs were found to be 880 (± 5), 932 (± 6) and 240 (± 3) Bq kg^{-1} at 4-6 cm (Site I), 2-4 cm (Site II) and 6-8 cm (Site III), respectively. All these depths were included in humus horizon of soil, in which the site III of the oldest stand age (100-120 years) gave the lowest ^{137}Cs level among all. Figure 3 shows distribution of mean ^{137}Cs activity concentration (Bq kg^{-1}) in each fraction (H_u; 2-4 cm, H_m; 4-6 cm, and H_l; 6-8 cm in depth range) of humus horizon, in which the highest concentration of ^{137}Cs was found in the middle fraction (H_m) of humus in all the three sites.

Fig. 2

Fig. 3

Absolute activity (Bq) of ^{137}Cs was calculated using equation (1) in Material and methods to compare the amount of ^{137}Cs remaining in each humus fraction within 2 cm in thickness (200 cm^{3} in volume). Figure 4 summarizes the results on ^{137}Cs activity (Bq) as well as the amounts of alkali metals (K, Rb and Cs) in each humus fraction of in the three sites. Contrary to the results in Fig. 3, the ^{137}Cs activity clearly increased

Fig. 4

towards deeper humus fraction to reach the maximum value at the deepest portion (H$_l$, 6-8 cm) in any sites. This fact confirms the ^{137}Cs deposited on the forest floor moved downward within humus horizon. The presence of a ^{137}Cs peak in organic (O$_h$, or O$_f$) horizons has been reported in the literature (e.g. Ylipieti et al. 2008; Karandeniz and Yaprak 2011; Alexery et al. 2014; Takahashi et al. 2017; Karandeniz et al. 2015a). However, few papers have addressed distribution of ^{137}Cs profile within humus horizon using sets of ^{137}Cs activity (Bq) data. The results must be the first finding to clarify the ^{137}Cs distribution in humus horizon.

The same trend was observed on each alkali metal (K, Rb and Cs) distribution in humus fraction in Fig. 4, in which the amount of alkali metal increased towards deeper portion of humus fraction ($H_u < H_m < H_l$) as in the case of ^{137}Cs. Possible interactions of atmospheric ^{137}Cs with lithogenic alkali metals occur in biological uptake of nutrients in surface organic horizons. Chao et al. (2008) determined ^{137}Cs and alkali metals (K, Rb and K) in a perhumid montane forest ecosystem. They found that the ^{137}Cs/Cs ratio varied with soil horizons depending on the persistent supply of lithogenic Cs via weathering processes of minerals in deeper soil layers. They concluded that the ^{137}Cs/Cs ratio can be an indicator for monitoring the distribution and uptake of ^{137}Cs in the forest ecosystem.

3.4 Downward migration of ^{137}Cs in humus horizon

In the previous sections two important findings were obtained on behavior of ^{137}Cs and alkali metals (K, Rb and Cs) in humus, i) ^{137}Cs moved downward within humus horizon and its highest proportion was in the lowest fraction (H_l, 6-8 cm in depth range) at the sampling time in 2014, and ii) the amounts of alkali metals increased with increasing fraction of mineral components (or decreasing SOM) suggesting them as lithogenic origin.

The following two possibilities are now considered as explanations of the distribution of ^{137}Cs within humus horizon: 1) clay minerals such as illite and smectite fixing ^{137}Cs in their structures may be contained in the humus, and 2) downward movement of ^{137}Cs within humus horizon may be accompanied with degrading SOM. The first possibility came from the results in the literature that some clay minerals fix ^{137}Cs strongly within their layered structure, especially on micaceous clay minerals (Kruyts and Delvaux 2002; Nakao et al. 2014; Fuller et al. 2015).

No appreciable diffraction peaks corresponding to these clay minerals were detected in the lower range of diffraction angle ($3^\circ < 2\theta < 18^\circ$) on X ray diffraction (XRD) spectra of all the humus fractions (H_u, H_m and H_l) investigated in this study. An example of X ray diffraction pattern of a humus sample in the site I was shown in our previous paper (Takahashi et al. 2017). Table 3 lists major diffraction peaks (2θ, d-spacing) and assignment of the mineral in each humus fraction in the site III investigated in this study, in which only quartz was detected in all the cases. Amorphous materials, such as some alumino-silicates, hydrous iron

Table 3

oxides and strongly resistant organic residues may be contained in the lower portion of the humus horizon. All of them are supposed to be insignificant for the ^{137}Cs uptake (April and Deller 1990; Zhao et al. 2016).

In our previous paper (Takahashi et al. 2017), the ecological half-life of ^{137}Cs in humus layer in the sites I, II and III were estimated to be 21.7, 19.8 and 24.8 years, respectively. A similar estimation reported by Alexey et al. (2014) gave values of the ^{137}Cs effective half-life in the fermentative forest litters to be 8-19 years in Ukraine and Russian Federation forest sites. Several authors acknowledged that forest humus represents a sink for radiocesium recycling in a forest ecosystem, and that this sink for radiocesium will persist for many years (Melin et al. 1994; Bunzl et al. 1995; Schell et al. 1996). Furthermore, the time scale for this persistence will be decades rather than years (Rafferty et al. 2000).

