

THESIS

RADIOCESIUM DYNAMICS IN IRRIGATION PONDS IN OKUMA, JAPAN

Submitted by

Ian Byrnes

Department of Environmental and Radiological Health Sciences

In partial fulfillment of the requirements

For the degree of Master of Science

Colorado State University

Fort Collins, Colorado

Spring 2017

Master's Committee

Advisor: Thomas Johnson

Alexander Brandl

Thomas Borch

Copyright by Ian Thomas Behnke Byrnes 2017

All Rights Reserved

ABSTRACT

RADIOCESIUM DYNAMICS IN IRRIGATION PONDS IN OKUMA, JAPAN

The objective of this research is to examine the variability in the vertical migration of ^{137}Cs in ponds located in the 10 km deposition zone surrounding the Fukushima Dai-ichi nuclear accident site in Fukushima Prefecture, Japan. Four ponds, utilized for the irrigation of local rice production, were selected for sampling. Three types of samples were collected from each pond. Sediment cores were extracted to a minimum depth of 30 cm. Grab samples of the top sediment layer were taken at each core location. Finally, water samples were obtained to determine hydrochemical properties and radiocesium concentration suspended in the water.

In the laboratory, the sediment cores were cut into sections and dried. Once the water was removed, the sediments sections were counted on a HPGe detector. Counting data results were used to construct a profile of the ^{137}Cs activity concentration by depth. Water samples, used to determine the solid-liquid distribution coefficient K_d , were filtered through a 0.45 micrometer filter and analyzed to determine the hydrochemical composition of the pond water as well as counted for ^{137}Cs activity concentrations. The grab samples were dried and packaged for HPGe counting and sequential extractions. Sequential extractions were used to determine the ratio of radiocesium that is bound to the sediments versus available to transfer through the environment. Two extractions were completed on the grab samples; an ammonium acetate extraction and an extraction with hydrogen peroxide solution (35%).

The results show that ^{137}Cs is very mobile in the top sediment layers. The highest concentrations of ^{137}Cs in the cores is not limited to the upper sediment layers suggesting some

combination of a higher mobilization of sediments in the ponds and post extraction mixing within the core resulting from the extraction technique. Similarly, the K_d values found are similar to those found in rivers in the affected areas of Fukushima. Collectively, the results suggest the ponds are open water bodies with the capacity to exchange ^{137}Cs with local agriculture.

ACKNOWLEDGEMENTS

This research would not have been possible without the encouragement and guidance of Dr. Thomas Johnson. He drove me to seek further education at Colorado State University (CSU) and kept me focused on completing my research in a timely fashion. I would also like to thank my committee members for supporting me as my research was developed into my Thesis.

I am very grateful to Dr. Alexander Brandl for providing technical support and instruction on the core of health physics. I would also like to thank Dr. Ralf Sudowe for illuminating me to the finer points of radiochemistry.

This research was completed in conjunction with the Institute of Environmental Radioactivity (IER) at Fukushima University. To collect the necessary samples, support from numerous members of the IER was required. Many thanks to Dr. Thomas Hinton, who coordinated the exchanges between CSU and the IER and provided many insights on radioecology as well as learning to live in Japan. I am deeply grateful to Dr. Alexei Konoplev, who helped design the project and explain the theory to me. A big thanks to Dr. Hirofumi Tsukada who was especially patient and held my hand through the chemistry work with sequential extractions. Thanks to Dr. Yoshifumi Wakiyama for helping me with field work and thank you to Dr. Tsugiko Takase for counting so many sediment samples and returning the data to me quickly. Finally, I would thank Dr. Kenji Nanba who welcomed me to the IER and Japan and introduced me to the incredible work Fukushima Prefecture is doing to respond to the Fukushima accident.

TABLE OF CONTENTS

ABSTRACT	ii
ACKNOWLEDGEMENTS	iv
METHODS	6
2.1 Site Description	6
2.2 Pond Sediment Core Samples	9
2.3 Pond Water Samples	12
2.4 Bottom Sediment Grab Samples	13
2.5 Sequential Extractions	14
RESULTS	15
3.1 Pond Sediment Core Profiles	15
3.2 Solid-Liquid Distribution Coefficient	20
3.3 Ion Chromatography and ICP/MS	20
3.4 Sequential Extractions	21
DISCUSSION	23
4.1 Sediment Core Profiles	23
4.2 Solid-Liquid Distribution Coefficient	28
4.3 Cation Measurements	28
4.4 Sequential Extractions	29
REFERENCES	33
APPENDIX 1: Raw Data from Sediment Cores	36
5.1 Inkyozaka	36
5.2 Suzuuchi	37
5.3 Funasawa	38
5.4 Kashiramori	39
APPENDIX 2: Sediment Densities by Depth	40
APPENDIX 3: Raw Data for Solid-Liquid Distribution Coefficient Calculation	42
APPENDIX 4: Copyright Information	43

