THESIS

VERTICAL DISTRIBUTION OF RADIOCESIUM IN SOIL DEPOSITS ON THE CONTAMINATED AREAS AFTER THE FUKUSHIMA DAIICHI NUCLEAR POWER PLANT ACCIDENT

Submitted by

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ABSTRACT

VERTICAL DISTRIBUTION OF RADIOCESIUM IN SOILS DEPOSITS ON THE CONTAMINATED AREAS AFTER THE FUKUSHIMA DAIICHI NUCLEAR POWER PLANT ACCIDENT

An accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP) occurred on March 11, 2011 which resulted in an environmental contamination with the radiocesium species ^{134}Cs and ^{137}Cs. Vertical distribution of radiocesium is important as it impacts the area dose rate. The vertical distribution of radiocesium is sensitive to wash-off by surface runoff, wind resuspension, and soil to plant transfer. Soil core samples were extracted to develop soil profiles.

The purpose of this research is to study the vertical distribution of radiocesium in different soils contaminated after the accident, and to characterize the mechanisms by which the element moves through the soil. The results were compared to data on radiocesium vertical migration observed in Fukushima contaminated area for the year 2015 (Konoplev et al. 1992; Konoplev et al. 2016). The hypothesis is that reliable predictions of future soil contamination can be made based on the results from our soil samples. Predictions regarding radiocesium movement in soils will assist and improve remediation efforts in the Fukushima District. The vertical distribution of radiocesium was found to have a rate of movement of up to 12 cm/y in fluvisol type soils of Inkyozaka, 1 cm/y in andosol soils (Funasawa) and 3 cm/y in terrestrial regosol soils (Kashiramori). The results compared well with previous studies. Movement of radiocesium in Fukushima soils is most likely due to the high precipitation rate, combined with the weak bonding of cesium to fluvisol type soils.

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INTRODUCTION

The Fukushima Daiichi nuclear power plant station (FDNPP), located on the east coast of Honshu Island in Northeastern Japan, experienced a seismic event of 9.0 magnitude on 11 March 2011 that resulted in the release of various radionuclides into the atmosphere and consequently led to contamination of the soil surrounding the FDNPP and adjacent areas. The radioisotopes released included ^{132}Te, ^{131}I, ^{132}I, ^{133}I, ^{133}Xe, ^{131m}Te, which have decayed to below detection limits[8]. The radiocesium isotopes ^{134}Cs and ^{137}Cs are now responsible for the continued emission of radiation from the soil contamination. ^{134}Cs has a half-life of 2.07 years, and ^{137}Cs has a half-life of 30.17 years. The ratio of ^{134}Cs/^{137}Cs isotopes at the time of the accident was approximately 1[8]. The ratio has since changed because of the ^{134}Cs shorter half-life.

The observed mechanism of radiocesium soil retention is influenced by movement as dissolved cations by infiltration flow, fixation (sorption/desorption) to soil, ion exchange, bonding to organic matter, and bioturbation [1]. Cesium soil sorption is via a cation exchange process at varying levels of selectivity and reversibility depending on the exchange site [2,3]. Mobile radiocesium may be transported as solid particles by infiltration flow through pores, cracks, and cavities (bioturbation), and the rate is affected by particle size. Retention of Cs on soil particles increases as the particle size decreases [3]. Various cesium activity depth distribution studies have been carried out since the accident in Chernobyl to determine the mechanisms by which the cesium isotopes migrated in soil [1][4] [12]. Understanding the mechanisms by which cesium is distributed in Fukushima soils can help to improve remediation methods for the area. The study of radioecology investigates how different mechanisms affect a material's migration in the environment. A significant part of the decision-making process for

remediation, and part of the goal for radioecology, is predicting how the condition of contamination may advance. The data described in this study can contribute to the improvement of predictive simulation models.

The hypothesis is that reliable predictions of future soil contamination can be made based on the results from current soil samples. The prediction of cesium movement down the soil profile was found to be correct. The vertical distribution of radiocesium was estimated to have a rate of movement of up to 4 cm/y in fluvisol type soils of Inkyozaka, 1 cm/y in andosol soils (Funasawa), and 3 cm/y in terrestrial regosol soils (Kashiramori).

METHODS

2.1 Description of sampling sites

Honshu, the main island of Japan, is the largest island and the most populous. Geographically, Honshu accounts for about sixty percent of the total area of Japan and is about 1300 km in length with a width that ranges from 50 to 300 km. Honshu is characterized by a humid subtropical climate which has four distinct seasons. The rainy season occurs between mid-June to mid-July (samples were taken during this time) and the precipitation can vary from up to 1800 mm per year. The average precipitation expected for the area of this study is between 1200 and 1800 mm per year [14]. The average annual precipitation rate based on historical data from 1991-2015 is 1627.5 mm per year.

2.2 Soil collection and sampling procedure

Soil samples were extracted to a depth of 30 cm with the use of a hammer and a liner sampler DIK-110C (DAIKI, JAPAN: www.daiki.co.jp) containing a 5-cm diameter plastic cylinder insert, as pictured in Figure 1.

Figure 1: The soil core sampling equipment is used here to collect a virtually undisturbed soil core sample for environmental study.

Several soil cores representing the major soil types were collected in a 10-km zone of the FDNPP. The 10-km zone includes Okuma town. Details on the samples are provided in Table 1.

Table 1: Sample location and details (indicated sample coordinates which are unavailable)*

Sample Number	Date of Sample	Town where sample taken	North GPS Coordinates of sample location	East GPS Coordinates of sample location	Total sample Depth (cm)	Dose Rate, µSv/h
SPS-05	2016/06/09	Suzuuchi	37.41579722	140.9796972	29	16.4
SPS-06	2016/06/09	Suzuuchi	37.415775	140.9791139	29	15.4
SPS-07	2016/06/09	Suzuuchi	37.41588333	140.9800278	29	14.5
SPF-05	2016/06/22	Funasawa	*	*	28.5	4.5
SPF-06	2016/06/22	Funasawa	*	*	28.5	
SPF-07	2016/06/22	Funasawa	*	*	28	
SPK-05	2016/06/09	Kashiromori	*	*	29	1.1
SPK-06	2016/06/09	Kashiromori	*	*	29	
SPK-07	2016/06/09	Kashiromori	*	*	29	
SPI-05	2016/06/09	Inkyozaka	37.42510833	141.0171806	29.5	5.6
SPI-06	2016/06/09	Inkyozaka	37.42538056	141.017	28.5	6.0
SPI-07	2016/06/09	Inkyozaka	37.42598333	141.0174361	28.5	8.0

The extracted soil core samples were labeled, cut and placed into separated containers for radiocesium analysis (Figure 2 and Figure 3) to the depth increments summarized in Table 2.

Table 2: Thickness and number of sections, including depth increments, of each soil core slice based on location

Location	Section Depth Increments, in cm	Number of increments
Suzuuchi, Funasawa, Kashiromori and Inkyozaka	0-1, 1-2, 2-3, 3-5, 5-7, 7-10, 10-13, 13-17, 20-25, 25-30	10

Figure 2: Dr. A. Konoplev demonstrates how to prepare soil core samples.

Figure 3: In the above image are all the samples provided from one soil core extracted around Suzuuchi Pond.

It should be noted that depth increments in Table 2 were used as a guide, therefore, the increments on some cores deviate slightly from the size guide. The location where the soil cores were extracted was determined with a GPS model GARMIN Oregon 550TC. Additionally, the dose rates in the sampling areas were determined using a Gamma ray survey meter ALOKA pocket survey meter PDR-111. Soil cores were placed into containers after being separated at the specified depth increments and dried at 70° C for a minimum of 72 hours. The samples were ground with mortar and pestle tools to make a homogenized sample.

2.3 Radiocesium analysis of soil

Radiocesium concentrations of the soil samples were measured by gamma spectrometry using a high-purity germanium detector (HPGe), model CANBERRA GC3018 pictured in Figure 4.

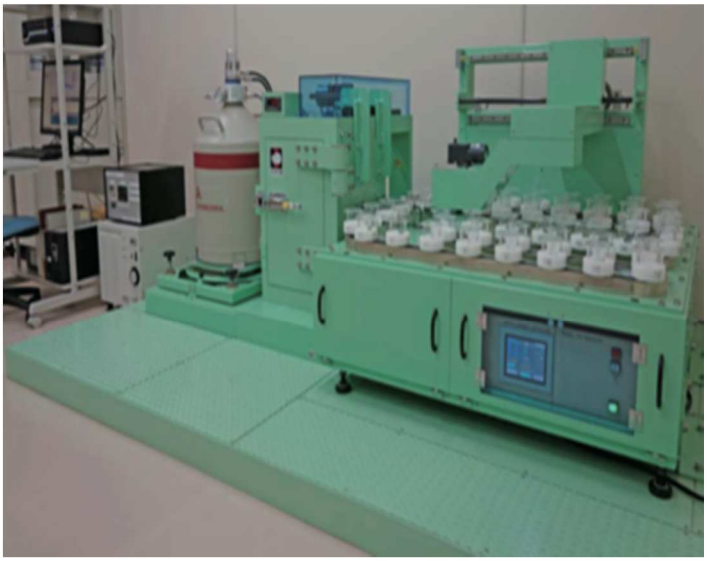


Figure 4: The high purity germanium detector (HPGe), model CANBERRA GC3018 located at the Institute of Environmental Radioactivity facility at Fukushima University

2.4 Sequential extraction of ^{134}Cs and ^{137}Cs

Sequential extraction analysis of the soil core samples was performed in three fractions, exchangeable, bound to organic matter, and nonexchangeable (residual).

2.4.1 Exchangeable Fraction

The top 10 centimeter depths of the first extracted soil cores from Suzuuchi, Inkyozaka, Funasawa, and Kashiromori were processed to determine the exchangeable fraction of radiocesium using sequential extraction. The depth increments processed were 0-2, 2-4, 4-6, 6-8, and 8-10 cm. Each soil increment was placed into separate beakers. Individual sections were passed through a 2-mm sieve to remove particles such as leaves, stones and roots to make a final homogenous sample. A solution of CH_3COOMH_4 and 3 g of the soil was prepared using a ratio of 1 part soil to 10 parts CH_3COOMH_4 (approximately 3 g of soil to 30 g of CH_3COOMH_4) and filtered into centrifuge tubes (Figure 5 and Figure 6).

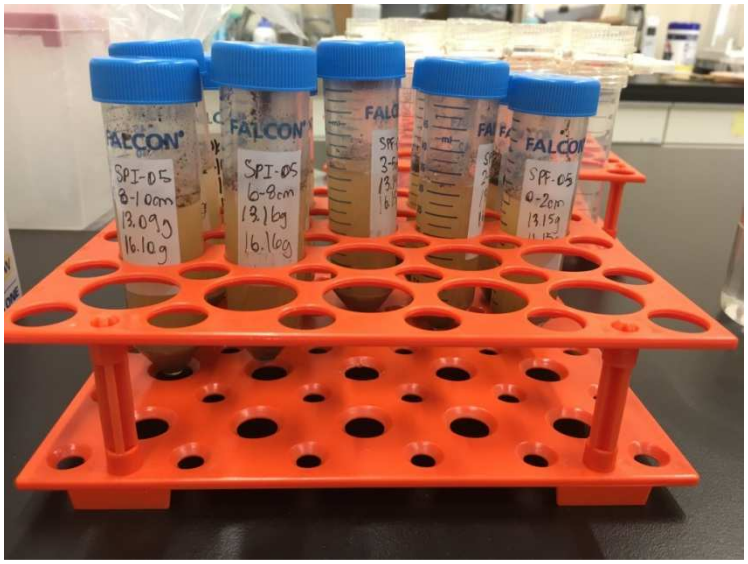


Figure 5: A collection of samples for the exchangeable fraction of sequential extraction. The samples were prepared at a ratio of 1 part soil to 10 part of the chemical (3 g of soil to 30 g of CH_3COOMH_4) and filtered into centrifuge tubes.