The mean depth of the humus layer was obtained in each site and ranged from 3.6 to 5.4 cm using Eq. (3). When considering the elapsed time after the Chernobyl accident in 1986 (28 years as of 2014), the ^{137}Cs vertical migration rate was estimated to be in the range of 0.13-0.19 cm yr^{-1} (n=45) in this study. Similar values have been reported in the literature (Belli et al. 1994; Arapis and Karandinos 2004; Almgren and Isaksson 2006; Karandeniz et al. 20015b) in other forest soils under different geological and climatic conditions. Takahashi et al. (2017) estimated apparent burial rate from humus to mineral soil horizon to be in the range of 0.04-0.08 cm yr^{-1} (n=27). Higher migration rate of ^{137}Cs within humus layer compared with that into mineral horizon supports previous findings in the literature in which humus does not strongly retain ^{137}Cs within its structure compared with weathered micaceous minerals (Rigol et al.

2002). Kruyts and Delvaux (2002) reviewed main processes and parameters controlling the mobility of radiocesium in semi-natural soils. They showed that thick humus facilitates transfer of ^{137}Cs from soil to plant (biorecycling), because of weak interaction between ^{137}Cs and soil organic matter. They further investigated mobility of radiocesium in forest floors with different humus types of different mixing of mineral and organic residues (Kruyts et al. 2004). They suggested that the mixing of organic residues with Cs-fixing minerals is a key process in ^{137}Cs mobility. Another important evidence for very low ^{137}Cs migration rate into mineral horizon was that there was a great gap in ^{137}Cs activity concentration between the lowest humus fraction (H_l) and the uppermost mineral phase as shown in Fig. 2. Further observations should be continued to elucidate dynamics of ^{137}Cs remaining in lower humus fraction.

It should be noted that all the fractions of humus horizon in this study did not contain appreciable amounts of clay minerals, especially illites and smectites as determined by X-ray diffraction analysis (Table 3). However, it is likely that mineral components of little or no ^{137}Cs retention, such as amorphous oxides (aluminum, iron and manganese) and quartz, were contained in each humus fraction, because the proportion of organic components was not higher than 90 % even in the upper-most fraction (H_u) of humus horizon as suggested in Fig. 1. The results, including the downward migration rate of ^{137}Cs, indicate that organic components of humus have certain capacity for retaining ^{137}Cs within each humus fraction (Fuller et al. 2015).

Although little is known about interaction of ^{137}Cs with SOM in humus horizon in the literature, possible role of organic matter in radiocesium adsorption in soils was reviewed by Staunton et al. (2002), focusing largely on their own results in the laboratory. They observed that soil organic matter appreciably reduced the ability to immobilize Cs (decreasing distribution coefficient of radiocesium) on reference clay minerals (illite and montmorillonite), despite the weak and non-specific interaction of Cs with soil organic matter.

Regarding possible interactions of ^{137}Cs with SOM, soil microflora and microfauna may play a role in retaining radiocesium in their bodies (Kruse-Imer and Giani 2003; Karandeniz et al. 2015a).

It is now speculated that radiocesium may be released from dead microorganisms, and then incorporated again into newborn organisms; that is, radiocesium is circulated within the surface soil including humus horizon. To elucidate the possible relationship between microbial activity and deposited radiocesium in the profiles of forest soil, a change in their population with depth needs to be considered. Susyan et al. (2006) investigated the state of the microbial community in forest soils in Russia by determining microbial biomass and respiration, and population densities of micro-organisms. They found that the microbial activity decreased with increasing soil depth. More than 50 % of microbial biomass in the entire soil profile was concentrated in the upper portion (0-24 cm in depth) of the soil (humus horizon).

Decay of organic matter and nutrient cycling in forest soils are influenced by the interrelationships among climate, litter types and the variety and abundance of soil organisms. Regardless of considerable research on the effects of climate and litter quality, it is difficult to link the structure of soil communities to soil processes due to the lack of appropriate methods applicable to complex microbial communities (Leckie et al. 2004). To address this subject, they investigated the microbial communities (total microbial biomass and community composition) in forest floors of two forest sites contrasting in nitrogen availability in Canada. They found that bacterial and fungal communities were distinct in the fermentation (F), upper humus and lower humus layers of the forest floor and total biomass decreased in deep layers. Their results may support the results obtained in this study that abiotic properties of humus horizon were changed between upper and lower fractions.

A genetic technique on DNA and RNA quantification was applied to distinguish activity distribution of microbial biomass, especially bacterial and fungal biomass, in coniferous forest sites in the Czech Republic (Baldrian et al. 2012). They found that bacterial and fungal communities differ in their spatial distributions with fungal taxa more distinctly confined to either the litter of the organic horizon of soil and more heterogeneously distributed in the ecosystem. They also suggested that low abundance of several fungal taxa sometimes contributes to decomposition processed in soil. Several studies have indicated the importance of mycelia on the accumulation of radiocesium in surface soils (Guillitte et al. 1994; Brückmann and Woters 1994; Vinichuk and Johanson 2003; Yoshida et al. 2014). Regarding the distribution of fine roots in soils under coniferous stands (*Picea abies*), Børja et al. (2008) found that the highest root biomass was in the upper soil layers (humus layer and upper portion of mineral soil). Pertritan et al. (2011) also indicated that most fine roots were concentrated in the humus layer (more than 45 %) and in the top mineral soil (0-5 cm, about 15 %). This type of biological cycling is strongly associated with the downward movement of ^{137}Cs.

