



Article

Influence of Soil Characteristics and Land Use Type on Existing Fractions of Radioactive ^{137}Cs in Fukushima Soils

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Abstract: Radioactive cesium (^{137}Cs) in distinct soil fractions provides key information to its bioavailability, and therefore determining the effect of soil characteristics and land use types on existing fractions of ^{137}Cs in soils is important for predicting future ^{137}Cs mobility in Fukushima. Thus, the objective of this study was to investigate the influence of soil characteristics and land use types on sequentially extracted fractions of ^{137}Cs in Fukushima. In this study, five coniferous forest soils, eight arable soils, and eight paddy rice soils were sampled in 2012 and 2013. The ^{137}Cs in the soils were separated into four fractions; water-soluble, exchangeable, organic matter-bound and residual fractions. More than 90% of the soil ^{137}Cs fraction for arable and paddy rice soils was found in the residual fraction, implying significantly reduced bioavailable ^{137}Cs with higher fixation. In contrast, forest soils measured higher exchangeable and organic matter-bound fractions of 5%–33% and 9%–44%, respectively, implying future ^{137}Cs mobility in the forest ecosystem. Correlation analysis showed a significant negative correlation ($p < 0.05$) between the organic matter fraction and residual fraction in both arable and paddy rice soils. There was a significant positive correlation ($p < 0.05$) for both exchangeable and residual fractions with cation exchange capacity (CEC), total carbon (TC) and total nitrogen (TN) values in arable soils. Organic matter content influenced both exchangeable and residual fractions. It was not clear whether organic matter played a direct role in ^{137}Cs fixation or mobility in the agricultural soils. In paddy rice soils, the organic matter fraction showed a significant negative correlation with TC and TN values. Soil pH was significantly negatively correlated ($p < 0.05$) with both water-soluble and residual fractions in forest soils but positively ($p < 0.1$) with the organically bound ^{137}Cs fraction.

Keywords: agricultural soils; ^{137}Cs bioavailability; forest soils; sequential extraction; Fukushima soils

1. Introduction

The Fukushima Dai-ichi Nuclear Power Plant accident, which resulted from the large tsunami which was an aftermath of the 11 March 2011 Tohoku-Oki earthquake [1], has led to the contamination of the soil environment with radiocesium, mainly cesium-134 (^{134}Cs) and cesium-137 (^{137}Cs) in Fukushima and its neighboring prefectures [2]. Cesium-137 is of serious environmental concern due to its high biological availability [3] and relatively long half-life of 30.17 years compared to ^{134}Cs ($t_{1/2} = 2.06$ years). The existence of ^{137}Cs in various forms or fractions is one identifiable key factor for characterizing the fate of radionuclides in the environment and plays an essential role in the migration of ^{137}Cs in the soil. In addition, knowledge about ^{137}Cs speciation, which is largely dependent on the soil physicochemical properties, is of relevance for a better understanding of its dispersion, transport and biological impact in the soil environment [4].

Early studies conducted in contaminated soils in Fukushima showed that ^{137}Cs is retained in the top 5 cm of surface soils, with considerably reduced amounts below the 5 cm depth [5]. The retention of ^{137}Cs in soils could largely result from the adsorption onto clays [6]. Owing to their large surface areas, a dominant proportion of ^{137}Cs was reported to be accumulated within the clay fractions (69%–93% of ^{137}Cs was found in clay fraction for soil with 24%–27% clay content [7]). Because ^{137}Cs retention in soils can increase with increasing clay content [8], clay content has been considered as one of the crucial factors in regulating ^{137}Cs retention [1]. Also, several studies have shown that clay mineral composition, mainly an abundance of micaceous clay mineral, has the ability to influence the long-term stabilization of ^{137}Cs [9,10]. Cesium exists as a univalent cation in solution with small hydration energy, and it is easily adsorbed to negatively charged constituents in soil, especially illitic clay mineral [11]. The strong affinity is supposed to come from the presence of frayed edge sites (FES), which accounts for a minor part of the cation exchange capacity (CEC) of soil clay mineral [6,12]. Cesium fixation increases with time as the cation penetrates the lattice [4]. The adsorption capacity of the clay fraction for Cs is important even when soil clay minerals only make up a small part of the clay fraction [6].