INTRODUCTION

The Great East Japan Earthquake that occurred on March 11, 2011 triggered a tsunami that crippled the Fukushima Dai-ichi Nuclear Power Plant (FDNPP), operated by Tokyo Electric Power Company (TEPCO) (Steinhauser, Brandl, & Johnson, 2014). The following day, a release commenced of radioactive material that continued until March 15, 2011 and led to extensive soil contamination throughout the local ecosystem. Three towns near the accident site were evacuated; Okuma, Futaba, and Namie. After the accident, the International Atomic Energy Agency (IAEA) declared the accident an INES (International Nuclear and Radiological Event Scale) 7 meaning a “Major Accident” (Ministry of Agriculture, Forestry, and Fisheries, 2015). A public exposure risk exists because radiocesium can migrate through the environment and potentially into the food chain.

The accidental release of radioactive materials from FDNPP marks the second worst nuclear disaster in history after the Chernobyl accident in Ukraine in 1986. At the time of the incident, FDNPP comprised of 6 boiling water reactors (BWRs) operating at a combined power of 5480 MWe. Three reactors were in operation and three were being refueled or under repairs. The estimated total radioactive material released from the accident at FDNPP is about 15% of the radioactive material released by Chernobyl. The total activity of radioactive material released from Chernobyl is estimated to be 5.3×10^{18} Bq across 450,000 km², whereas from Fukushima the release is estimated to be 6.3×10^{17} Bq across 8,000 km² (Kortov & Ustyantsev, 2013). The release from FDNPP was only a gas phase release and occurred in pulses over the course of a week, allowing the operators to wait for advantageous weather conditions. After accounting for the full release, it was determined that 80% of the radioactive material was

released over the ocean (Steinhauser, Brandl, & Johnson, 2014). It is also worth noting that the release from Fukushima only affected land in Japan, while the release from Chernobyl affected several countries beyond Ukraine. To date, there are no human deaths attributed to the release of radioactive materials from the FDNPP accident.

Of the radionuclides deposited in the environment, radioactive isotopes of cesium are of primary concern due to a long half-life. Radiocesium, generally, appears in two forms; ^{134}Cs ($T_r = 2.06$ years) and ^{137}Cs ($T_r = 30.17$ years). ^{137}Cs can originate as both a direct fission fragment of ^{235}U or as a decay product of other fission fragments (a fission product). Therefore, it has a high cumulative yield of 6.221% (of fission products of ^{235}U) (International Atomic Energy Agency, 2017). The cumulative yield is defined as the sum of the independent yield of the isotope as well as the independent yield of all the precursor isotopes (Baum, Ernesti, Knox, Miller, & Watson, 2010). By comparison, the cumulative yield of ^{134}Cs is 0.0000121% (International Atomic Energy Agency, 2017). The yield for ^{134}Cs is significantly lower for ^{134}Cs as it does not have any precursor isotopes; meaning no isotopes decay to ^{134}Cs . In combination with a shorter half-life than ^{137}Cs , the activity of ^{134}Cs decreases more swiftly in samples. The decay process of ^{137}Cs is characterized by the emission of a unique photon with energy of 662 keV; making the isotope easy to identify and measure in environmental samples. ^{137}Cs primarily decays by beta emission to a metastable state of $^{137\text{m}}\text{Ba}$ which in turn drops to a stable ^{137}Ba by emitting a 662 keV photon (Brookhaven National Laboratory, 2017). With a significantly longer half-life and greater fission product yield, ^{137}Cs is a more persistent issue than ^{134}Cs . As of June 6, 2011, 1.7×10^{16} Bq of ^{137}Cs was released from the Fukushima accident (Baba, 2013).