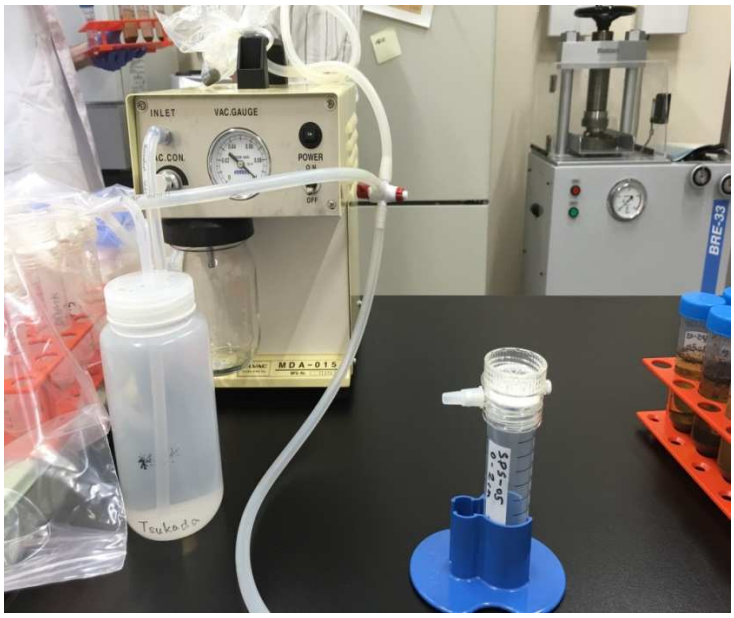


Figure 6: Filtration system for preparing the exchangeable fraction samples.

The centrifuge tubes were placed inside the centrifuge, Hitachi himac CT6E, assuring that the masses were balanced within the centrifuge. The samples, a total of 21 (including a blank), were spun for 1 hour in the centrifuge. The liquid samples were then analyzed for cesium content using the high-purity germanium detector (HPGe), model CANBERRA GC3018.

2.4.2 Bound by Organic Matter Fraction

The second fraction of sequential extraction analysis was performed using only the top soil layer, 0-2 cm, of the four sample sites (Suzuuchi, Funasawa, Kashiromori, and Inkyozaka). One mL of H_2O_2 (hydrogen peroxide) was dispensed into each of the four sample solutions (Figure 7) and left for >12 hours to allow oxidation reactions to complete.

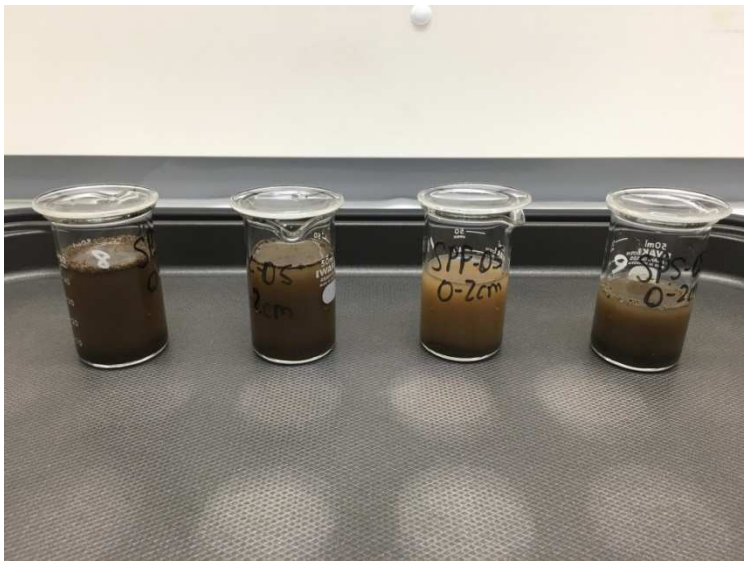


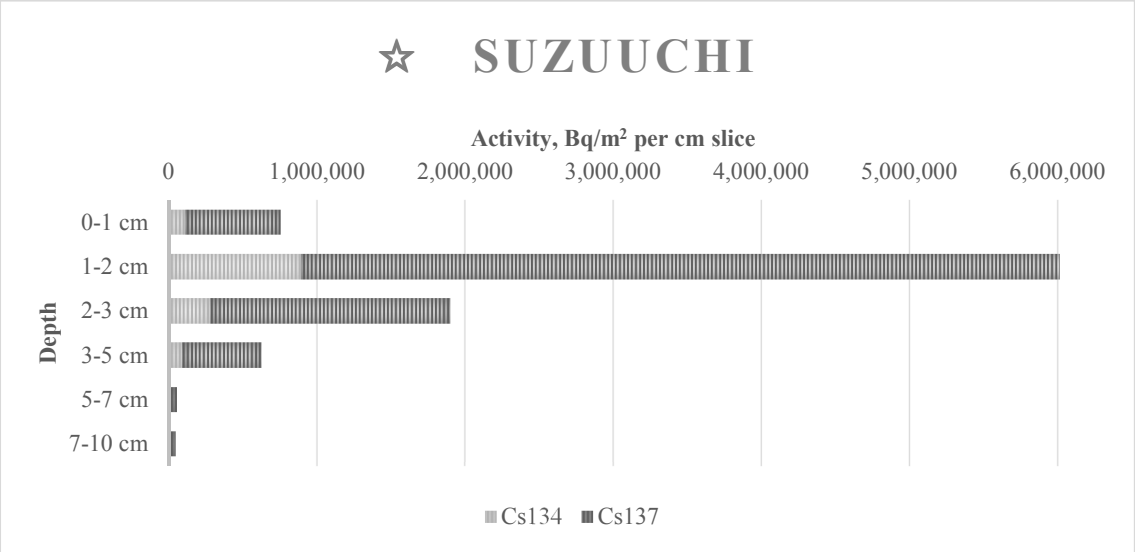
Figure 7: Samples of the Bound to Organic Matter fraction that were left to react over a 12 hour time period.

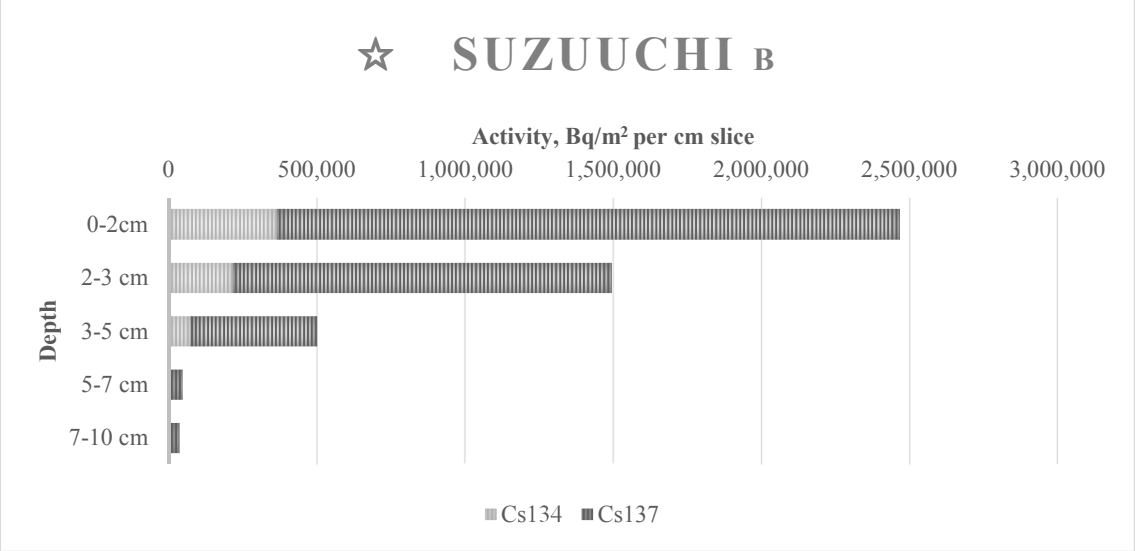
The samples were heated to 80°C and cooled to room temperature. The liquid samples were analyzed for cesium content using the high-purity germanium detector (HPGe), model CANBERRA GC3018.

RESULTS

3.1 Soil profile of radiocesium distribution

The soil activity concentration profile for all particles (original sampling) and particle size under 2 mm are denoted in Figure 8 for Suzuuchi and Funasawa. The isotopic ratio of the radiocesium species was found to vary between 0.17-0.18 (^{134}Cs to ^{137}Cs).





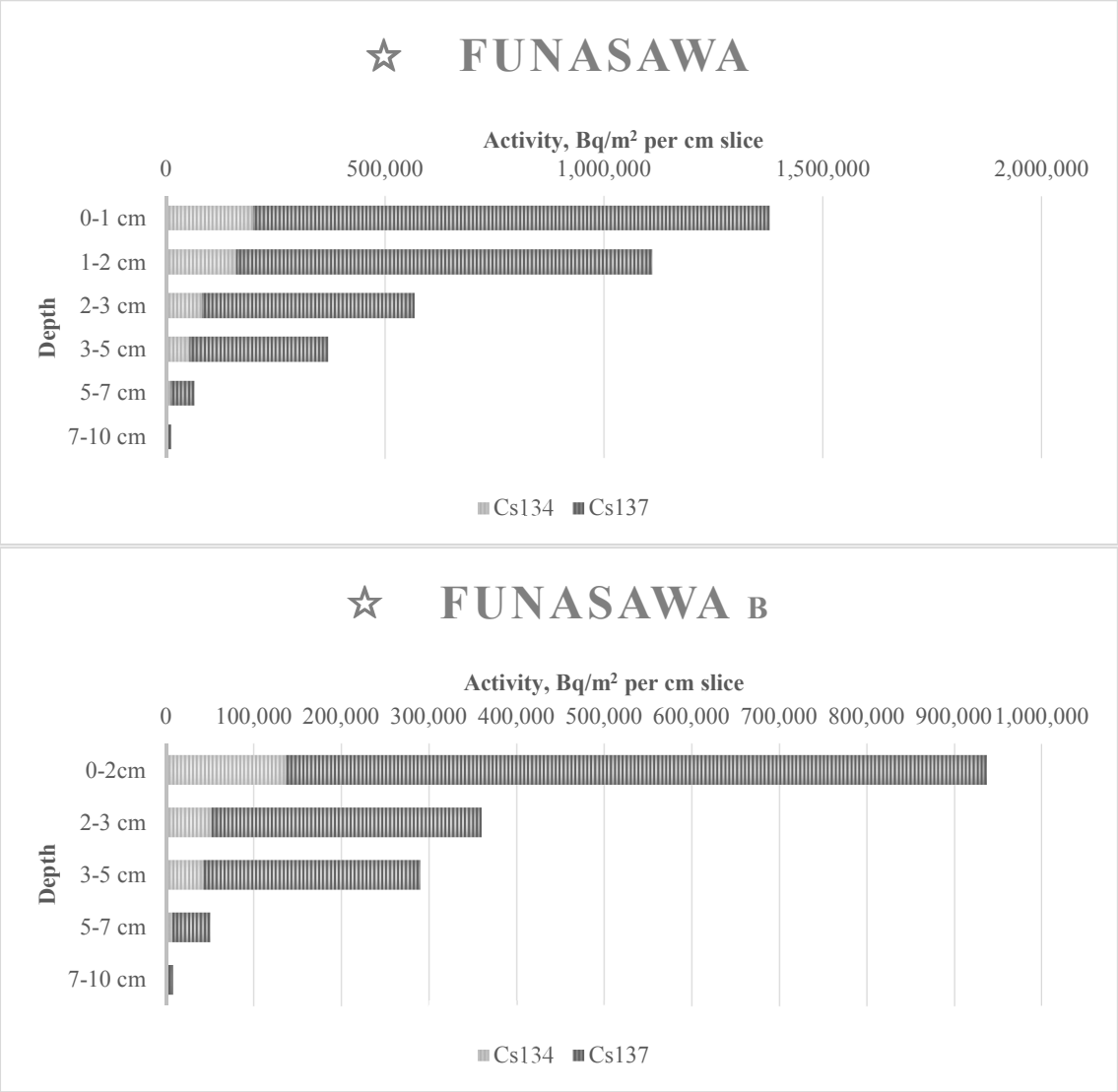


*Figure 8: Vertical distribution radiocesium soil profiles were provided for the samples taken around each of the four ponds. *B denotes the soil profile for a particle size under 2-mm.*

Sequential extraction and additional sieving were carried out on the first iterations of each set of soil cores extracted around the 4 ponds, that is, the soil core serial numbers which ended in "05" (Table 1). Only the first extraction of each sample set was used to compare all particle size to under 2 mm particle size in Figure 8. Soil data for all soil extractions around the four ponds are provided in the appendix. As seen in the soil profiles (Figure 8), there was a

steady decrease in activity as soil depth increased, and the profiles followed a natural log function (Figure 9).