Based on these findings and reports in the literature, various organic compounds of the humus layer are likely to retain ^{137}Cs within their structures. Decomposition of the organic matter in soil is largely a biological process that occurs naturally. Humus is regarded as successive decomposition products with complex chemical structures, remaining in soil for relatively long time. It is expected that properties of humus may change gradually with degrading organic matter, i.e. with depth, in undisturbed forest soil, and that the organic compounds in lower humus fraction (H_l) may be much more resistant to microbial oxidation than those in the upper fraction (H_u) (Gonet et al. 2008; Schaeffer et al. 2015). Dynamics of soil organic matter in humus could be considered with a parameter, mean residence time, of organic components. Different values of mean residence time and even different concept on humus formation were reported, which were inevitably caused by the complexity of organic matter in composition and structure, environmental parameters and separation techniques (Certini et al. 2004; Delula and Aplet 2008; Schmidt et al. 2011). Dynamics of soil humus should be investigated through carbon isotopes (^{13}C and ^{14}C) technique, which enables possible sources (and date) of carbon contained in the humus component (Paul et al. 1964; Fujiyoshi et al. 2010, 2012). Decomposition of soil organic matter therefore depends both on biotic and abiotic environment, thus ecosystem property (Lehmann and Kleber 2015).

As shown in Fig. 4, the ^{137}Cs activity (Bq) remained in humus fraction decreased consistently in the order: $H_U < H_m < H_l$. The result together with those on ^{137}Cs mean downward migration rate (0.1-0.2 cm yr^{-1}) estimated in this study suggests that ^{137}Cs moved downward within humus horizon. Assuming that ^{137}Cs is released within a humus fraction due to the decomposition of host organic components, the downward migration of ^{137}Cs may depend on degradation rate of the organic components. If this assumption is appropriate, mean residence time of the host organic components may give similar mean residence time of ~30 (29~36) years to that of ^{137}Cs estimated from the ecological half-life of ^{137}Cs in humus horizon (Site I; 21.7, Site II; 19.8 and Site III; 24.8 years) estimated in our previous study (Takahashi et al. 2017). It is suggested that speciation of ^{137}Cs in humus fractions may be a potential approach to identify a target organic component, and that mean residence time of the humus components should be evaluated to assess a mechanism of downward migration of ^{137}Cs in humus horizon, while applying powerful tools like genetic and isotope (stable and radioactive) techniques.

4 Conclusions

Bulk soil and its humus horizon were separately collected from three coniferous forest sites in the Czech Republic in September 2014. The highest activity concentration of ^{137}Cs (Bq kg^{-1}) appeared in the upper portion of the soil (mainly in humus horizon) about 30 years after the Chernobyl accident. Increasing activity of ^{137}Cs (Bq) was found in the lower humus horizon (H_l, 4~6 cm depth range) in which no appreciable amounts of micaceous clay minerals like illite and smectite were detected in all the sites. Similar depth distribution profiles appeared for lithogenic alkali metals (K, Rb and Cs) in humus as in the case of fallout nuclide ^{137}Cs, suggesting that mineral components with little retention ability for ^{137}Cs increased at lower humus fraction. It is likely that organic components of humus layer have certain capacity of retaining ^{137}Cs in their structures. Downward migration rate of ^{137}Cs within the humus horizon (0.13-0.19 cm yr^{-1}) estimated in this study was about three times higher than those (0.04-0.06 cm yr^{-1}) previously obtained from the humus to mineral horizon. These findings suggest that ^{137}Cs moves downward with degrading organic matter within the humus horizon. Comparing depth distribution profiles between ^{137}Cs activity (Bq) and the amount (g) of alkali metals (K, Rb and Cs) may reflect possible association of ^{137}Cs with the alkali metals during biological activity within humus. Further investigation is needed to elucidate mechanism of ^{137}Cs migration within humus horizon, especially interactions of ^{137}Cs and degrading organic components.