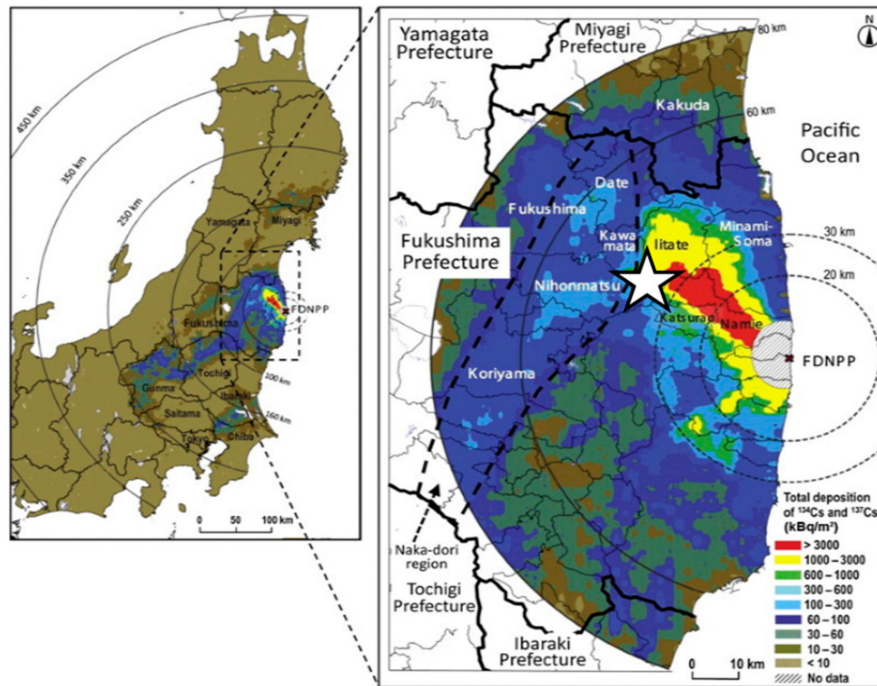
Soil organic matter also has the ability to retain ^{137}Cs . Several studies have shown that ^{137}Cs is more easily available to biological systems in organic matter-rich soils [13,14]. However, the adsorption of ^{137}Cs to organic matter is determined by its concentration in relation to other competing cations and the cation exchange capacity (CEC) of the specific organic substance [11,15]. Organic matter has a smaller affinity for ^{137}Cs in comparison to clay minerals, and the extent of adsorption is thus limited due to its low selectivity for ^{137}Cs [11,16]. According to Dumat et al. [12], either clay minerals prevent Cs adsorption to organic matter, or the binding of Cs to organic matter is so weak that it is easily reversible. This notwithstanding, the study of Dumat and Staunton [17] confirmed the general phenomenon that the presence of humic substance decreased the adsorption of cesium on mineral clays, as indicated in an earlier study by Staunton and Roubaud [18].

The retention of ^{137}Cs in soils is defined by the balance between organic matter content and clay mineral composition. This balance, however, is not only dependent on soil variety but also land-use type. Forest soils have fewer disturbances and more preserved mineral horizons compared to agricultural land uses [19]. In addition, there are usually no artificial inputs such as fertilizer, manure, and pesticides occurring in forest soils, with plant communities relying on their inherent soil nutrient cycling to support their nutritional needs. Important characteristic features of forest soils such as the presence of litter and organic horizons and the depth distribution of roots are considered to be important for ^{137}Cs availability [9,20]. Thus, it is hypothesized that, in addition to soil characteristics, different land-use types can influence the existing ^{137}Cs fractions in soils in Fukushima. The purpose of this study was to investigate the existing ^{137}Cs fractions in various soils in Fukushima by a sequential extraction procedure and to determine the influence of soil characteristics on the existing ^{137}Cs fractions, comparing ^{137}Cs mobility in both forest and agricultural land uses.

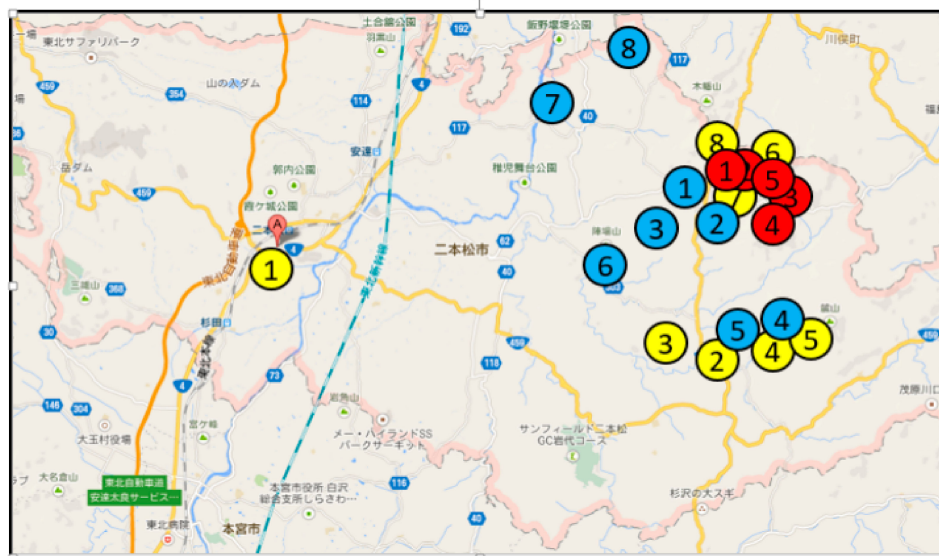
2. Materials and Methods

2.1. Soil Sampling and Preparation

Soil samples were collected from eight arable fields on 5 and 6 August 2013—referred to as Arable 1 to Arable 8—eight paddy rice fields on 8 and 9 December 2013—referred to as Paddy 1 to Paddy 8—and five coniferous forest sites on 31 October, 27 and 28 November 2012—referred to as Forest 1 to Forest 5—in Nihonmatsu City, Fukushima Prefecture, Japan, which is located 50 km away from the nuclear power plant (Figure 1) (Table 1).



(a)



● Upland fields ● Paddy rice fields ● Forest sites

(b)

Figure 1. Map of (a) study area and (b) sampling sites of soils.