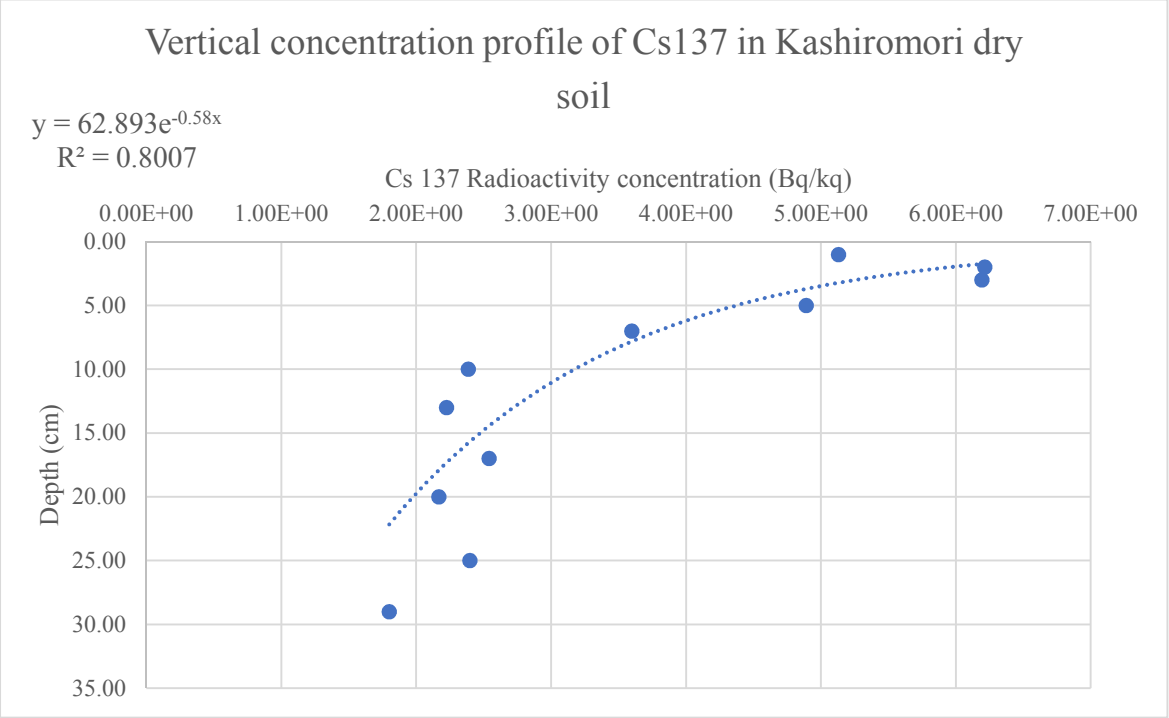


Figure 9: Vertical concentration of Cs 137 profile in dry soil around Kashiromori pond. Cesium 137 concentration (Bq/kq) was log transformed to provide a depiction of how the isotope was distributed in the soil surrounding Kashiromori pond.

The highest radiocesium content was found in the 0-2 cm layer across all ponds except for Inkyozaka where the highest concentration was observed at 2-4 cm. The highest concentration was not expected to be on the very top layer but within the first five centimeters, for fluvisol soils which contain a weak upper layer (see Table 3).

Table 3: Different soil types around the FDNPP

Site	Soil type	Distance from FDNPP, km	^{137}Cs depositon Bq/m^2
Suzuuchi	Fluvisol	3.75	6777
Inkyozaka	Fluvisol	0.24	2273
Kashiramori	Terrestrial Regosol	7	425
Funasawa	Andosol	3.5	3650

Soil types such as fluvisols and andosols lack a uniform upper layer and are characterized by the materials deposited from rivers and lakes. As seen in the Suzuuchi profile in Figure 8, the 1-2 cm layer had a higher activity concentration than the 0-1 cm layer. Nonetheless, the bulk of the activity concentration was observed in the top 10 cm of soil for all extractions across the four ponds.

3.2 Sequential Extraction

3.2.1 Fraction 1 – Exchangeable Fraction

The extracted quantity of cesium increased with depth. At the most contaminated site, Suzuuchi, the exchangeable fraction varied from 0.4-3.1% (Table 4). The less contaminated pond areas, Funasawa, Inkyozaka, and Kashiromori, had exchangeable fractions which varied from 3.0-9.2%, 3.6-23.2%, and 3.9-28.3% respectively (Table 4). Extracted proportions appeared to vary strongly with depth at the Inkyozaka and Kashiromori site, both achieved an extraction fraction of over 20% in the 7-10 cm layer (Table 4).

Table 4: Results of sequential extraction - exchangeable and bound to organic matter fraction.

Extracting solution					
Soil			Cs-137	Exchangeable fraction	Organic matter fraction
Date	ID	Depth layer	Bq	%	%
2016/6/9	SPS05	0-2cm	9.88	0.4	0.7
2016/6/9	SPS05	2-3 cm	8.26	1.2	
2016/6/9	SPS05	3-5 cm	2.62	1.4	
2016/6/9	SPS05	5-7 cm	0.40	2.7	
2016/6/9	SPS05	7-10 cm	0.45	3.1	
2016/6/9	SPF05	0-2cm	20.19	3.0	2.5
2016/6/9	SPF05	2-3 cm	9.98	3.3	
2016/6/9	SPF05	3-5 cm	5.54	3.7	
2016/6/9	SPF05	5-7 cm	1.64	9.2	
2016/6/9	SPF05	7-10 cm	0.40	9.0	

2016/6/9	SPI05	0-2 cm	10.79	3.6	2.6
2016/6/9	SPI05	2-4 cm	5.62	5.0	
2016/6/9	SPI05	4-6 cm	5.08	12.0	
2016/6/9	SPI05	6-8 cm	4.06	17.2	
2016/6/9	SPI05	8-10 cm	1.76	23.2	
2016/6/9	SPK05	0-2 cm	3.88	3.9	0.5
2016/6/9	SPK05	2-3 cm	5.16	12.2	
2016/6/9	SPK05	3-5 cm	2.41	13.2	
2016/6/9	SPK05	5-7 cm	0.59	19.7	
2016/6/9	SPK05	7-10 cm	0.17	28.3	

3.2.2 Fraction 2 – Bound to Organic Matter

The second fraction of sequential extraction was performed to find what fraction of radiocesium was bound to organic matter. The organic matter was destroyed to allow the release of cesium. A summary of both the exchangeable and organic fraction is illustrated in Figure 10.

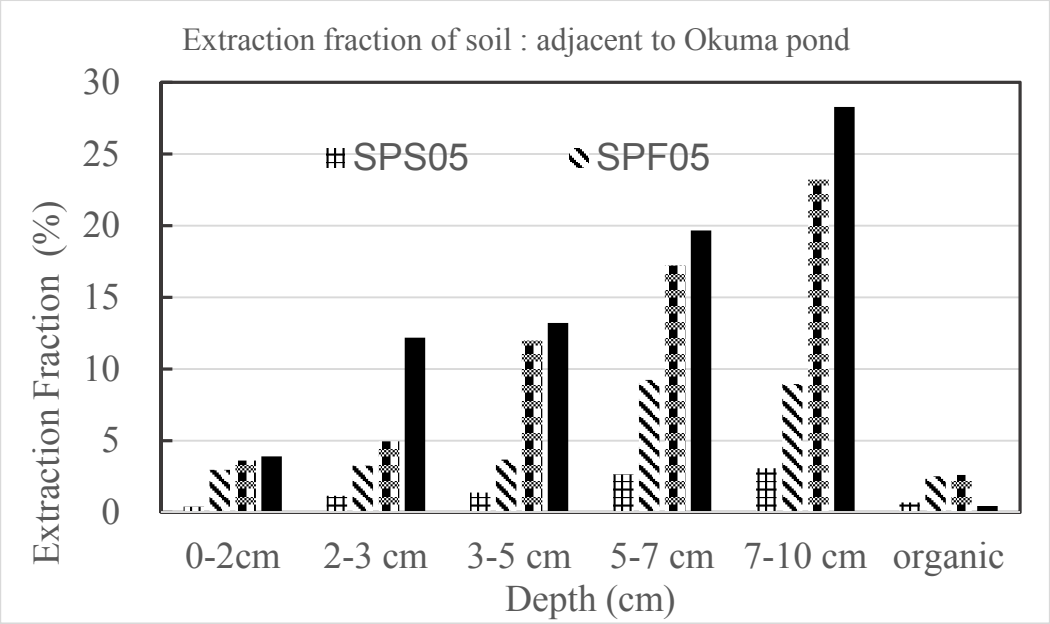


Figure 10: Summary of extraction fraction for sequential extraction

3.3 Comparison of radiocesium analyzed in soil cores for 2015

Fig. 11-14 provides a graphical representation of data from 2015 versus 2016 soil cores. The top soil layer appeared to have a lower contamination activity while the lower layers experienced an increase in activity. Around the Funasawa pond, there appeared to be an accumulation of contamination on the top soil layer, possibly washed in. Additionally, there was an increase in the lower layers. Inkyozaka experienced the same decrease in top soil layer activity that was seen in Suzuuchi, as well as in increase in the deeper layer activities. The 2016 Kashiromori soil cores showed a decrease in the activity of the top soil layer.

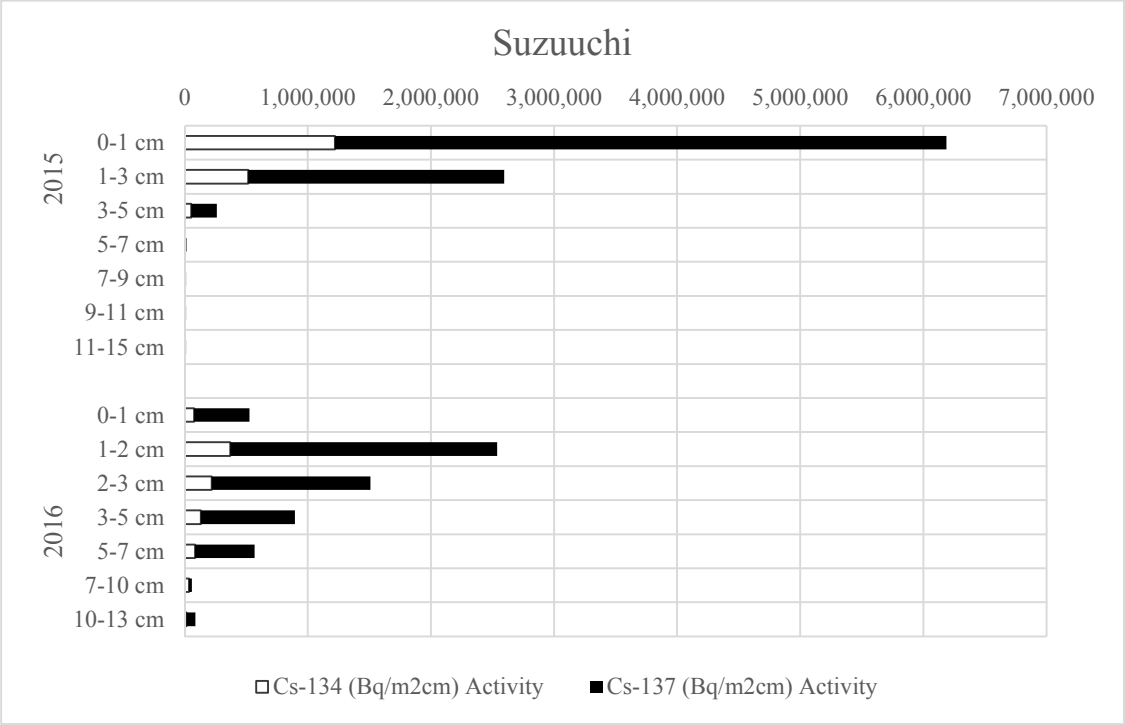


Figure 11: Comparison of soil data collected for the Suzuuchi site in 2015 and 2016 soil cores.