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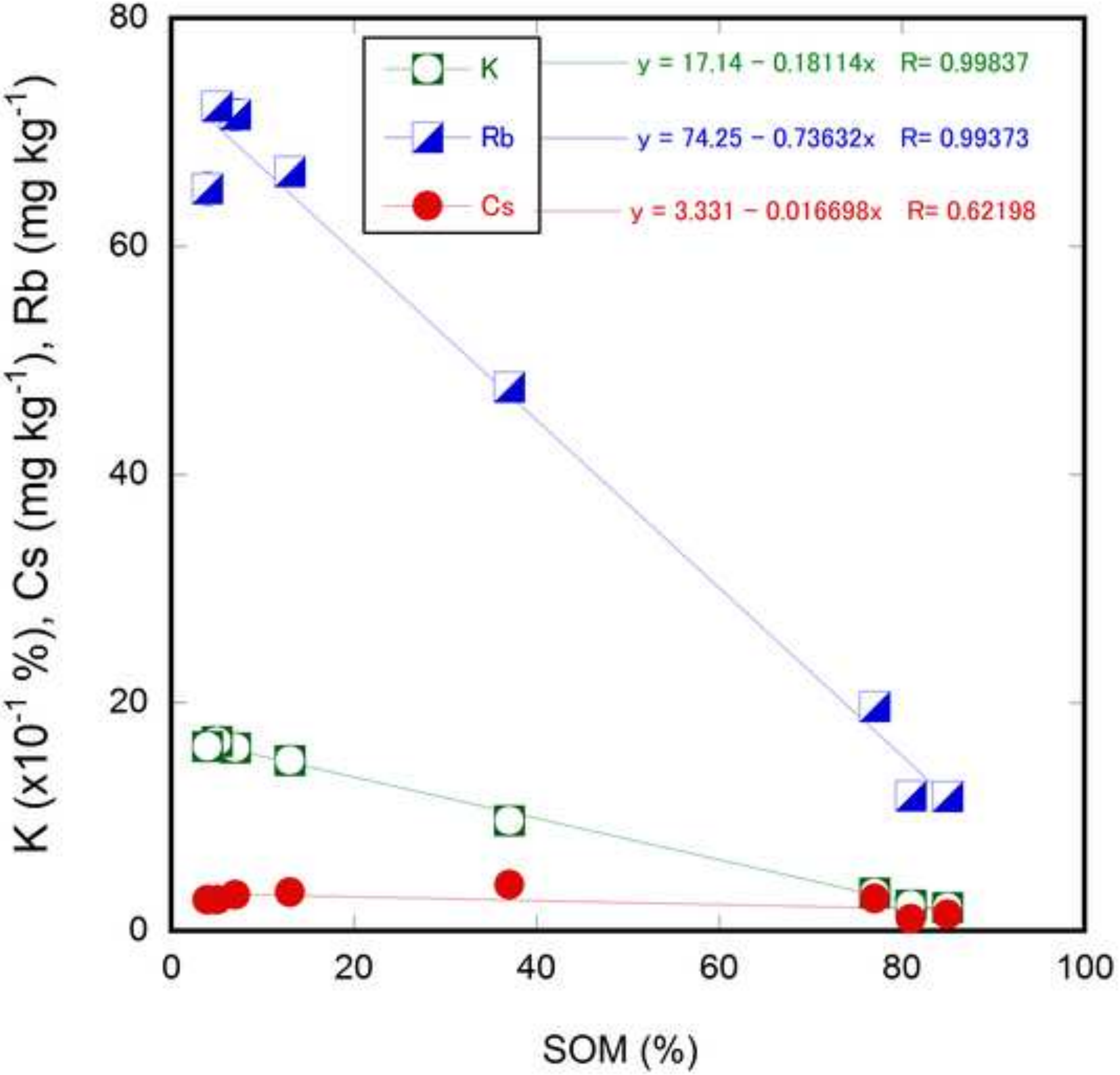
K
Rb
Cs
y = 17.14 − 0.18114x R= 0.99837
y = 74.25 − 0.73632x R= 0.99373
y = 3.331 − 0.016698x R= 0.62198
K (x10^-1 %), Cs (mg kg^-1), Rb (mg kg^-1)
0
20
40
60
80
0
20
40
60
80
100
SOM (%)