Table 1. GPS location, soil classification, surface geology and sampling dates of sampling sites.

| Field | Location | | Soil Group (World Reference Base, 2006) | Surface Geology | Sampling Date |
|----------|----------|-----------|--|-------------------|------------------|
| | N | E | | | |
| Arable 1 | 37.34.41 | 140.25.43 | Haplic Fluvisol | Granite | 6 August 2013 |
| | 37.33.26 | 140.35.22 | Gleyic Fluvisol | Granite | 5 August 2013 |
| | 37.33.26 | 140.34.05 | Cambisol | Granite | 5 August 2013 |
| | 37.33.32 | 140.36.21 | Andosol | Granite | 5 August 2013 |
| | 37.33.36 | 140.36.40 | Andosol | Granite | 5 August 2013 |
| | 37.36.28 | 140.35.03 | Andosol | Granite | 6 August 2013 |
| | 37.36.22 | 140.35.02 | Cambisol | Granite | 6 August 2013 |
| | 37.36.09 | 140.34.38 | Cambisol | Granite | 8 December 2013 |
| Paddy 1 | 37.36.37 | 140.35.03 | Haplic Fluvisol | Mud, sand, gravel | 8 December 2013 |
| | 37.36.10 | 140.34.55 | Gleyic Fluvisol | Mud, sand, gravel | 8 December 2013 |
| | 37.35.24 | 140.33.46 | Gleyic Fluvisol | Granite | 8 December 2013 |
| | 37.36.14 | 140.34.43 | Andosol | Granite | 9 December 2013 |
| | 37.33.41 | 140.36.30 | Gleyic Fluvisol | Granite | 9 December 2013 |
| | 37.34.52 | 140.32.45 | Gleyic Fluvisol | Granite | 9 December 2013 |
| | 37.37.31 | 140.31.41 | Gleyic Fluvisol | Granite | 9 December 2013 |
| | 37.38.32 | 140.33.11 | Gleyic Fluvisol | Granite | 9 December 2013 |
| Forest 1 | 37.35.58 | 140.35.20 | Cambisol | Granite | 27 November 2012 |
| | 37.35.59 | 140.35.24 | Cambisol | Granite | 27 November 2012 |
| | 37.35.54 | 140.36.52 | Cambisol | Granite | 31 October 2012 |
| | 37.35.38 | 140.36.52 | Cambisol | Granite | 28 November 2012 |
| | 37.36.05 | 140.36.29 | Cambisol | Granite | 31 October 2012 |

The general soil types found at the sampling sites were Andosol, Cambisol, Haplic Fluvisol and Gleyic Fluvisol [21] (Table 1). Composite soil samples were collected at a surface soil plow depth of 0–10 cm by five-point diagonal sampling to give representative samples of each field, with the most extreme points, 1 and 5, measuring 16.2 km apart. For forest soils, a litter layer or cover consisting of easily recognizable, originally shaped organic components made up of periodically falling raw litter of mainly dead fallen leaves, branches, and early fermented and fragmented litter components were removed from the soil surface before sampling. Homogenized soils were air-dried for 2 weeks, crushed and sieved through a 2 mm mesh, and plant roots were removed. All analyses were carried out in 2014.

2.2. Soil Analysis

2.2.1. Soil Physical and Chemical Analysis

Soil particle size distribution analysis was performed by sedimentation and the pipette method [22] to determine the soil texture. Soil pH was determined by the electrode method. The pH was measured in a 1:2.5 (*w/v*) ratio of 10 g soil to 25 mL water supernatant suspension, after shaking for 1 h, using the Beckman PKG-260 pH meter (Beckman Coulter, California, USA) [23]. Total carbon (TC) and total nitrogen (TN) contents were determined using an automatic highly sensitive CN analyzer (Sumigraph NC-220F; Sumika Chemical Analysis Service Co., Osaka, Japan). Soil organic matter (OM) content was determined using the hydrogen peroxide digestion method [24]. Cation exchange capacity (CEC) was determined by the indophenol blue colorimetric method with a UV-visible spectrometer (UV mini 1240, Shimadzu, Japan). Exchangeable cations were extracted with 1 M ammonium acetate (NH₄OAc, pH 7) using the Schollenberger method [25]

2.2.2. Measurement of ¹³⁷Cs Activity

To measure the activity concentration of ¹³⁷Cs in the soil, an automatic gamma counter (2480 Wizard, Perkin-Elmer, Tokyo, Japan) equipped with a well-type NaI detector, with lead shielding, coupled to a multi-channel analyzer calibrated for the energy range 15–2048 keV with a maximum dead time of 2.5 μs was used. The energy and efficiency calibrations for this detector were performed using a

blank (background) sample and a reference ^{137}Cs standard (Spectrum Techniques, Knoxville USA) with an activity of 10.878 kBq. Samples were loaded into 20 mL vials, and the ^{137}Cs activity of each sample was determined by counting gamma emissions for 1200 s. Cesium-137 activity concentrations were measured in 2014. The activity concentration in samples were expressed in Bq kg^{-1} dry weight. The detector efficiency for ^{137}Cs was 47% and the energy resolution was <10%.

2.2.3. Sequential Extraction of ^{137}Cs

Cesium-137 was sequentially extracted from 20 g soil (dry weight), which was sieved through a 2 mm sieve as mentioned previously for soil physical and chemical analysis. Prior to extraction, weighed samples were placed in 20 mL vials, and the initial ^{137}Cs activity of soil samples was measured. A modification of the sequential extraction procedure [26,27] was carried out as shown in Table 2, with four different ^{137}Cs fractions in soil extracted: water-soluble (W), exchangeable fraction (E), organic matter-bound fraction (O) and residual fraction (R).