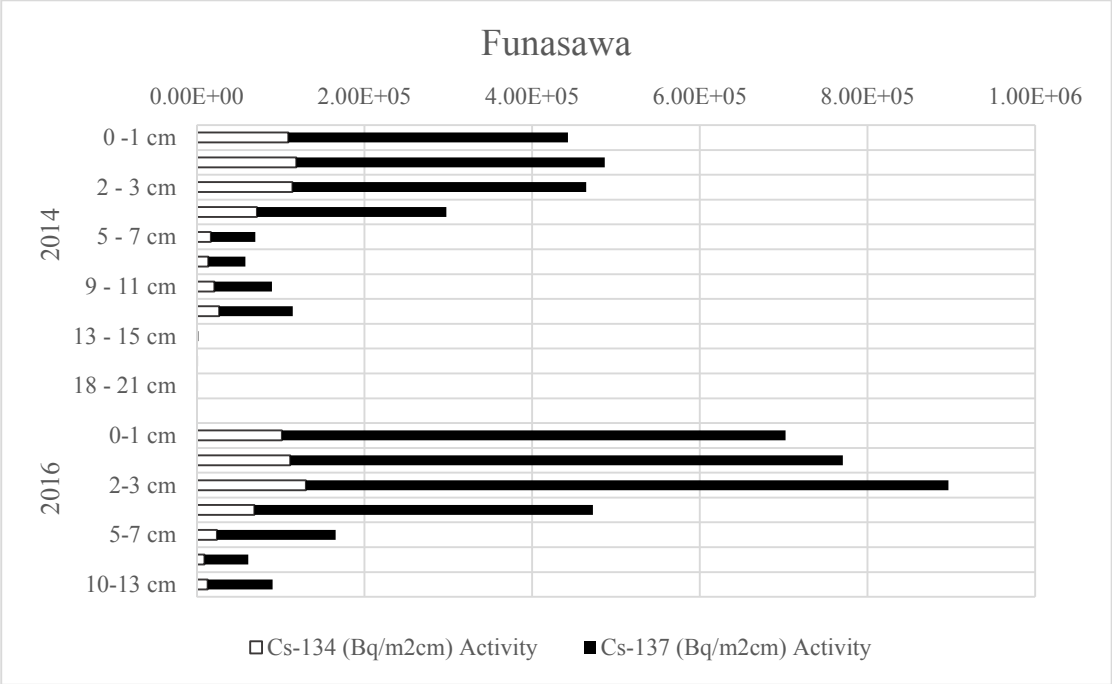


Figure 12: Comparison of soil data collected for the Funasawa site in 2014 and 2016 soil cores.

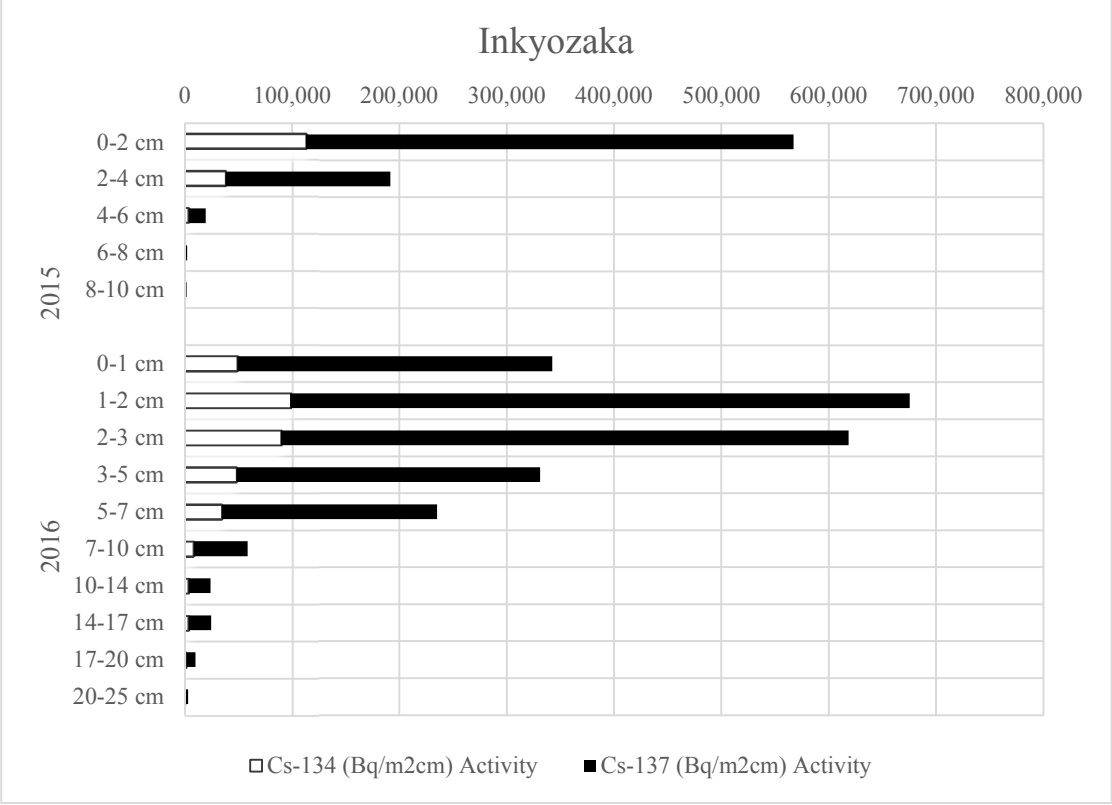


Figure 13: Comparison of soil data collected for the Inkyozaka site in 2015 and 2016 soil cores

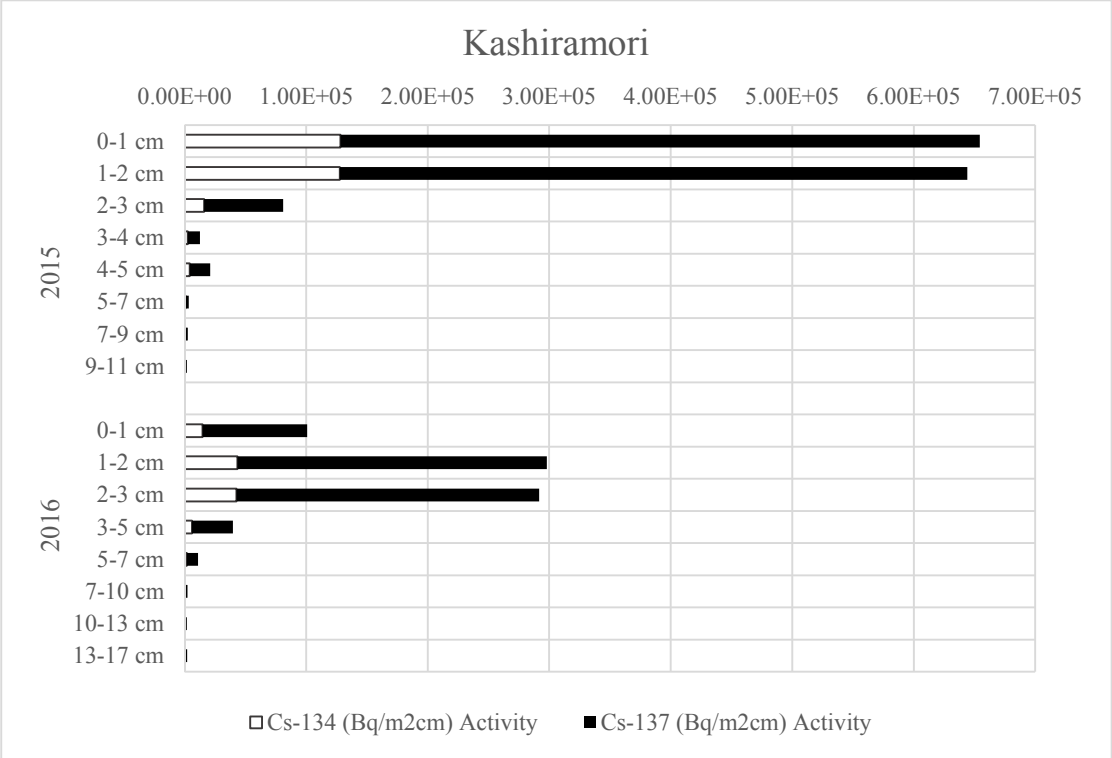


Figure 14: Comparison of soil data collected for the Kashiromori site in 2015 and 2016 soil cores.

Figures 11-14: *A collection of graphs that represent soil core data collected from the previous year compared to most recent sampling.*

Soil particle size under 2 mm for all samples were taken 2016.JUN.09 (Table 5). The ID for soil extractions around each pond are labeled SP(S,F,I,K), where SP is soil profile and the third letter corresponds to the first letter of the pond name. An ID was added to the serial number (05, 06, 07) to denote the three separate soil extractions.

Table 5: Summary of soil extraction for first samples of all pond areas.

Sample		Cs-134 (Bq/kg)		Cs-137 (Bq/kg)		Cs134/Cs137
ID	Depth	Activity	Uncertainty	Activity	Uncertainty	
SPS05	0-2cm	1.39E+05	3.91E+02	8.01E+05	1.41E+02	0.17
SPS05	2-3 cm	4.05E+04	1.50E+02	2.39E+05	5.43E+02	0.17
SPS05	3-5 cm	1.08E+04	5.06E+01	6.33E+04	1.83E+02	0.17
SPS05	5-7 cm	8.55E+02	8.79E+00	5.04E+03	3.11E+01	0.17
SPS05	7-10 cm	8.35E+02	7.98E+00	4.82E+03	2.81E+01	0.17
SPF05	0-2cm	3.86E+04	1.82E+02	2.26E+05	6.59E+02	0.17
SPF05	2-3 cm	1.71E+04	1.02E+02	1.02E+05	3.73E+02	0.17
SPF05	3-5 cm	8.59E+03	4.30E+01	5.01E+04	1.54E+02	0.17
SPF05	5-7 cm	1.03E+03	1.02E+01	5.93E+03	3.63E+01	0.17
SPF05	7-10 cm	2.46E+02	5.09E+00	1.49E+03	1.80E+01	0.17
SPI05	0-2 cm	1.70E+04	1.56E+02	9.94E+04	5.64E+02	0.17
SPI05	2-4 cm	6.38E+03	4.34E+01	3.77E+04	1.57E+02	0.17
SPI05	4-6 cm	2.38E+03	2.47E+01	1.41E+04	8.92E+01	0.17
SPI05	6-8 cm	1.37E+03	1.08E+01	7.85E+03	3.86E+01	0.17
SPI05	8-10 cm	4.47E+02	5.49E+01	2.53E+03	1.93E+01	0.18
SPK05	0-2 cm	5.77E+03	4.74E+01	3.32E+04	1.69E+02	0.17
SPK05	2-3 cm	2.39E+03	8.66E+01	1.41E+04	2.84E+02	0.17
SPK05	3-5 cm	1.04E+03	2.21E+01	6.07E+03	7.73E+01	0.17
SPK05	5-7 cm	1.80E+02	4.07E+00	9.92E+02	1.37E+01	0.18
SPK05	7-10 cm	3.69E+01	1.36E+00	2.05E+02	4.38E+00	0.18

DISCUSSION

4.1 Factors affecting ^{137}Cs distribution in soil layers

Radiocesium distribution in soil has been observed to be affected by bioturbation, fixation (sorption/desorption) to soil, ion exchange, and binding to organic matter. Studies of the vertical distribution of radiocesium in soils found that, in undisturbed forest and grassland soils, radiocesium dispersion is quicker in forest soils compared to grassland soils [1]. This observation might be due to, among other things, differences in density between forest soils and grassland soils. The Fukushima prefecture is over seventy percent forested. ^{137}Cs was observed to remain concentrated in the top ten centimeters of the Fukushima soil profile.