Once deposited in the environment, the presence of radiocesium in both soluble and particulate form, can lead to potential uptake by flora and fauna. Radiocesium can enter flora

through aerial deposition and surface absorption, or by root uptake. Intake in fauna occurs by inhalation, ingestion, or by surface absorption (Whicker & Schultz, 1982). Soil properties play a major role in the transport of radiocesium in the ecosystem. Particularly, high clay content can immobilize cesium effectively by chemical binding (Whicker & Schultz, 1982). Conversely, sandy soils have a low cation exchange which makes radiocesium more available to migrate through different parts of the ecosystem. The ability of soils and sediments, when suspended in solution with water, to bind radiocesium is described by the solid-liquid distribution coefficient (K_d). In a water body, the K_d is given by the ratio of the concentration of radiocesium in the suspended sediments to the concentration of radiocesium dissolved in the water (Whicker & Schultz, 1982). A higher K_d is an indication that the suspended sediments are more adept at binding radiocesium in the water. Conversely, a lower K_d means there is more radiocesium dissolved in the water, which is problematic in terms of ecosystem transport and bioavailability.

There are several cations that effect the binding of radiocesium to soils; primarily potassium ions (K^+), ammonium (NH_4^+), and stable cesium ($^{133}Cs^+$) (Whicker & Schultz, 1982). Cesium is chemically analogous to potassium which is a vital nutrient for plants and animals (NCRP 154, 2006). In biotic systems with an abundance of potassium, ^{137}Cs binding to soils is reduced since potassium cations compete for the same binding locations. The inverse is true as well, a vacancy of potassium provides availability for ^{137}Cs to bind to soils. Similarly, in aquatic ecosystems, anoxic conditions can affect the mobilization of ^{137}Cs . Anoxia, an issue in aquatic environments where oxygen levels are low, is characterized by an increase in ammonium concentrations creating an environment that competes for sorption locations, similar to potassium cations, with ^{137}Cs (NCRP 154, 2006). Finally, the presence of stable cesium is key to the transport of radiocesium as it is significantly more competitive at acquiring binding locations

than potassium and ammonium in clay soils. Specifically, $^{133}\text{Cs}^+$ is about 1000 times more competitive than potassium and 200 times more competitive than ammonium (Cremers, Elsen, De Preter, & Maes, 1988). However, stable cesium only constitutes a tiny fraction ($\sim 7 \mu\text{g/g}$) of the elements present in the environment (NCRP 154, 2006). Generally, it is expected to have minimal impact on ^{137}Cs retention.

In radioecology, ecosystems are often described in terms of compartments. Radiocesium is transferred from one compartment to another. A compartment that receives radiocesium but does not release any is termed a “sink” (NCRP 154, 2006). Ponds and lakes often represent sinks in the environment. Radiocesium transfers through the ecosystem and becomes bound to the clay in a pond and slowly descends into deeper strata of the sediments. At this point, there is negligible biological contact and protection from erosion so the radiocesium is buried (Whicker & Schultz, 1982). Unfortunately, an actual pond or lake is never a perfect sink because it is not a closed body of water. An important distinguishing characteristic is between an open or a closed water body. An open body is one that freely exchanges its water with the habitat around it. A river is an example of an open water body. Conversely, a closed body retains its water with little to no exchange (Bulgakov, et al., 2002).

In Japan, the areas surrounding the Fukushima accident site contain a range of small irrigation ponds. There are 150 similar water bodies in Okuma alone (Ministry of Agriculture, Forestry, and Fisheries, 2015). The ponds in the affected area were primarily used for regulating the irrigation of rice paddies, meaning there is a lot of water exchange. Compared to Ukraine, the ecosystem in Fukushima contains significantly more moisture. Annual rainfall averages about 1105 mm in Fukushima compared to 483 mm in Ukraine (Konoplev A. , et al., 2016). The sequestration of radiocesium in the bottom sediment of irrigation ponds is the next step in

understanding the extent of long term contamination in the ecosystem. The long half-life of ^{137}Cs allows it to remain in the environment and potentially be consumed further up the food chain. Determining the volume of radiocesium that can be transported is important in two ways. First, understanding the transport of radiocesium can provide key insights into the radioactive contamination clean-up that is ongoing in Fukushima Prefecture. The second is to provide a data point in the construction of the overall radiation exposure to the public. The clean-up resulting from the Fukushima Dai-ichi accident has been a key environmental effort in Japan since 2011. It also resulted in misunderstanding and misinterpreting the local area radioactive contamination. In this study, ^{137}Cs activity concentrations in pond sediments were found by examining extracted sediment cores. Combining core data with hydrochemical characteristics of irrigation ponds can shed light on the distribution and potential migration of ^{137}Cs as a result of the deposition from FDNPP. Developing an understanding of the environmental contamination will support a wider effort in Japan to disseminate information on the accident and the extent of the damage. By educating the population, Fukushima Prefecture hopes to calm concerns and improve its economy.