Table 2. Procedure for sequential extraction.

| Fraction | Extractant | Treatment | Reference |
|----------------------|---|---|--------------------------------|
| W, Water-soluble | 200 mL distilled H_2O | Shaken for 1 min at room temperature | Forsberg and Strandmark (2001) |
| E, Exchangeable | 200 mL 1 M NH_4OAc (pH 7) | Shaken for 2 h at 20 °C | Forsberg and Strandmark (2001) |
| O, Organic matter | 10 mL 30% H_2O_2 (pH 2) | Shaken for 5 h at 80 °C | Puhakainen et al. (2001) |
| R, residual fixation | Residue | Remaining ^{137}Cs activity content after organic matter fraction extraction | Forsberg and Strandmark (2001) |

For the extraction procedure, each soil sample was weighed out into a 300 mL plastic bottle with the respective amounts of extractants added, and the samples were treated as described in Table 2. The extractants were distilled water, ammonium acetate (NH_4OAc), and hydrogen peroxide (H_2O_2). For organic matter-bound extracted ^{137}Cs , soil samples were weighed out into a 500 mL glass beaker with 10 mL of 30% H_2O_2 to allow for organic matter digestion. Samples were further heated to ensure complete organic matter digestion, with 1 mL H_2O_2 added in time intervals until the supernatant was clear. All supernatants were filtered through a 0.45 μm membrane filter with a suction pump after shaking at the required temperatures, as described in Table 2. Prior to the next extraction, the soil extracts were washed by adding 100 mL distilled H_2O . The difference in measured ^{137}Cs concentration between the immediate extraction and the previously extracted fraction was defined as the amount for each fraction. The residual fraction, which is mainly clay mineral-bound and strongly fixed, was determined from the remaining soil after the extraction of the organic matter fraction. The activities in each extract were expressed as a percentage of the initial activity in the soil.

2.3. Statistical Analysis

Correlations among ^{137}Cs fractions, between ^{137}Cs fractions and soil characteristics, and among soil characteristics were analyzed by Pearson Product Moment Correlation (SigmaPlot 12.5, Systat Software, San Jose, CA, USA).

3. Results and Discussion

3.1. Soil Physiochemical Properties

The pH was acidic to moderately alkaline, with values ranging from 5 to 7.5, 5.8 to 6.8 and 4.6 to 5.4 for arable, paddy rice and forest soils, respectively. Among the three land uses, arable soil recorded higher pH values, while forest soil recorded lower pH values (Table 3).

CEC, TC, and TN were highest in forest soils and least in paddy rice soils, with some exceptions. Forest 4 recorded the highest CEC and TN values of 20.7 $\text{cmol } 100 \text{ g}^{-1}$, and 7.1 g kg^{-1} respectively, but the highest TC value of 124 g kg^{-1} was recorded in Forest 5. The lowest CEC and TC values of 5.7 $\text{cmol } 100 \text{ g}^{-1}$ and 8.7 g kg^{-1} , respectively, were recorded in Paddy 4. TN values in arable and paddy rice

soils were similar, except in Arable 4 and Arable 8, with values of 6.0 and 4.3 kg⁻¹. The soil texture of paddy rice soils was sandy loam for all fields except Paddy 1, which was sandy clay loam, and Paddy 3 and Paddy 4, which were featured by a loamy sand texture. Arable and forest soils were sandy loam to clay loam, depending on the relative amount of sand and clay fractions.

Table 3. Basic characteristics of soils at study sites.

| Field | ¹³⁷ Cs ^a | pH (H ₂ O) | CEC ^b | TC ^c | TN ^d | Soil Texture ^e | | |
|----------|--------------------------------|--------------------------|------------------|-----------------|-----------------|-----------------------------|-----------------------|-----------------------|
| | (Bq kg ⁻¹) | | | | | (cmol 100 g ⁻¹) | (g kg ⁻¹) | (g kg ⁻¹) |
| Arable 1 | 1718 | 5.7 | 6.9 | 15.6 | 1.4 | 34.0 | 19.8 | 46.3 |
| 2 | 1068 | 5.6 | 8.8 | 16.3 | 1.5 | 12.2 | 19.9 | 67.9 |
| 3 | 895 | 5.7 | 17.8 | 12.9 | 1.3 | 27.6 | 28.1 | 44.3 |
| 4 | 3019 | 7.1 | 11.4 | 64.3 | 6.0 | 22.5 | 25.4 | 52.1 |
| 5 | 2341 | 6.3 | 10.5 | 20.6 | 1.9 | 31.6 | 29.9 | 38.5 |
| 6 | 569 | 5.0 | 9.5 | 18.8 | 2.0 | 19.9 | 17.6 | 62.5 |
| 7 | 605 | 7.1 | 9.3 | 18.0 | 1.9 | 9.8 | 12.0 | 78.2 |
| 8 | 1167 | 7.5 | 15.8 | 44.5 | 4.3 | 20.5 | 27.5 | 51.9 |
| Paddy 1 | 792 | 5.9 | 9.3 | 9.6 | 0.9 | 21.5 | 26.3 | 52.2 |
| 2 | 1083 | 6.2 | 9.4 | 25.7 | 2.1 | 17.3 | 18.4 | 64.3 |
| 3 | 1963 | 6.8 | 6.3 | 13.2 | 1.1 | 5.8 | 7.1 | 87.1 |
| 4 | 1270 | 6.4 | 5.7 | 8.7 | 1.0 | 1.2 | 16.2 | 82.6 |
| 5 | 1200 | 6.2 | 7.0 | 14.4 | 1.3 | 2.9 | 24.1 | 72.9 |
| 6 | 1708 | 5.8 | 9.3 | 17.7 | 1.6 | 9.9 | 21.0 | 69.1 |
| 7 | 1842 | 6.0 | 7.7 | 16.4 | 1.6 | 9.0 | 18.6 | 72.5 |
| 8 | 1664 | 6.1 | 7.7 | 15.8 | 1.4 | 14.9 | 26.1 | 60.0 |
| Forest 1 | 114 | 5.5 | 15.7 | 77.7 | 4.1 | 11.0 | 41.7 | 47.2 |
| 2 | 229 | 5.0 | 15.8 | 77.6 | 3.8 | 43.1 | 24.2 | 32.7 |
| 3 | 690 | 5.1 | 12.7 | 80.5 | 4.9 | 6.5 | 27.8 | 65.7 |
| 4 | 5268 | 5.4 | 20.7 | 116.0 | 7.1 | 26.9 | 22.6 | 50.5 |
| 5 | 461 | 4.6 | 18.9 | 124.0 | 6.7 | 28.9 | 18.9 | 52.2 |