The retention of radiocesium is significantly influenced by cation exchange, and it is sorbed on the surface of clay particles [3]. Soils that are high in organic matter may inhibit the adsorption of radiocesium by clay content. Therefore, forest soils which are rich in organic matter are expected to have highly mobile radiocesium, as the specific adsorption on clay minerals is decreased [2]. The contamination in Fukushima appeared to be mobile based on the sequential extraction findings that there was no significant amount bound to organic matter. Additionally, the cesium appeared to be more easily exchanged by ion exchange mechanisms down the soil profile.

Radiocesium not immobilized in surface soil may be transported based in direct and indirect ways. The methods for travel include movement by infiltration flow, soil pores and cracks, as well as disturbance by wildlife. Precipitation rates are important to consider for soils of this matter. Precipitation that deposits on the ground may cause radiocesium to infiltrate the soil or move across the area as surface runoff. Mobile radiocesium and radiocesium adsorbed to

clay particles might be transported by the infiltration flow, as influenced by saturation from precipitation, moving through soil pores and cracks. The precipitation may have been the most important factor in this study for the movement of the particles. Another factor affecting the radiocesium distribution to consider is bioturbation, that is, disturbance of the soil deposits by plants and animals. Bioturbation is important in the case of the Fukushima prefecture considering a significant fraction of it is forested. A high cesium retention rate in the forested soils, if due to immobile cesium, may remain highly concentrated in the litter layer available for root uptake [2][3]. The results of the study suggest that the cesium migrated down the soil profile; therefore plants (and animal bioturbation) may not be an important factor in the movement of radiocesium (Figures 11-14).

Fluvisol are those soils characterized by periodically flooded surface waters such as river floodplains. This soil type has a weak uppermost layer containing materials which have been deposited from rivers and lakes. The content of organic carbon decreases with depth. Both Suzuuchi and Inkyozaka share the fluvisol soil type (Table 3). It is expected that the radiocesium will continue to migrate down the profile in these areas while the uppermost layer decreases in activity (Figures 11-14). Organic bounding down the soil profile may change, and thus alter the movement of radiocesium deeper into the soils, but insufficient time was available to test soil deeper than 0-2 cm. Although deeper layers were not examined to determine organic bonding, if organic bonding to radiocesium is similar at depths deeper than 0-2 cm, further migration into the soil is expected. Less than three percent of the material in the top layer was bound to organic matter. The organic carbon content of this soil type decreases with depth. It would not be expected that the cesium migration would be halted down the profile if this is truly the case.

Andosols are soils characterized by physical properties of clay that are dominated by allpohane and imogolite, they also have high accumulations of organic matter. Andosols have high organic matter content and low bulk densities. The Funasawa soil is characterized as Andosol. Since the area is dominated by clay properties, this might be a reason for the increase in top soil activity. The increase in activity may have been due to an influx of radioceisum that was not bound in nearby soils. The activity in this area might increase due to this factor but it is expected that the isotopic ratio inventory will continue to decline naturally due to radioactive decay.

Regosol, another of the soil groups as classified by the Food and Agriculture Organization (FAO) system, is characterized by shallow, medium to fine textured, unconsolidated soil or sediments. Additionally, the soil type lacks a significant soil horizon layer because of climatic conditions [13]. In regard to this soil type, Kashiromori is the site of interest. Kashiromori was the one site to show a decrease in the upper and lower soil layers. The activity at this site is expected to see further decline by downward migration or remediation efforts.

4.2 Relevance of air dose rates across sampling sites

Immediately following the substantial release of radiation and contamination of soils, evacuation orders were issued at a 3-km radius about the FDNPP. The government eventually extended the evacuation radius to 20 km. The area where annual cumulative dose was expected to reach 20 mSv per year was designated as “restricted”. As of 7 August 2013, evacuation orders have been lifted in areas where the cumulative dose is expected to be less than 20 mSv per year. In the event that evacuees decide to return home, it can prove to be advantageous to quantify the area dose rates across sampling sites. Low dose radiation is expected to be of little health risk to the public.

4.3 Comparison of soil profile results

The radiocesium activity down the soil profile showed a decrease with depth in the soils analyzed around each pond. As observed in previous soil studies, the ^{137}Cs remained highly concentrated in the top soil layers [1][3]. The highest activities were observed in the soil around Suzuuchi pond. Assuming the external dose rate is representative, this finding is consistent with the 15 μSv/h area readings, which were measured around Suzuuchi pond (Table 1). In contrast, the soil around Kashiromori pond was found to have the lowest radiocesium concentration, where the external dose rate was 1.1 μSv/h. Geographically, Kashiramori pond is located 7 km away from the FDNPP. Radiocesium inventory ratios were found to be in the range of 0.17-0.18.

4.4 Sequential extraction summary

The sequential extraction of twenty samples (SPS05, SPF05, SPI05, SPK05: 0-2, 2-3, 3-5, 5-7, and 7-10 cm) showed that 0.4-28.3% of ^{137}Cs was leached by 1M CH_3COONH_4. While the 28.3% may seem significant it is worth mentioning that the higher end of these extraction fractions were observed toward the bottom layers where there was not a high amount of cesium. In the top 10 cm of the soils for Suzuuchi and Funasawa, only minor fractions of cesium were readily available for exchange (0.4-9%). The soils around Inkyozaka and Kashiromori experienced higher amounts of cesium available for exchange deeper down the profile (12-28.3%). The top layers were low in bound cesium (3.6-3.9%) as with the two other sites, Suzuuchi and Funasawa. This suggests that the retention of adsorbed radiocesium in the top soil layer is not strongly affected by the ion-exchange process.

Small quantities of radiocesium were found to be bound by organic matter. Due to time and funding constraints, only the top soil layer, 0-2 cm, was analyzed for the second fraction of sequential extraction, bound by organic matter. Fluvisol soil types (Suzucchi and Inkyozaka)

contain organic matter content that decreases with depth. The Andosol soil type (Funasawa) is expected to have a high accumulation of organic matter. Despite the high organic matter of the Andosol, only trace amounts of cesium were found to be bound by organic matter at all sites (0.7-2.6%). The sequential extraction of the study suggests, in relation to exchangeable fraction and organic matter content, the cesium is not strongly retained in the top layers by the fluvisol or regisol soil types.

4.5 Comparison with 2015 Fukushima soil profile results

Results of 2016 data, when compared with those of 2015, show there has been a decrease in the top soil layer activity apart from the Funasawa site which saw an increase. This may be due to precipitation rate in Fukushima. Samples were taken June-August which is a time of year characterized by the monsoon climate. The average annual precipitation rate based on historical data from 1991-2015 is 1627.5 mm per year. It should be noted that there was a decrease in radiocesium concentration inventories in 2016, compared to 2015. Soil core data of 2015 had a widely varying inventory ratio of ^{134}Cs to ^{137}Cs of 0.20-0.33 while the 2016 soil core data varied from 0.17-0.18 for similar areas. This ratio decrease was determined to be due to decay using the equation :

$$A = A_0 e^{-\lambda t}$$

where A= the final activity
A_0= the intial activity
λ = the decay constant
t=time elapsed

given that in the 2015 soil core data (SPK-3, 0-1 cm) ^{137}Cs and ^{134}Cs activities were 5.26×10^5 Bq/m^{2}cm and 1.28×10^5 Bq/m^{2}cm respectively. The time between sampling was approximately 1.13y.

$$A(\mathrm{Cs}-137) = 5.26\times10^{5}\,\frac{\mathrm{Bq}}{\mathrm{m^2cm}}\cdot\ e^{-(0.0231\ \mathrm{y^{-1}})(1.13\ \mathrm{y})}$$

$$A(\mathrm{Cs}-134) = 1.28\times10^{5}\,\frac{Bq}{m^2cm}\cdot\ e^{-(0.3364\ \mathrm{y^{-1}})(1.13\ \mathrm{y})}$$

$$=\frac{A(\mathrm{Cs}-134)}{A(\mathrm{Cs}-137)}=\frac{\left(\frac{8.75\times10^{4}\mathrm{Bq}}{\mathrm{m^2cm}}\right)}{\left(\frac{5.12\times10^{5}\mathrm{Bq}}{\mathrm{m^2cm}}\right)}=0.17$$

Using another example, 2015 soil core data for SPI-3 (0-2cm) gave ^{137}Cs and ^{134}Cs activities of 4.54×10^5 Bq/m^{2}cm and 1.13×10^5 Bq/m^{2}cm respectively. The time between sampling was approximately 1.13y.

$$A(\mathrm{Cs}-137) = 4.54\times10^{5}\,\frac{\mathrm{Bq}}{\mathrm{m^2cm}}\cdot\ \mathrm{e}^{-(0.0231\ \mathrm{y^{-1}})(1.13\ \mathrm{y})}$$

$$A(\mathrm{Cs}-134) = 1.13\times10^{5}\,\frac{\mathrm{Bq}}{\mathrm{m^2cm}}\cdot\ \mathrm{e}^{-(0.3364\ \mathrm{y^{-1}})(1.13\ \mathrm{y})}$$

$$=\frac{A(\mathrm{Cs}-134)}{A(\mathrm{Cs}-137)}=\frac{\left(\frac{7.73\times10^{4}\mathrm{Bq}}{\mathrm{m^2cm}}\right)}{\left(\frac{4.42\times10^{5}\mathrm{Bq}}{\mathrm{m^2cm}}\right)}=0.17$$

For the remaining sites where the highest concentration was not observed in the top layer, an increased activity was measured down the profiles. These findings compare well with previous studies as it has been observed that the cesium species, while strongly retained in the top soil layer (litter layer), can penetrate the profile deeper once they have reached the soil [2].

4.6 Implication of results

It can be expected that radiocesium will continue to migrate down the soil profile. Comparatively, 2015 versus 2016 soil cores, a travel of 4 cm/y (Inkyozaka), 3 cm/y (Kashiramori) and about 1 cm/y (Funasawa, Suzuuchi) can be estimated. These estimates were determined by observing significant quantities of radiocesium measured down the profile in the 2016 soil cores that were not present during 2015. For example, in the Inkyozaka soil cores,

activities on the order of 10^4 were measured only up to the 4-6 cm layer in 2015. In the following year, activities of the same magnitude were measured in the 14-18 cm layer down the soil profile. After a year, although the activity down the profile has increased, most of it is still contained in the top ten centimeters. Furthermore, while more accumulation of activity can be expected at the Funasawa site, which may be due to surface runoff, the inventory will continue to decline by decay. It is difficult to say how much could accumulate annually due to runoff.

CONCLUSION

The soil depth profile showed an overall decrease in relation to depth in the soils analyzed around four different pond sites. Results of 2016 data, when compared with those of 2015, show that the deeper layers of the soil profile are experiencing an increase in activity, while the top layers have declined, with the exception of Funasawa. The movement of radiocesium to deeper layers may be due to the precipitation rate in Fukushima, because sequential extraction showed that cesium is not strongly adsorbed in the exchangeable or bound by organic matter fractions in fluvisol and regosol soils found in Suzuuchi, Inkyozaka, and Kashiromori. Exchangeability was found to be very low in the top soil layers which suggest that mobility and bioavailability should be low. Additionally, the exchangeable fraction appeared to move along the soil profile, because it was more easily exchanged by ion exchange mechanisms down the soil profile.