The objective of this research is to assess the extent of ^{137}Cs contamination in irrigation ponds in Okuma, Japan. Evaluating these ponds involves examining ^{137}Cs concentrations in the bottom sediments as well as in the water. The calculation of K_d values for the ponds provides a measure of binding to clay minerals and a comparison point to previous studies. Chemical extraction methods can also be used to examine how much of the ^{137}Cs is binding to the clays in the sediments. All these data points can provide a greater picture of the contamination of ^{137}Cs in irrigation ponds, 5 years after the accident at FDNPP.

METHODS

2.1 Site Description

All samples were collected from the 10 kilometer (km) exclusion zone around FDNPP, in Okuma, Japan. The irrigation ponds selected for sampling were Inkyozaka, Suzuuchi, Funasawa, and Kashiramori. A map of the Japanese town, Okuma, with the locations of the four ponds is shown in Figure 1. Table 1 shows the general characteristics of the target ponds (Konoplev A. , et al., 2015).

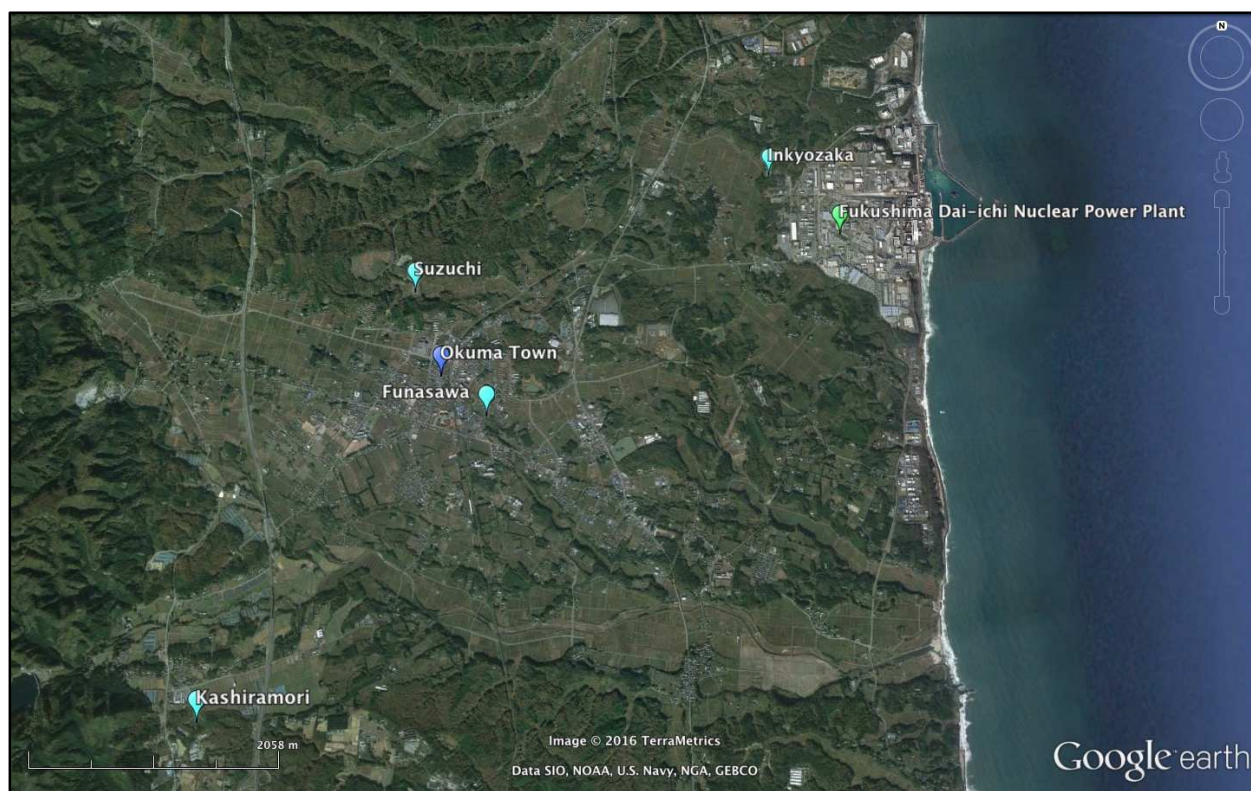


Figure 1: Map of Pond Locations in Okuma

Table 1: Pond Characteristics

Location	Site	Distance to FDNPP (km)	Depth (m)	Surface Area (m ²)	Sediment Type
<i>Inkyozaka</i>	N35°25.499' E141°01.05'	0.24	2.0	6500	Fluviosol
<i>Suzuuchi</i>	N37°24.950' E140°58.791'	3.75	1.0	4100	Fluviosol
<i>Funasawa</i>	N37°24.363' E140°59.173'	3.50	2.5	10700	Terrestrial Rigosol
<i>Kashiramori</i>	N37°22.996' E140°56.400'	7.00	5.0	8100	Andosol