^a ¹³⁷Cs concentration in soil—Measuring date; Arable, 4 March 2014; Paddy, 28 July 2014; Forest, 20 October 2014.

^b Cation exchange capacity (CEC). ^c Total carbon (TC) content in soil. ^d Total nitrogen (TN) content in soil. ^e Clay, <0.002 mm; silt, 0.002–0.02 mm; sand, 0.02–2 mm.

3.2. ¹³⁷Cs Concentrations and Sequential Extraction

Cesium-137 activity concentrations were higher than 1.00 × 10³ Bq kg⁻¹ with few exceptions (Table 3). In arable soils, high activity concentrations of 3.02 × 10³ Bq kg⁻¹ and 2.34 × 10³ Bq kg⁻¹ were measured in soils from Arable 4 and Arable 5. Arable 3, Arable 6 and Arable 7 recorded activity concentrations less than 1000 Bq kg⁻¹, measuring 8.95 × 10², 5.69 × 10² and 6.05 × 10² Bq kg⁻¹, respectively. In paddy rice soils, ¹³⁷Cs activity exceeded 1000 Bq kg⁻¹ except for Paddy 1, which had a value of 7.92 × 10² Bq kg⁻¹ (Table 3). The highest ¹³⁷Cs activity in paddy rice soils was measured at Paddy 3 with a value of 1.96 × 10³ Bq kg⁻¹. Forest soils had the highest and lowest ¹³⁷Cs activity of 5.27 × 10³ Bq kg⁻¹ and 1.14 × 10² Bq kg⁻¹ at Forest 4 and Forest 1, respectively, from all measured points.

The results of sequential extraction showed that, in arable soils, both water-soluble and exchangeable fractions ranged from 1%–5% of the total ¹³⁷Cs extracted, while the organic matter-bound fraction ranged from 6%–20% for all sites (Figure 2a).