The lowered isotopic ratio is an indication that, as expected, the ^{134}Cs is decaying much faster than ^{137}Cs due to its considerably shorter half-life. Earlier Fukushima soil studies found that the ^{137}Cs remained highly concentrated in the top soil layers [1][3], such was the case in the study as well. Additionally, it was observed that the cesium species, despite their strong retention in the top soil layer, can penetrate deeper into the soil with time.

There was insufficient time to perform the second fraction of sequential extraction to determine the quantity of radiocesium bound to organic matter on the remaining soil samples. Only the top soil layers 0-2 cm was used to determine this fraction. It would have been beneficial to see if this finding held true throughout the entire soil profile. It is recommended that future work determines the soil percentage of clay and sand particles to understand the retention

of radiocesium. It would also be beneficial to perform different methods of sequential extraction to better understand the radiocesium mobility mechanisms in the area, because the bound to organic matter and exchangeable fraction did not appear to be important modes of retention. This study found that while the deeper layers of the soil profiles have increased in activity, there was a decrease in the top soil layers. In regards to remediation, the data described in this study can contribute to the improvement of predictive simulation models. A simulation model can provide a prediction of the contaminant distribution and allow for the design of improved monitoring plans and cleanup arrangements.

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APPENDIX

Appendix 1: Raw data of soil cores by site

Suzuuchi

ID				Cs-137 (Bq/kg) Activity
SPS05	Core Soil	0-1 cm	0.5	3.83E+05
SPS05	Core Soil	1-2 cm	1.5	5.17E+05
SPS05	Core Soil	2-3 cm	2.5	2.02E+05
SPS05	Core Soil	3-5 cm	4	4.84E+04
SPS05	Core Soil	5-7 cm	6	3.37E+03
SPS05	Core Soil	7-10 cm	8.5	3.64E+03
SPS05	Core Soil	10-13 cm	11.5	5.35E+02
SPS05	Core Soil	13-17 cm	15	4.66E+02
SPS05	Core Soil	17-20 cm	18.5	1.82E+02
SPS05	Core Soil	20-25 cm	22.5	1.45E+02
SPS05	Core Soil	25-29 cm	27	3.00E+02

ID				Cs-137 (Bq/kg) Activity
SPS06	Core Soil	0-1 cm	0.5	3.62E+05
SPS06	Core Soil	1-2 cm	1.5	1.51E+05
SPS06	Core Soil	2-3 cm	2.5	5.37E+04
SPS06	Core Soil	3-4 cm	3.5	4.23E+04
SPS06	Core Soil	4-5 cm	4.5	6.61E+04

SPS06	Core Soil	5-7 cm	6	1.69E+04
SPS06	Core Soil	7-10 cm	8.5	7.41E+03
SPS06	Core Soil	10-15 cm	12.5	4.97E+02
SPS06	Core Soil	10-15 cm-2	12.5	4.75E+02
SPS06	Core Soil	15-20 cm	17.5	3.17E+02
SPS06	Core Soil	15-20 cm-2	17.5	2.86E+02
SPS06	Core Soil	20-25 cm	22.5	2.36E+02
SPS06	Core Soil	25-29 cm	27	4.62E+02

ID				Cs-137 (Bq/kg) Activity
SPS07	Core Soil	0-1 cm	0.5	5.67E+04
SPS07	Core Soil	1-2 cm	1.5	3.34E+05
SPS07	Core Soil	2-3 cm	2.5	1.02E+06
SPS07	Core Soil	3-5 cm	4	8.07E+05
SPS07	Core Soil	5-7 cm	6	9.73E+04
SPS07	Core Soil	7-10 cm	8.5	7.12E+04
SPS07	Core Soil	10-13 cm	11.5	1.49E+04
SPS07	Core Soil	13-17 cm	15	3.60E+03
SPS07	Core Soil	17-20 cm	18.5	2.74E+03
SPS07	Core Soil	20-25 cm	22.5	1.33E+03
SPS07	Core Soil	25-29 cm	27	7.83E+02

Funasawa

ID				Cs-137 (Bq/kg) Activity
SPF05	Core Soil	0-1 cm	0.5	2.38E+05
SPF05	Core Soil	1-2 cm	1.5	1.62E+05
SPF05	Core Soil	2-3 cm	2.5	7.40E+04
SPF05	Core Soil	3-5 cm	4	3.55E+04
SPF05	Core Soil	5-7 cm	6	5.45E+03
SPF05	Core Soil	7-10 cm	8.5	1.21E+03
SPF05	Core Soil	10-13 cm	11.5	7.36E+02
SPF05	Core Soil	13-17 cm	15	4.38E+02
SPF05	Core Soil	17-20 cm	18.5	3.19E+02
SPF05	Core Soil	20-25 cm	22.5	1.43E+02
SPF05	Core Soil	25-28.5 cm	26.7	1.87E+02

ID				Cs-137 (Bq/kg) Activity
SPF06	Core Soil	0-1 cm	0.5	1.15E+05
SPF06	Core Soil	1-2 cm	1.5	1.30E+05
SPF06	Core Soil	2-3 cm	2.5	1.09E+05
SPF06	Core Soil	3-5 cm	4	4.04E+04
SPF06	Core Soil	5-7 cm	6	3.25E+03

SPF06	Core Soil	7-10 cm	8.5	3.56E+03
SPF06	Core Soil	10-13 cm	11.5	6.42E+02
SPF06	Core Soil	13-17 cm	15	3.32E+01
SPF06	Core Soil	17-20 cm	18.5	4.03E+01
SPF06	Core Soil	20-25 cm	22.5	4.90E+01
SPF06	Core Soil	25-28.5 cm	26.8	1.10E+02

ID				Cs-137 (Bq/kg) Activity
SPF07	Core Soil	0-1 cm	0.5	5.47E+04
SPF07	Core Soil	1-2 cm	1.5	6.50E+04
SPF07	Core Soil	2-3 cm	2.5	6.81E+04
SPF07	Core Soil	3-5 cm	4	5.46E+04
SPF07	Core Soil	5-7 cm	6	4.22E+04
SPF07	Core Soil	7-10 cm	8.5	1.20E+04
SPF07	Core Soil	10-13 cm	11.5	2.47E+04
SPF07	Core Soil	13-17 cm	15	5.24E+03
SPF07	Core Soil	17-20 cm	18.5	2.65E+03
SPF07	Core Soil	20-25 cm	22.5	1.09E+03
SPF07	Core Soil	25-28 cm	26.5	1.68E+03

Inkyozaka

ID				Cs-137 (Bq/kg) Activity
SPI05	Core Soil	0-2 cm	1	1.05E+05
SPI05	Core Soil	2-4 cm	3	3.85E+04
SPI05	Core Soil	4-6 cm	5	1.31E+04
SPI05	Core Soil	6-8 cm	7	8.17E+03
SPI05	Core Soil	8-10 cm	9	2.63E+03
SPI05	Core Soil	10-14 cm	12	3.05E+03
SPI05	Core Soil	14-18 cm	16	2.05E+03
SPI05	Core Soil	18-22 cm	20	3.09E+02
SPI05	Core Soil	22-26 cm	24	5.00E+02
SPI05	Core Soil	26-29.5 cm	28	1.97E+02

ID				Cs-137 (Bq/kg) Activity
SPI06	Core Soil	0-1 cm	0.5	2.00E+05
SPI06	Core Soil	1-2 cm	1.5	1.35E+05
SPI06	Core Soil	2-3 cm	2.5	6.34E+04
SPI06	Core Soil	3-5 cm	4	2.20E+04
SPI06	Core Soil	5-7 cm	6	2.67E+03
SPI06	Core Soil	7-10 cm	8.5	6.44E+02
SPI06	Core Soil	10-13 cm	11.5	9.32E+02

SPI06	Core Soil	13-17 cm	15	1.26E+03
SPI06	Core Soil	17-20 cm	18.5	8.20E+02
SPI06	Core Soil	20-25 cm	22.5	2.48E+02
SPI06	Core Soil	25-28.5 cm	26.7	1.45E+02

ID				Cs-137 (Bq/kg) Activity
SPI07	Core Soil	0-1 cm	0.5	1.47E+05
SPI07	Core Soil	1-2 cm	1.5	3.11E+05
SPI07	Core Soil	2-3 cm	2.5	4.90E+05
SPI07	Core Soil	3-5 cm	4	1.43E+05
SPI07	Core Soil	5-7 cm	6	4.75E+04
SPI07	Core Soil	7-10 cm	8.5	1.07E+04
SPI07	Core Soil	10-14 cm	12	4.32E+03
SPI07	Core Soil	14-17 cm	15.5	2.83E+03
SPI07	Core Soil	17-20 cm	18.5	7.17E+02
SPI07	Core Soil	20-25 cm	22.5	2.00E+02
SPI07	Core Soil	25-28.5 cm	26.7	1.20E+02

Kashiromori

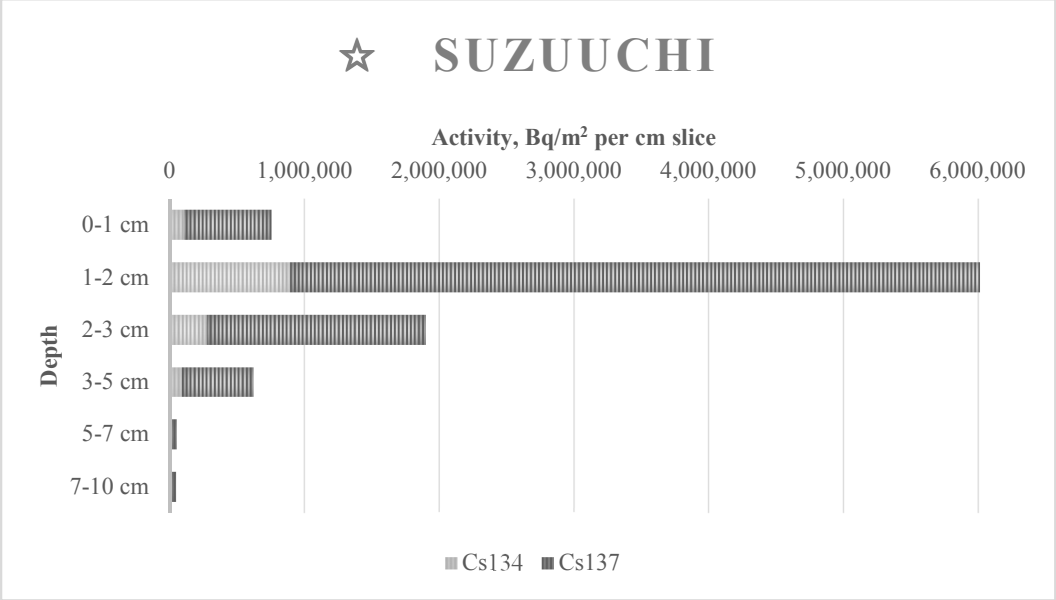
ID				Cs-137 (Bq/kg) Activity
SPK05	Core Soil	0-1 cm	0.5	3.99E+04
SPK05	Core Soil	1-2 cm	1.5	3.29E+04
SPK05	Core Soil	2-3 cm	2.5	1.37E+04
SPK05	Core Soil	3-5 cm	4	6.04E+03
SPK05	Core Soil	5-7 cm	6	9.82E+02
SPK05	Core Soil	7-10 cm	8.5	1.94E+02
SPK05	Core Soil	10-13 cm	11.5	1.81E+02
SPK05	Core Soil	13-17 cm	15	1.60E+02
SPK05	Core Soil	17-20 cm	18.5	1.61E+02
SPK05	Core Soil	20-25 cm	22.5	1.21E+02
SPK05	Core Soil	25-29 cm	27	7.71E+01