Fig. 1

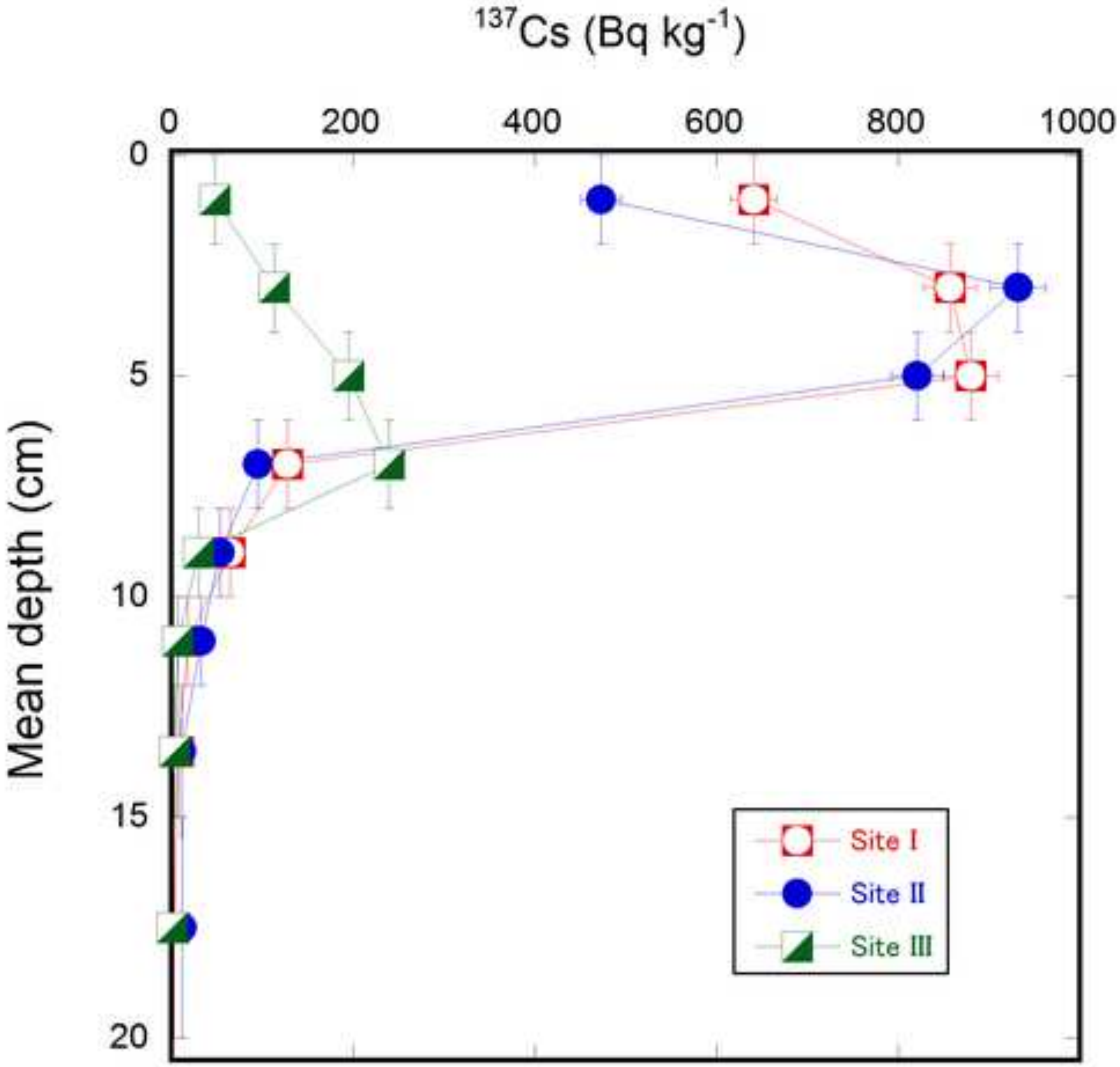
^{137}Cs (Bq kg^{-1})
0
200
400
600
800
1000
Mean depth (cm)
0
5
10
15
20
Site I
Site II
Site III

Fig.2

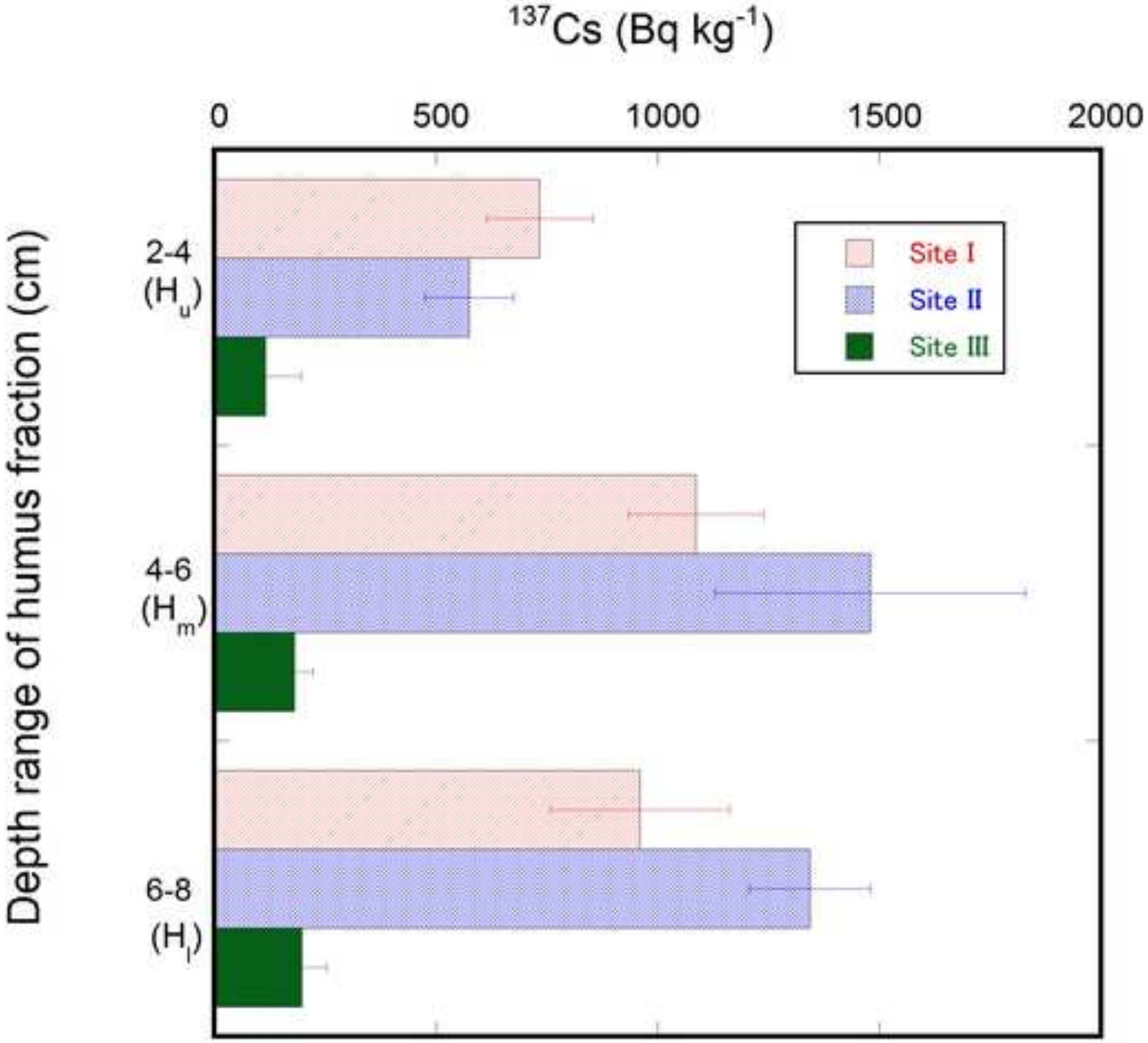
^{137}Cs (Bq kg^{-1})
0
500
1000
1500
2000
Depth range of humus fraction (cm)
2-4
(H$_u$)
4-6
(H$_m$)
6-8
(H$_l$)
Site I
Site II
Site III