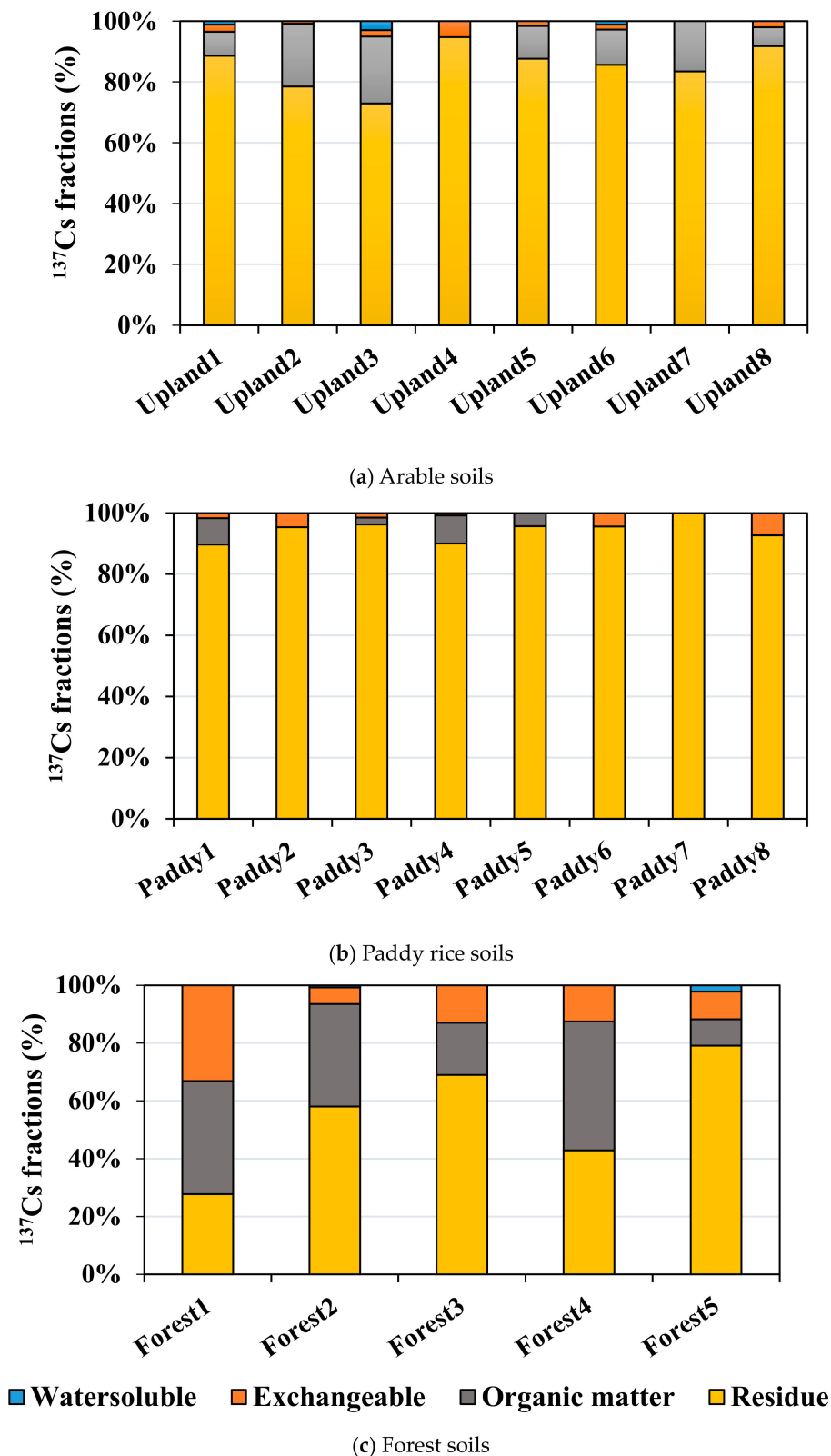


Figure 2. Proportions of soil ¹³⁷Cs fractions in (a) upland soils, (b) paddy rice soils and (c) forest soils.

For paddy rice soil, the water-soluble fraction was not detected as exchangeable and organic matter-bound fractions ranged from 1%–7% and 5%–10% respectively (Figure 2b). More than 90% residual fraction-extracted ¹³⁷Cs in both arable and paddy rice soils showed a stronger fixation

compared to the earlier ^{137}Cs sequential extraction study by Saito et al. [28] on soils taken from eastern Fukushima on 20 April 2011. The latter study reported no adsorption of ^{137}Cs on the water-soluble fraction but 14% in the ion exchangeable state, 4.7% in the oxide state, 5.4% in the organic matter fraction, 58% in the strongly fixed state and 17.5% in the residual fractions. The existing fraction of radioactive Cs when added to soil changes in the following order: water-soluble fraction, ion exchangeable and finally fixed fraction [29,30], with most radioactive Cs remaining strongly fixed to soil [15]. Previous research works, similar to this study, also showed a high fixation of ^{137}Cs to soil within two years after fall out [31–33] indicating progressive ^{137}Cs fixation with time, especially in the paddy rice soils (89%–97% residual fraction) as compared to in arable soils (72%–97%) (Figure 2a,b). In forest soils, the water-soluble fraction represented 0%–0.02% of total ^{137}Cs extracted with higher percentages of 5%–33% and 9%–44% representing exchangeable and organic matter-bound fractions, respectively, in contrast to those of both arable and paddy rice soils (Figure 2c). This shows a greater amount of easily mobile and bioavailable ^{137}Cs in forest soil than in the farmland soils. The residue, however, recorded lower fractions of 27%–79% in comparison to those of arable and paddy rice soils. Results from two independent preliminary surveys conducted six months after the accident by the Forestry and Forest Products Research Institute and Forestry Agency of Japan (FFPRI and FAJ) and by the Ministry of Education, Culture, Sports, Science and Technology (MEXT) indicated a significant transfer of radiocesium from trees to the forest floor via litterfall and rain and further transfer to mineral soils (0–5 cm) via organic matter decomposition, within a one-year interval (August–September 2011 and 2012) of the survey. In this study, forest soils were sampled in 2012, which implies that this was the stage of radioactive ^{137}Cs transfer, or immediate transfer, from the forest floor and the sedimentation of organic matter to the soil. Thus, a lower amount of ^{137}Cs was bound to soil minerals and a higher proportion was bound to the mobile fractions (exchangeable and organic matter) in the forest soils.

In further discussions, the influence of soil characteristics and land use on the chemical fraction of ^{137}Cs will be analyzed.

3.3. Correlation of ^{137}Cs Fractions and Soil Characteristics

Results of correlation analysis among ^{137}Cs fractions showed a significant negative correlation between the organic matter fraction and the residual fraction for both arable ($r^2 = 0.97$, $p < 0.05$) and paddy rice soils ($r^2 = 0.96$, $p < 0.05$) (Table 4).

Table 4. Pearson correlation coefficients among ^{137}Cs fractions.

| | | E | O | R |
|--------|---|-------|-------|----------|
| Arable | W | 0.02 | 0.38 | −0.50 |
| | E | | −0.46 | 0.54 |
| | O | | | −0.97 * |
| Paddy | W | NA | NA | NA |
| | E | | −0.49 | −0.18 |
| | O | | | −0.96 * |
| Forest | W | −0.45 | −0.69 | 0.69 |
| | E | | 0.34 | −0.75 |
| | O | | | −0.88 ** |

W represents water-soluble fraction. E represents exchangeable fraction. O represents organic matter fraction. R represents residual fraction. *, ** stand for significant correlation at $p < 0.05$ and $p < 0.1$ respectively; NA means non applicable, as water-soluble fraction was not detected.