Figure 2: Inkyozaka – arrows indicate approximate core locations



Figure 3: Suzuuchi – arrows indicate approximate core locations

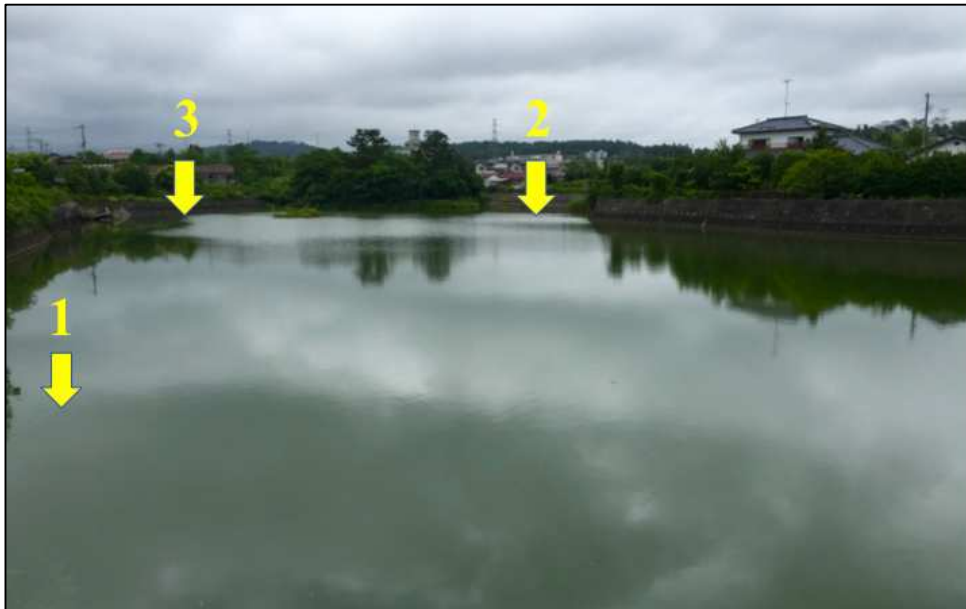


Figure 4: Funasawa – arrows indicate approximate core locations

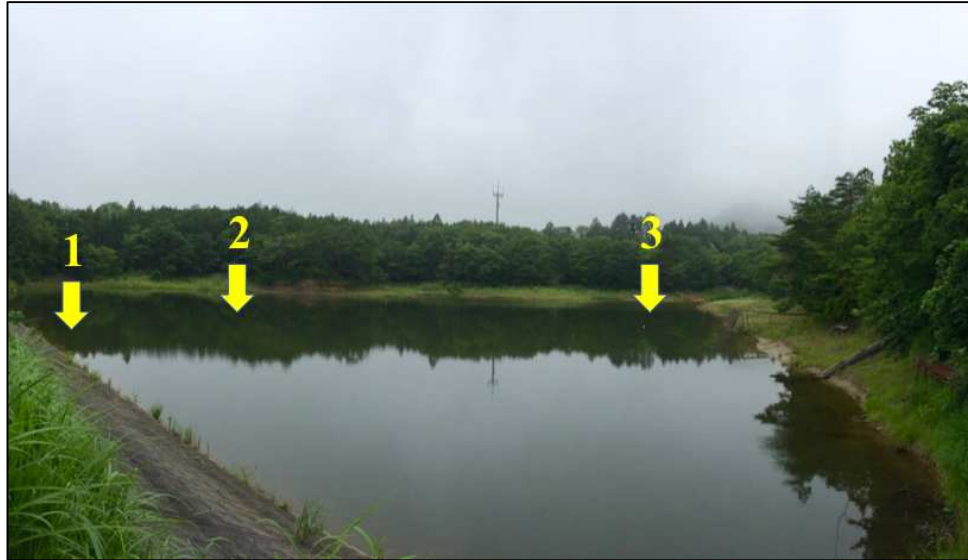


Figure 5: Kashiramori – arrows indicate approximate core locations

Three types of samples were collected: three bottom sediment cores at depths of 30 – 50 cm, three top layer grab samples, and small volume (50 mL) and large volume (4 L) water samples. Photographs of each pond with core locations indicated with arrows are shown in Figures 2 – 5.