Fig.3

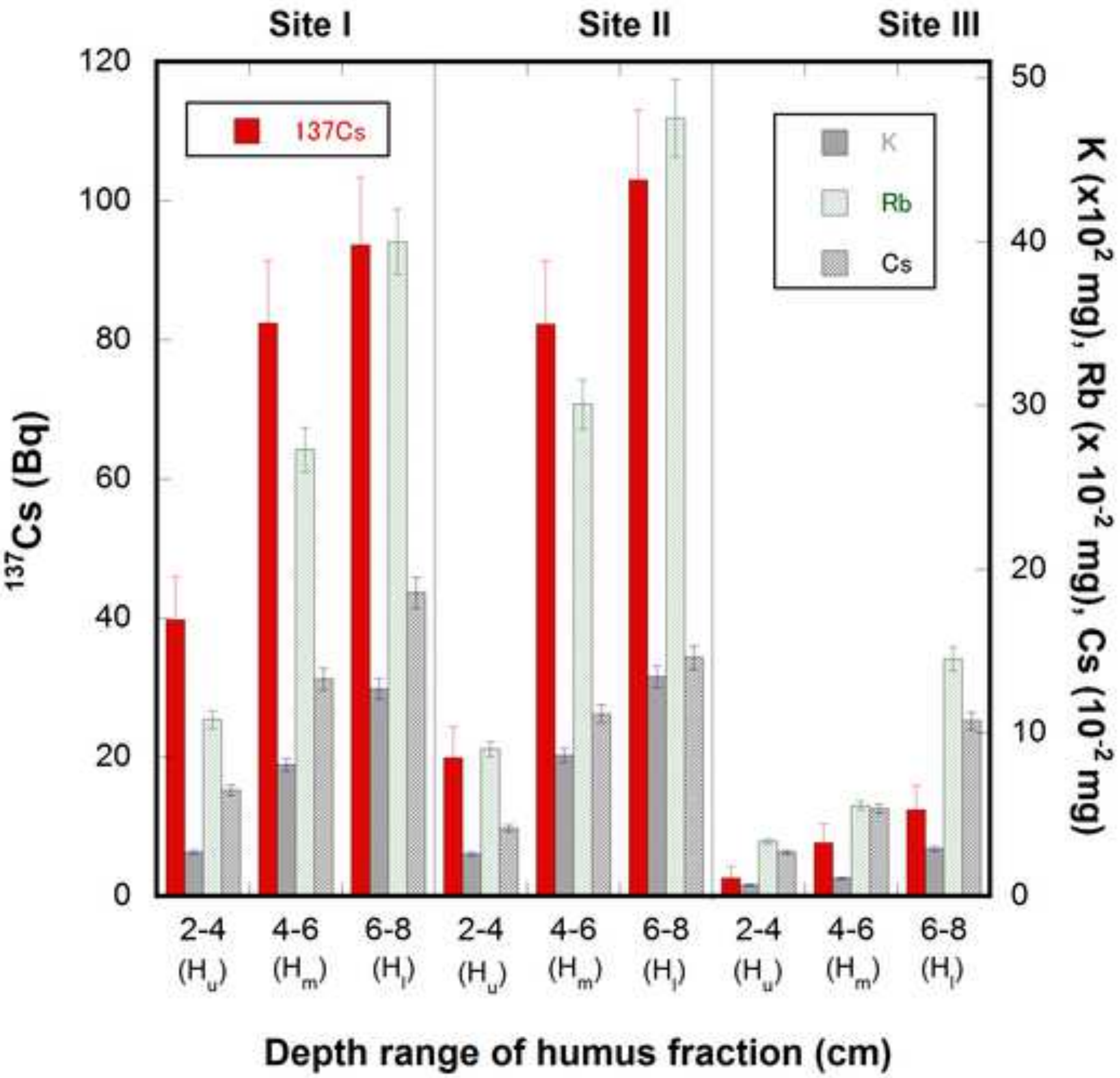
Site I
Site II
Site III
137Cs
K
Rb
Cs
^{137}Cs (Bq)
0
20
40
60
80
100
120
K (x10^2 mg), Rb (x 10^{-2} mg), Cs (10^{-2} mg)
0
10
20
30
40
50
2-4 (H$_u$)
4-6 (H$_m$)
6-8 (H$_l$)
2-4 (H$_u$)
4-6 (H$_m$)
6-8 (H$_l$)
2-4 (H$_u$)
4-6 (H$_m$)
6-8 (H$_l$)
Depth range of humus fraction (cm)