The negative correlation is evident as the lower amounts of organic-bound ^{137}Cs were mainly due to greater fixation over time. This was implied to be an inhibitory effect of the residual fraction on the organic-bound ^{137}Cs fraction and vice-versa. Larger proportions of ^{137}Cs are known to be associated with clay fractions, and clay adsorption largely accounts for ^{137}Cs retention in soils [6]. The retention of ^{137}Cs can increase with increasing clay content as shown by [7] who reported 69%–93% of ^{137}Cs to

be found in the clay fraction of soils with a clay content of 24%–27%. Another suggested implication was the adsorption of ^{137}Cs from the organic fraction into the residual fraction over time. Similarly, a significant negative correlation between organic matter and the residual fraction was observed in forest soil ($r^2 = -0.88$, $p < 0.1$). A possible explanation for this relationship can be attributed to the inhibitory effect of organic matter bound to clay surfaces by limiting access to adsorption sites [34,35] which indirectly declined ^{137}Cs adsorption in the organic-rich forest soils (77–124 g kg^{-1} TC) (Table 3). As organic matter has weak selectivity for ^{137}Cs , it may lead to more easily mobile and bioavailable fractions compared to agricultural soils.

3.4. Influence of Soil Characteristics on ^{137}Cs Availability

A significant positive correlation was obtained between the exchangeable fraction and CEC ($r^2 = 0.76$, $p < 0.05$), TC ($r^2 = 0.80$, $p < 0.05$) and TN ($r^2 = 0.78$, $p < 0.05$) values in arable soils ($r^2 = p < 0.05$) (Table 5). Cs is present as a monovalent cation in soil and is retained at a cation exchange site whose capacity is expressed as CEC.

Table 5. Pearson correlation coefficients between ^{137}Cs fractions and soil chemical fractions.

| | | pH | CEC | TC | TN | Clay | Silt | Sand | TC/Clay | TN/Clay |
|--------|---|---------|--------|---------|---------|-------|--------|-------|---------|---------|
| Arable | W | −0.55 | −0.38 | −0.45 | −0.44 | 0.42 | 0.17 | −0.35 | −0.62 | −0.61 |
| | E | 0.23 | 0.76 * | 0.80 * | 0.78 * | 0.42 | 0.49 | −0.50 | 0.44 | 0.39 |
| | O | −0.30 | −0.74 | −0.61 | −0.59 | −0.41 | −0.13 | 0.33 | −0.15 | −0.13 |
| | R | 0.58 | 0.84 * | 0.73 * | 0.72 * | 0.28 | 0.27 | −0.31 | 0.50 | 0.48 |
| Paddy | W | NA | NA | NA | NA | NA | NA | NA | NA | NA |
| | E | −0.25 | 0.48 | 0.43 | 0.49 | 0.53 | 0.30 | −0.49 | −0.49 | −0.51 |
| | O | 0.14 | −0.35 | −0.83 * | −0.80 * | −0.14 | 0.10 | 0.03 | 0.55 | 0.51 |
| | R | 0.03 | 0.04 | 0.62 | 0.54 | −0.24 | −0.33 | 0.33 | −0.26 | −0.19 |
| Forest | W | −0.91 * | 0.33 | 0.57 | 0.34 | 0.48 | −0.60 | −0.15 | −0.47 | −0.46 |
| | E | 0.67 | −0.17 | −0.34 | −0.30 | −0.66 | 0.92 * | 0.13 | 0.33 | 0.42 |
| | O | 0.84 ** | 0.25 | −0.27 | −0.21 | 0.15 | 0.41 | −0.49 | −0.29 | −0.28 |
| | R | −0.92 * | −0.11 | 0.35 | 0.30 | 0.21 | −0.75 | 0.30 | 0.07 | 0.01 |

W represents water-soluble fraction. E represents exchangeable fraction. O represents organic matter fraction. R represents residual fraction. CEC is the cation exchange capacity. TC and TN represent the total carbon and total nitrogen respectively. *, ** stand for significant correlation at $p < 0.05$ and $p < 0.1$ respectively; NA means not applicable, as a water-soluble fraction was not detected.

A similar positive relationship was obtained by Matsunaga et al. [36], extracting water-soluble and exchangeable fractions in the same manner as in this study. In contrast, studies by Puhakainen et al. [27] and Koarashi et al. [8] showed no significant correlation of CEC with the exchangeable fraction and Cs retention respectively. CEC was therefore not considered to be a good measure for ^{137}Cs retention in soil. In addition, a significant positive correlation was obtained between ^{137}Cs in residue and CEC ($r^2 = 0.84$, $p < 0.05$), TC ($r^2 = 0.73$, $p < 0.05$) and TN ($r^2 = 0.72$, $p < 0.05$) values. Organic matter, as with clay minerals, is negatively charged, has high CEC and is capable of ^{137}Cs adsorption [8]. In the arable soils, the organic matter amounts and CEC values showed a proportionally high relationship, and it could be implied that the organic matter abundance influenced CEC, which further influenced the available exchangeable fraction and the strongly fixed residual fraction (Table 5). This notwithstanding, it could not be established clearly whether organic matter played a direct role in ^{137}Cs fixation or retention and mobility in arable soils.