2.2 Pond Sediment Core Samples

The bottom sediment cores were extracted using 2 meter (m) polyvinyl tubes of 4 centimeter (cm) inner diameter. Each tube was hammered into the sediment to a depth of 30 cm minimum. On the shore, the tubes were cut down, labeled, and sealed for transportation back to the lab, as shown in Figure 6.



Figure 6: The core is cut down and sealed, by cork and tape, on shore prior to transport to the IER

Once returned to the Institute of Environmental Radioactivity (IER), at Fukushima University, the sediment cores were separated into segments. The separation method is shown in Figure 7. Figure 8 shows the core after it has been partitioned.

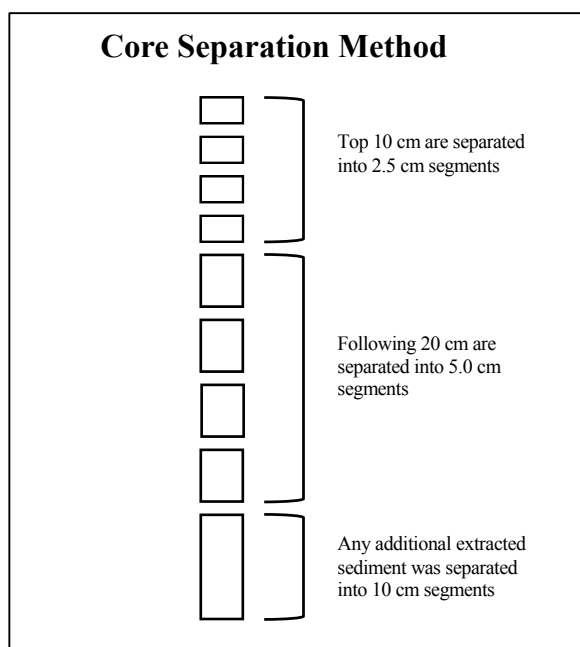


Figure 7: Core Separation Method



Figure 8: Core segments in pans prior to drying

All core portions were dried in a Panasonic MOV-212-PJ (Panasonic, Japan) dryer or a Yamato DY600 (Yamato, Japan) at 70 °C, for at least 48 hours. Once dry, each segment was prepared for gamma spectrometry. Dry segments were ground in a crucible and deposited in plastic cups, shown in Figure 9.



Figure 9: Dried core segment in the crucible for grinding (Left); Packaged core sample segments prepared for gamma spectroscopy (Right)

The mass of the dried sediment material was documented. All gamma spectrometry measurements were completed using a CANBERRA GC3018 (CANBERRA, USA) high purity germanium (HPGe) detector with a relative efficiency of 42.5%. The time of measurement was set to ensure a statistical uncertainty of $\leq 5\%$.

2.3 Pond Water Samples

Water samples were collected from each pond with a 2 L bucket on a 1 m pole in June, July, and August of 2016. The water samples were used to determine the concentration of suspended sediments and radiocesium in the ponds. In the laboratory, 4 L of water was filtered through a 0.45 μm pore cellulose acetate membrane filter. The filter was dried in a Yamato DY600 (Yamato, Japan) at 50 °C for at least 48 hours and packaged into a cup for gamma

spectrometry. 2 L of filtered water were also saved for gamma spectrometry. The small volume samples were also filtered through a 0.45 μm mesh syringe filter. The pH was measured for the filtered water. The remaining water was measured for dissolved K^+ , NH_4^+ , Na^+ , Ca^{2+} , and Mg^{2+} by ion chromatography with a DIONEX 1100 (Thermo-Fischer Scientific, USA). ICP Mass Spectroscopy was completed with an ELAN DRC 2 (Perkins Elmer, USA).

2.4 Bottom Sediment Grab Samples

Bottom sediment grab samples, used for sequential extractions, were taken at each core location with an Ekman-Berge Bottom Sampler shown in Figure 10. The total number of samples were three per pond.



Figure 10: Ekman-Berge Bottom Sampler

In the field, the samples were placed in a sealed plastic bag for transportation to the IER. In the laboratory, the samples were removed and placed into pans to be dried at 105 $^{\circ}\text{C}$ in a Panasonic MOV-212-PJ dryer (Panasonic, Japan), for at least one week. Once dried, 5 cm of sediment material were placed in a cup for gamma spectrometry with the Canberra GC3018 used

previously with the sediment cores. The remainder of the sediment material was used for sequential extractions.