Fig. 4

Figure caption

Figure. 1 Relationship between the amount of soil organic matter (SOM; %) and concentration of alkali metals (K; ×10^{-1} %, Rb; mg kg^{-1}, Cs; mg kg^{-1}) in surface soil down to a depth of 20 cm in the investigated sites (I, II, and III). Correlation coefficient of Cs with SOM was lower compared with those for K and Rb. Standard deviation (±1 S.D.) of each data set was 1.8~4.8, 1.1~3.5 and 0.98~4.5 % for K, Rb and Cs, respectively.

Figure 2 Depth distribution profiles of ^{137}Cs activity concentration (Bq kg^{-1}) in surface soil (<20 cm depth) in the sites I, II and III. Most of the Chernobyl derived ^{137}Cs still existed in the upper portion (< 8 cm depth) of the soil. The site III showed the lowest activity concentration of ^{137}Cs among the three sites.

Figure 3 Depth distribution profiles of ^{137}Cs activity concentration (Bq kg^{-1}) in each humus fraction of 2 cm in mean thickness. The highest ^{137}Cs activity concentration was found in the middle fraction (H_m) of humus in all the sites investigated.

Figure 4 Depth distribution of ^{137}Cs activity (Bq) and of the alkali metals (K, Rb and Cs) in each humus fraction (H_u, H_m, and H_l) in the sites I, II and III. Both the ^{137}Cs activity and the amounts of alkali metals consistently increased to deeper fraction of humus horizon, in which the site III gave the lowest levels compared with other sites.

Table 1 Summary of the site-specific properties of soil including those (location, climate, soil type, tree type, stand age, ^{137}Cs inventory, annual apparent burial rate of ^{137}Cs to mineral horizon and ecological half-life) described in our previous study (Takahashi et al. 2017). Also included properties (mean thickness, pH, moisture SOM and density) of humus fractions (H$_u$, H$_m$, H$_l$) in humus horizon.

Properties	Site								
	I (Přemyslov)			II (Opatovice)			III (Průhonice)		
Location	50.111ºN, 17.065 ºE			49.962ºN, 15.065 ºE			49.986ºN, 14.542 ºE		
Altitude (m)	800			390			320		
Annual mean temperature (ºC)	4-5			7-8			8-9		
Annual mean precipitation (ºC)	800-1000			500-550			500-550		
Soil type	Mesobasic cambisol			Modal cambisol			Modal cambisol		
Tree type	*Picea abies*			*Picea abies*			*Picea abies*		
Stand age (y)	60-80			80-100			100-120		
^{137}Cs inventory in 1986 (kBq m^{-2})	9.46			1.16			5.56		
Annual apparent burial rate of ^{137}Cs (cm y^{-1})	0.06 ± 0.01			0.04 ± 0.01			0.08 ± 0.01		
Ecological half-life of ^{137}Cs (y) in humus	19.8			24.8			21.7		
Humus horizon*	H$_u$	H$_m$	H$_l$	H$_u$	H$_m$	H$_l$	H$_u$	H$_m$	H$_l$
Thickness (mean, cm)	2	2	2	2	2	2	2	2	2
pH (mean)	4.5	3.7	3.4	3.7	3.5	3.6	3.5	3.4	3.5
Moisture (mean, %)	55	40	30	42	30	27	39	37	22
SOM (mean, %)	81	56	50	83	77	68	81	66	33
Density (dry, mean, g cm^{-3})	0.27	0.38	0.49	0.17	0.28	0.38	0.11	0.21	0.32

* Upper fraction (H$_u$), Middle fraction (H$_m$), Lower fraction (H$_l$)

Table 2 Results of the regression analysis between SOM (%) and the amounts of alkali metals (K, Rb and Cs) contained in each humus fraction in the sites I, II and III. The symbols r, n, p, a and X_{min} in the table denote Pearson's correlation coefficient, number of probability, slope and intercept of the regression line, respectively. The X_{min} value is the content (mg or g) of each alkali metal contained in mineral components of humus horizon with a thickness of 6 cm. Increasing ratio of ^{137}Cs (Bq) with decreasing SOM was also shown as a slope (a) in this table.

Element (radionuclide)	Site														
	I (Přemyslov)					II (Opatovice)					III (Průhonice)				
	r	n	p	a	X_{min}	r	n	p	a	X_{min}	r	n	p	a	X_{min}
K (g)	0.95	9	<0.01	0.29 (0.06)	2.6	0.99	9	<0.01	0.71 (0.14)	6.25	0.99	9	<0.01	0.05 (0.01)	0.44
Rb (mg)	0.96	9	<0.01	0.09 (0.02)	0.8	0.99	9	<0.01	0.25 (0.05)	2.20	0.99	9	<0.01	0.02 (0.01)	0.22
Cs (mg)	0.96	9	<0.01	3.58 (0.77)	0.35	0.96	9	<0.01	6.77 (1.33)	0.62	1.0	9	<0.01	1.67 (0.32)	0.16
^{137}Cs (Bq)	1.0	9	<0.01	1.75 (0.33)	-	0.93	9	<0.01	5.35 (0.66)	-	0.97	9	<0.01	0.20 (0.04)	-

Table 3 Major diffraction peaks (2θ and d-spacing) and assignment of the mineral observed in each humus fraction (H_u, H_m and H_l) in the site III (3° < 2θ < 38°).