In paddy rice soil, a significant negative correlation was obtained between organic matter fraction and TC ($r^2 = -0.83$, $p < 0.05$) as well as TN ($r^2 = -0.80$, $p < 0.05$) values. Previous studies have shown the soil removal in rice paddy fields in Fukushima to be a decontamination practice [37,38], and it has been suggested that the negative relationship was due to slower organic matter decomposition, most of which resulted from uncontaminated older residues after the decontamination practice.

A significant negative correlation was obtained between both water-soluble ($r^2 = -0.91, p < 0.05$) and residual fractions ($r^2 = -0.92, p < 0.05$). The fixation capacity of radioactive Cs is stronger in alkaline soil than in acidic soil. For example, the water-soluble fraction was increased at low pH, and the clay-fixed fraction was increased at high pH value [39]. In contrast to this study, as little as 0.02% (Figure 2c) water-soluble fraction of the extracted ^{137}Cs was found for the acidic forest soil (Table 3). Such a contrasting relationship was also observed in the residual fraction, where acidity was seen to promote ^{137}Cs fixation. A significant positive correlation between pH and organic matter fraction ($r^2 = 0.84, p < 0.1$) was observed. $^{137}\text{Cs}^+$ is adsorbed to the negatively charged carboxyl group of humus and forms a stable complex [15]. The negative charges increase with high pH values, and $^{137}\text{Cs}^+$ is more likely to be adsorbed to the carboxyl group. The relationship implies that with high pH, the organic matter fraction is increased. Nevertheless, the pH of forest soils was not high enough to promote ^{137}Cs adsorption onto organic matter in this study. A possible explanation could be the high amount of organic matter content. A positive correlation (however not significant) between CEC and TC and TN values may have reflected the higher CEC values at high amounts of organic matter content (Table 3) and promoted the adsorption of ^{137}Cs into organic matter.

3.5. Influence of Land Use Types on ^{137}Cs Availability in Soils

The fixation of ^{137}Cs was greater in paddy rice soil than in arable soil (Figure 1). There are some reports that a cycle of drying and wetting is required to enhance ^{137}Cs fixation [40,41]. This condition was considered to be enhanced in the paddy rice soils due to repeated irrigation and drainage. In addition, organic matter inclusion by fertilization was suggested to have influenced the relationship between organic matter content and organic matter fraction in both arable and paddy rice soils, as sampling sites are used as agricultural production fields by farmers and as experimental fields by researchers.

In the forest ecosystem, ^{137}Cs reaches the soil by a litterfall process (in addition to air deposition by throughfall) and serves as a temporal storage for large ^{137}Cs accumulation. Koarashi et al. [8] reported that 50%–90% of radioactive Cs accumulates in the litter layer of the forest floor. As a dynamic component of the forest ecosystem, forest litter generally undergoes microbial decomposition within a few years [42,43] and may become available for uptake by trees and microorganisms. The bioavailable ^{137}Cs can form complex compounds with soluble organic matter produced by litter decomposition and transferred from the litter layer to soil mineral layer by downward seepage [44]. Such phenomena of uptake by trees and microorganisms as well as ^{137}Cs -organic matter complexation are possible reasons for a low water-soluble fraction and a much increased organic matter fraction in the forest soils. In Japan, similar to as shown this study, most of the fallout ^{137}Cs in the 1960s remained in the shallow surface area even several decades after the main deposition in undisturbed forest [45]. Due to the typically high amounts of organic matter in the surface layer of forest, ^{137}Cs fixation in the mineral soil layer may be reduced [46]. Nonetheless, gradual oxidation and biodegradation can liberate ^{137}Cs adsorption to organic matter with time and thus further increase the available and easily extractable fractions, such as water-soluble and ion exchangeable fractions [32], which may not only be taken up by microorganisms and vegetation but also adsorbed into the mineral soil layer in forest soils (Table 5).

4. Conclusions

Results from the sequential extraction showed strong ^{137}Cs fixation in agricultural soils—in particular, paddy rice soils—in comparison to forest soils. Thus, the adverse effect on ^{137}Cs mobility and agricultural production is the gradual decrease of bioavailable ^{137}Cs for plant uptake. In contrast, the higher proportions of exchangeable and organic matter fractions observed in forest soils suggest future mobility and bioavailability of ^{137}Cs in forest ecosystems. A significant negative relationship ($p < 0.05$) between ^{137}Cs in the organic matter fraction and the residue in both arable and paddy rice soils suggest that the organic matter fraction inhibited ^{137}Cs fixation on soil minerals. In addition, CEC, TC, and TN showed positive relationships ($p < 0.05$) with both exchangeable fractions and

residue, which suggests that high organic matter content increases exchangeable ^{137}Cs fractions. Thus, although ^{137}Cs retention in the organic matter fraction could not be observed, it is inferred that a high organic matter content leads to higher ^{137}Cs mobility. In forest soils, pH showed a negative relationship ($p < 0.05$) with the water-soluble fraction and the residue but a positive relationship ($p < 0.1$) with the organic matter fraction. It is suggested that high pH promotes ^{137}Cs retention in soil organic matter, and therefore soil pH needs to be considered in all land use types.

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