2.5 Sequential Extractions

Sequential extractions were used to determine the fraction of radiocesium bound to inorganic and organic materials in the pond sediments. The sample sediments were taken from the grab samples collected with the Ekman-Berge sampler. The extraction procedure was completed per the method outlined by Hirofumi Tsukada in 2007. Two extractions were completed (Tsukada, Takeda, Hisamatsu, & Inaba, 2007). Samples were shaken in tubes with an ammonium acetate solution ($\text{CH}_3\text{COONH}_4$) and spun in a centrifuge for 10 minutes (min) at 4000 revolutions per minute (rpm). The second extraction was with a hydrogen peroxide solution (35%).

RESULTS

3.1 Pond Sediment Core Profiles

The dried segments of each core were counted, via a high purity germanium (HPGe) detector, and the results are presented in Becquerel per kilogram (Bq/kg), dry mass. Figures 11 - 14 show the results of the sediment core profiles by pond. Data can be found in Appendix 1.

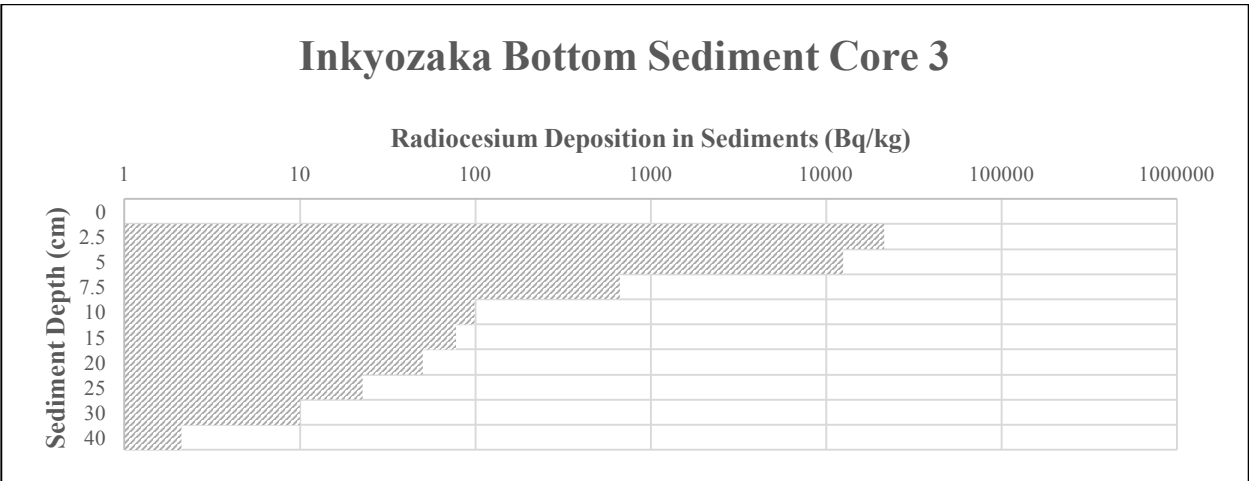
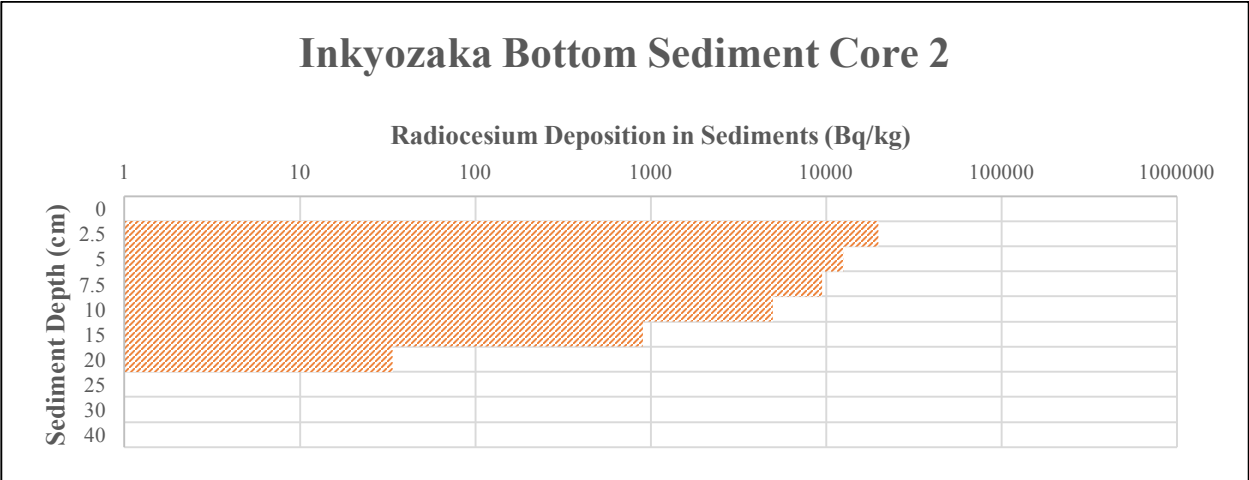
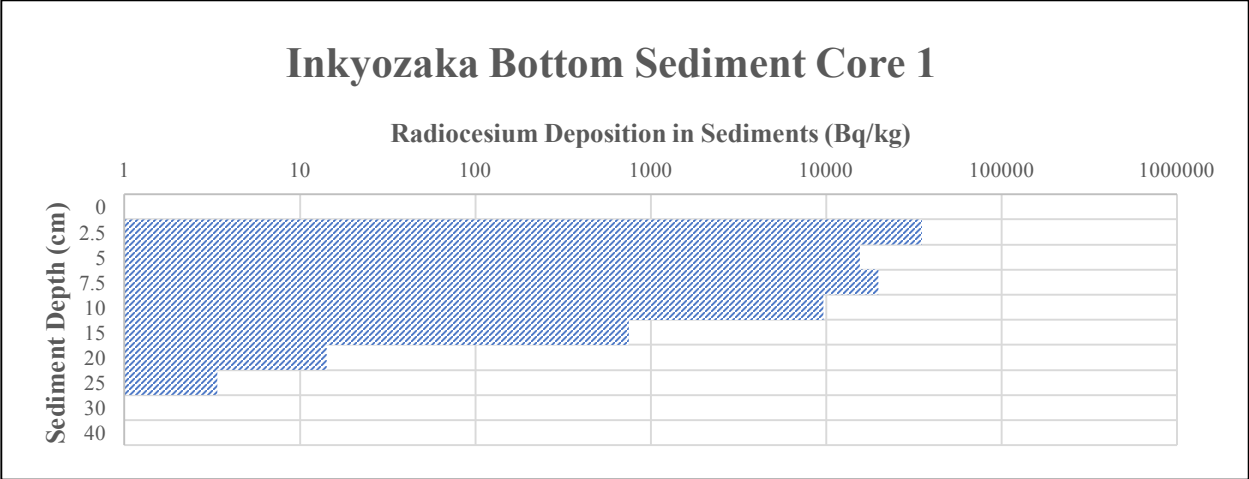