Site III	2θ °(CuKα)	d (Å)	Assignment
H_u	20.88	4.26	Quartz
	26.62	3.34	Quartz
	36.54	2.46	Quartz
H_m	20.88	4.26	Quartz
	26.61	3.34	Quartz
	36.57	2.46	Quartz
H_l	20.82	4.26	Quartz
	26.66	3.34	Quartz
	36.54	2.46	Quartz

Reply to the reviewer's comments and advices

We would greatly appreciate your help to refine our current revised manuscript. Reading the general comments of yours, we reconsidered the important findings obtained in the present study to shorten the extra discussion. We also thank you for suggesting many mistakes in English. We checked all of them to be revised as shown in red in the text (The corrected parts in blue were undertaken by the authors themselves). The followings were the reply to the reviewer's comments.

General comments

Page 21, lines 354-360

We erased whole the paragraph on biotic and abiotic effects on the sorption of stable cesium and strontium in soil components conducted in the laboratory (Seeprasert et al. 2016). As the reviewer suggested, this part was not directly connected to our results.

Page 22, lines 374-402

This part, concerned with "mean residence time" of soil organic components, may beyond the scope of this paper. We decided to shorten this part, and only to add a potential usefulness of carbon (^{13}C and ^{14}C) isotope technique to elucidate the source and date of soil organic components obtained in our previous studies (Fujiyoshi et al. 2010, 2012) as well as a work in the literature (Paul et al. 1964).

Typographical corrections

We corrected all the suggestions by the reviewer (as shown in red in the text), except for one:

Page 9, line 138

Unit of dry density is [g cm^{-3}]. The value 10^{-3} is multiplied to the right side in the equation (1).

The corresponding author, Ryoko FUJIYOSHI (July 25 2017)

July 26 2017

List of changes

The table below lists changes in the previous manuscript, according to the comments given by the Editor and Reviewer. All the corrections were written in red in the text of the revised manuscript.

Line(s)	Line(s) revised	Comment	Response	Note
		Correct numbering of Figures (Figs. 2, 3 and 4)	Done	Editor
Abstract				
20	20	Change the word	Done	Reviewer
21	21-22	Delete dangling phrases	Done	Reviewer
35	34	Delete dangling phrases	Done	Reviewer
41	39	Add “The”	Done	Reviewer
41	39	Change the phrase	Done	Reviewer
42	40-41	Re-write the phrase	Done	Reviewer
Introduction				
48	46-47	Revise the sentence	Done	Reviewer
53	51	Re-write the phrase	Done	Reviewer
59	56-57	Replace the words	Done	Reviewer
64	61	Replace the word	Done	Reviewer
65	62	Re-write the phrase	Done	Reviewer
79	76	Add “the”	Done	Reviewer
80	77	Re-write the phrase	Done	Reviewer
82	79	Add “the”	Done	Reviewer
92	89	Change the word	Done	Reviewer
96	93	Add “the”	Done	Also line 91
Material and methods				
99	96	Replace the phrase	Done	
138	135	Unit of dry density	Unit of dry density is [g cm^{-3}]. The value 10^{-3} is multiplied to the right hand side of the equation (1).	Explanation
Results and discussion				

166	163	Replace subsection title	Done	Reviewer
187-188	184	Replace the phrase	Done	Reviewer
210	208	Re-write the phrase	Done	Reviewer
211	208	Add “the”	Done	Reviewer
214	211	Replace the word	Done	Reviewer
277	274	Replace the word	Done	Reviewer
319	316	Replace the word	Done	Reviewer
324	321	Add “the”	Done	Reviewer
326	323-324	Re-write the sentence	Done	Reviewer
349	346	Move the word	Done	Reviewer
350	347	Define the word	Added the full spelling	Reviewer
354-360		Not directly connected to the present study	The whole paragraph on biotic and abiotic effects on the sorption of stable cesium and strontium in soil components conducted in the laboratory (Seeprasert et al. 2016).	Reviewer (Authors)
374-402	375-377	Beyond the scope of this study	This part was shortened, and added a potential usefulness of carbon (^{13}C and ^{14}C) isotope technique to elucidate the source and date of soil organic components obtained in our	Reviewer (Authors)

			studies (Fujiyoshi et al. 2010, 2012) as well as a work in the literature (Paul et al. 1964).	
411	387	Delete “then” and “highly”	Done	Reviewer
412	388	Replace the word	Done	Reviewer
413	390	Delete the phrase	Done	Reviewer
Conclusion				
423	399	Replace the word	Done	Reviewer
430	406	Adjust the style	Done	Reviewer
431	407	Delete the phrase	Done	Reviewer