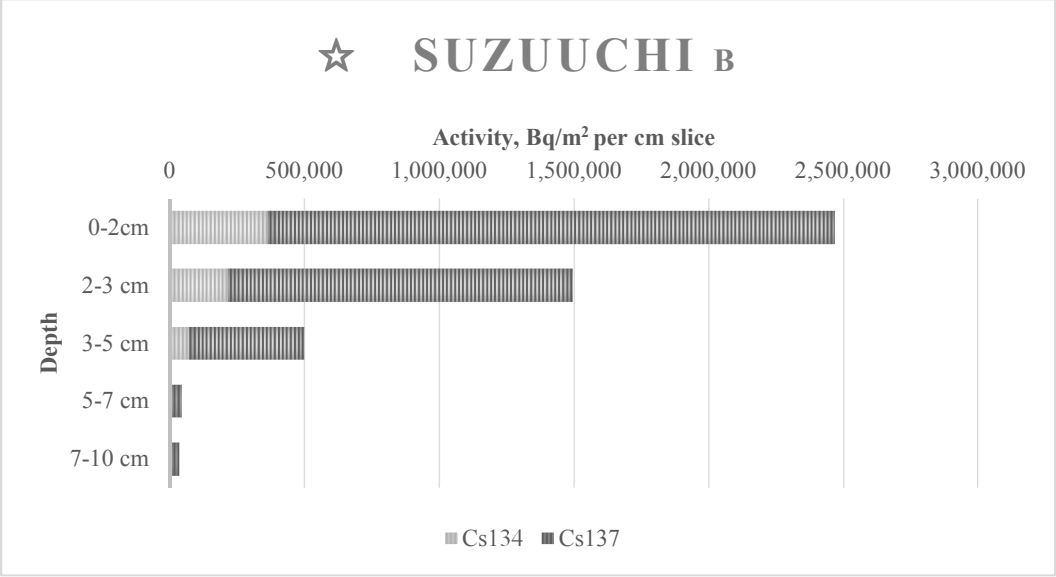
ID				Cs-137 (Bq/kg) Activity
SPK06	Core Soil	0-1 cm	0.5	6.71E+03
SPK06	Core Soil	1-2 cm	1.5	2.71E+03
SPK06	Core Soil	2-3 cm	2.5	1.73E+03
SPK06	Core Soil	3-5 cm	4	8.73E+02
SPK06	Core Soil	5-7 cm	6	1.24E+02
SPK06	Core Soil	7-10 cm	8.5	3.20E+02
SPK06	Core Soil	10-13 cm	11.5	1.85E+02
SPK06	Core Soil	13-17 cm	15	6.13E+01

SPK06	Core Soil	17-20 cm	18.5	1.89E+01
SPK06	Core Soil	20-25 cm	22.5	1.61E+00
SPK06	Core Soil	25-29 cm		

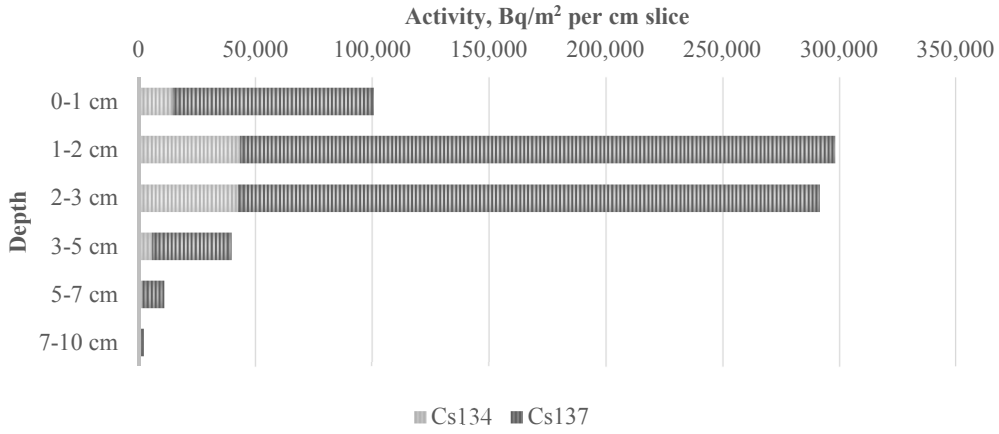
ID				Cs-137 (Bq/kg) Activity
SPK07	Core Soil	0-1 cm	0.5	2.27E+04
SPK07	Core Soil	1-2 cm	1.5	1.61E+04
SPK07	Core Soil	2-3 cm	2.5	5.11E+03
SPK07	Core Soil	3-5 cm	4	2.58E+03
SPK07	Core Soil	5-7 cm	6	1.09E+03
SPK07	Core Soil	7-10 cm	8.5	4.16E+02
SPK07	Core Soil	10-13 cm	11.5	1.80E+02
SPK07	Core Soil	13-17 cm	15	7.11E+01
SPK07	Core Soil	17-20 cm	18.5	2.85E+01
SPK07	Core Soil	20-25 cm	22.5	1.36E+01
SPK07	Core Soil	25-29 cm	27	7.19E+01

Appendix 2: Vertical distribution radiocesium soil profiles

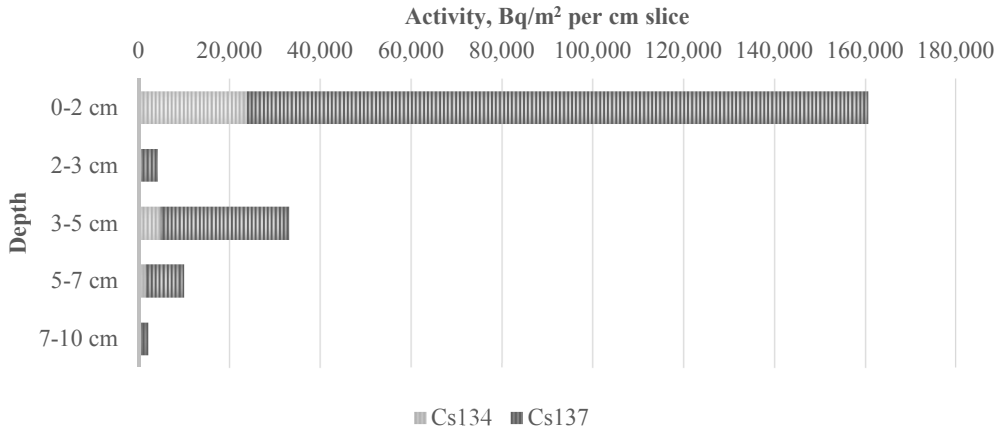


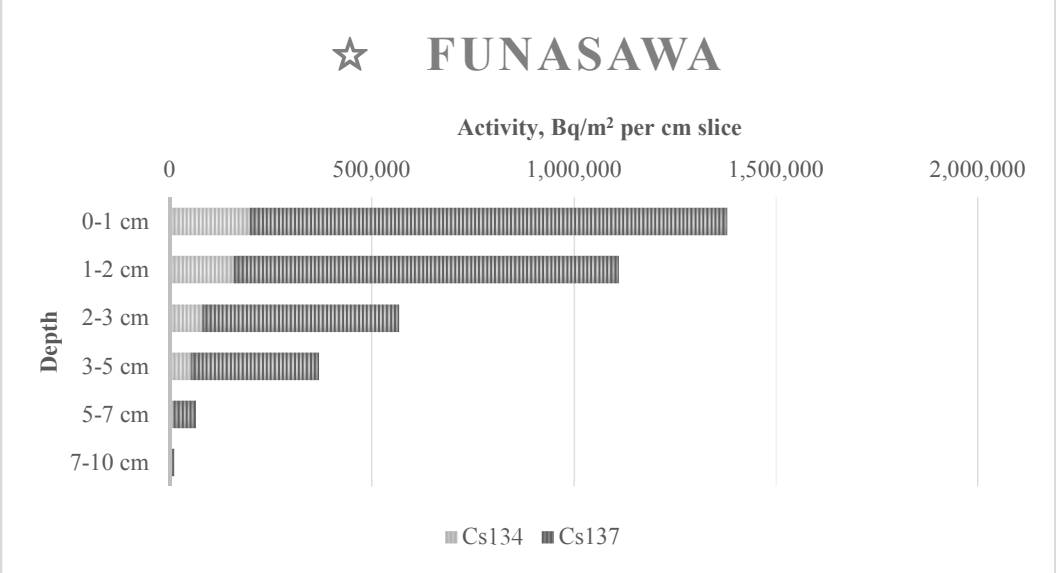


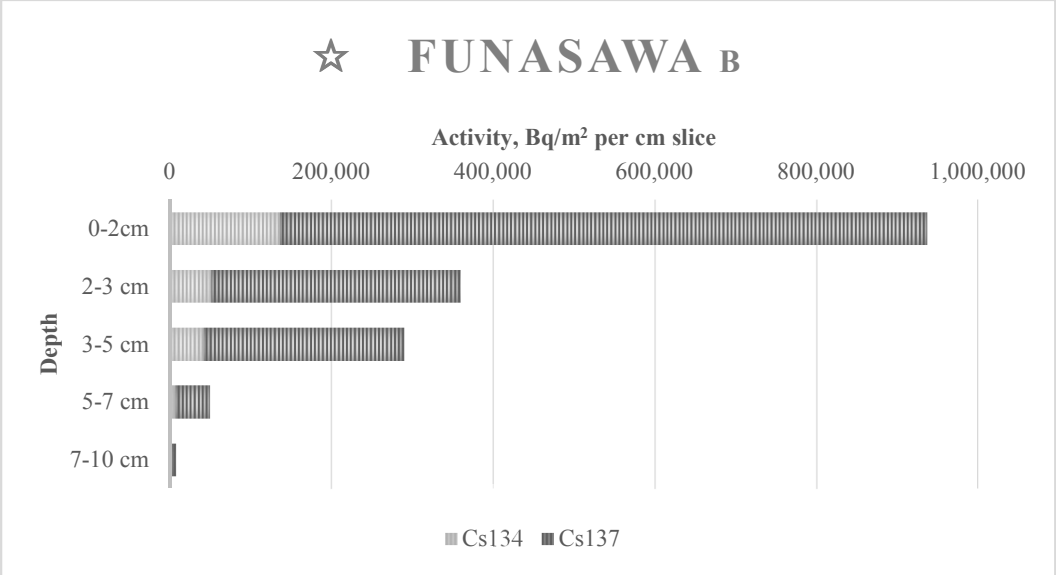
☆ KASHIROMORI

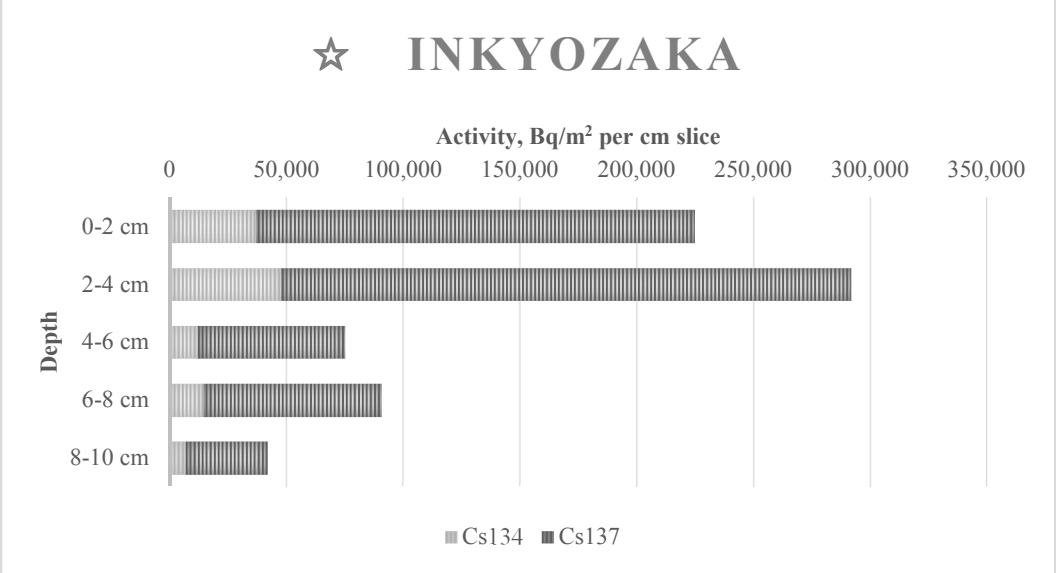


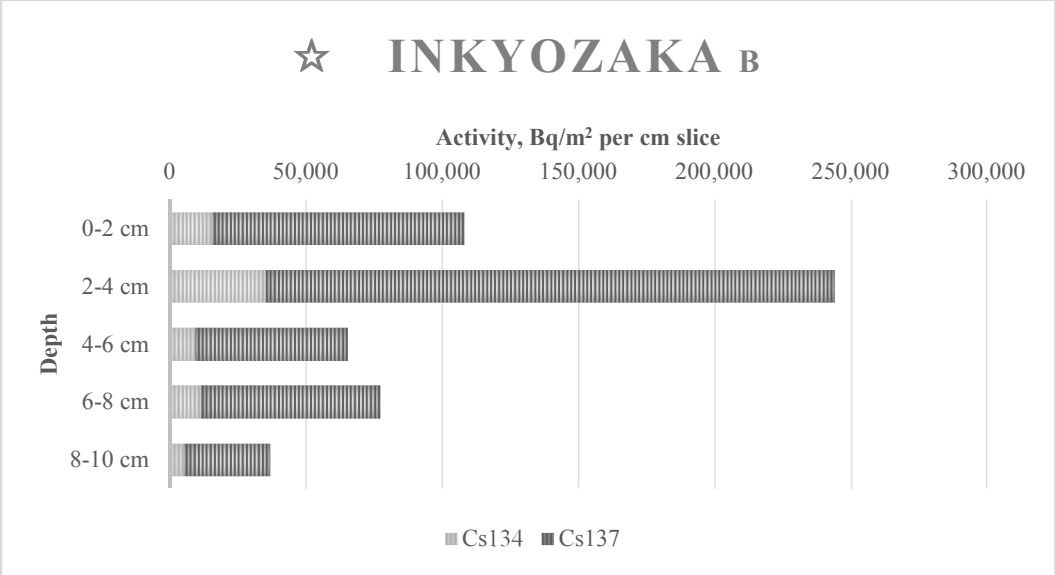
☆ KASHIROMORI B




☆ FUNASAWA
Activity, Bq/m² per cm slice
0
500,000
1,000,000
1,500,000
2,000,000
Depth
0-1 cm
1-2 cm
2-3 cm
3-5 cm
5-7 cm
7-10 cm
Cs134
Cs137

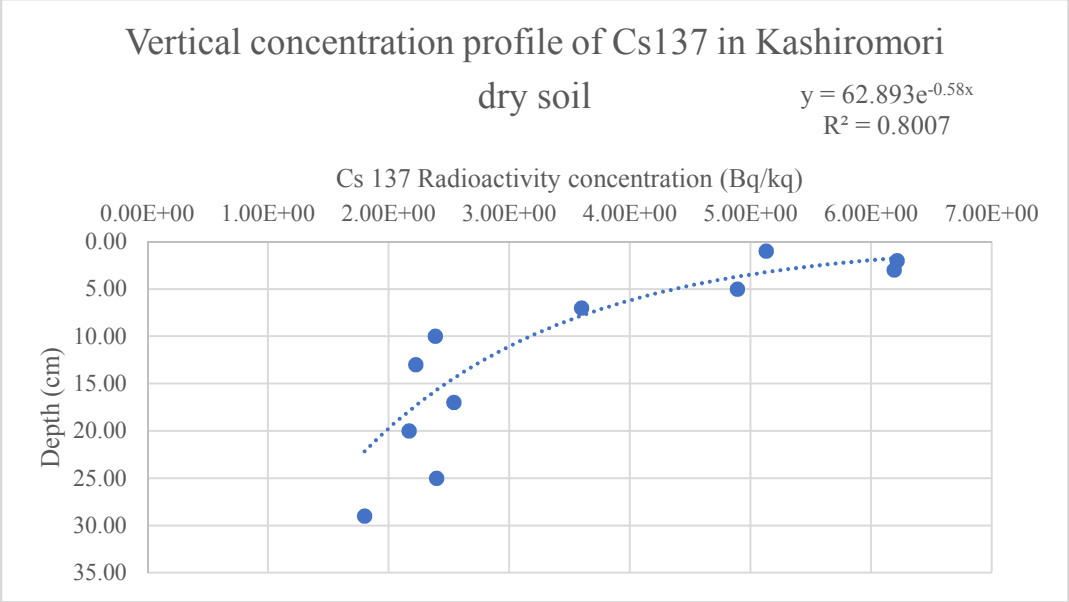

☆ FUNASAWA B
Activity, Bq/m² per cm slice
0
200,000
400,000
600,000
800,000
1,000,000
Depth
0-2cm
2-3 cm
3-5 cm
5-7 cm
7-10 cm
Cs134
Cs137


☆ INKYOZAKA
Activity, Bq/m² per cm slice
0
50,000
100,000
150,000
200,000
250,000
300,000
350,000
Depth
0-2 cm
2-4 cm
4-6 cm
6-8 cm
8-10 cm
Cs134
Cs137

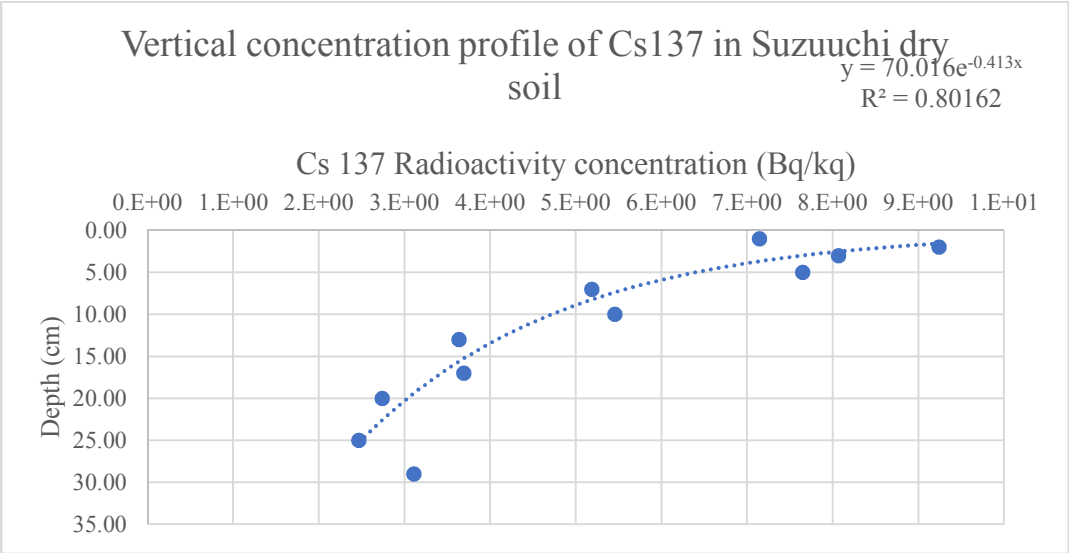

☆ INKYOZAKA B
Activity, Bq/m² per cm slice
0
50,000
100,000
150,000
200,000
250,000
300,000
Depth
0-2 cm
2-4 cm
4-6 cm
6-8 cm
8-10 cm
Cs134
Cs137

Appendix 3: Graphs of log transformed activity concentrations

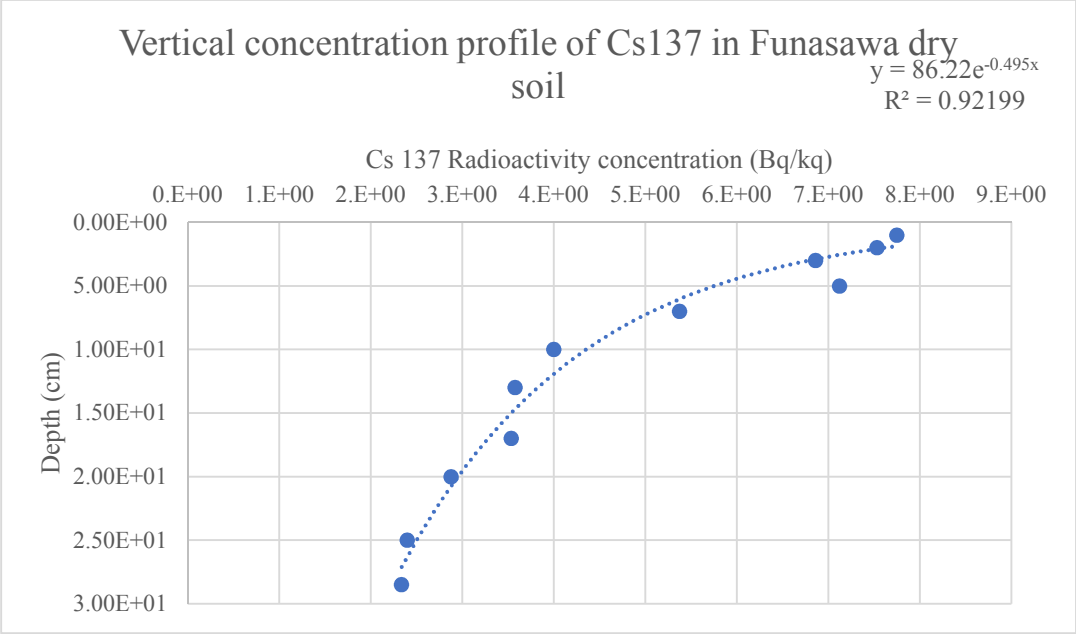
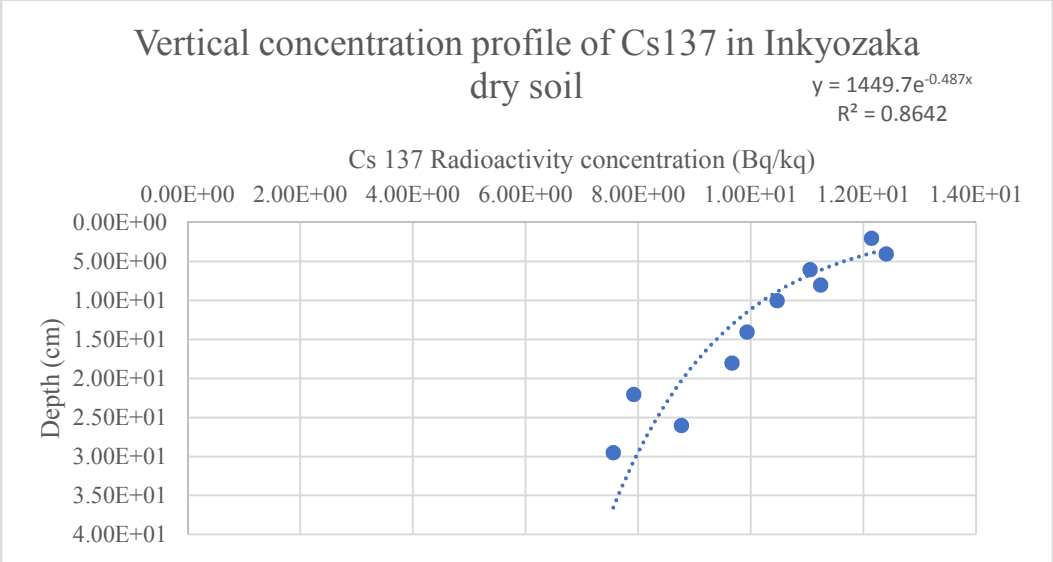
Isotope concentration vs depth for a soil core of 30 cm in length at Kashiromori



Isotope concentration vs depth for a soil core of 30 cm in length at Suzuuchi



Isotope concentration vs depth for a soil core of 30 cm in length at Funasawa



Isotope concentration vs depth for a soil core of 30 cm in length at Inkyozaka