Figure 11: Core Profiles Extracted from Inkyozaka

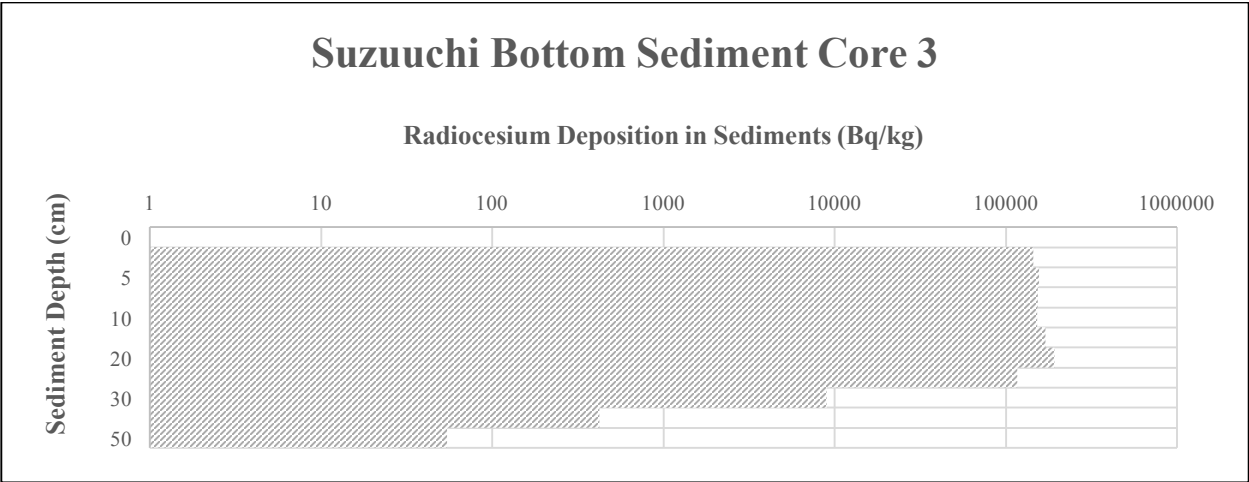
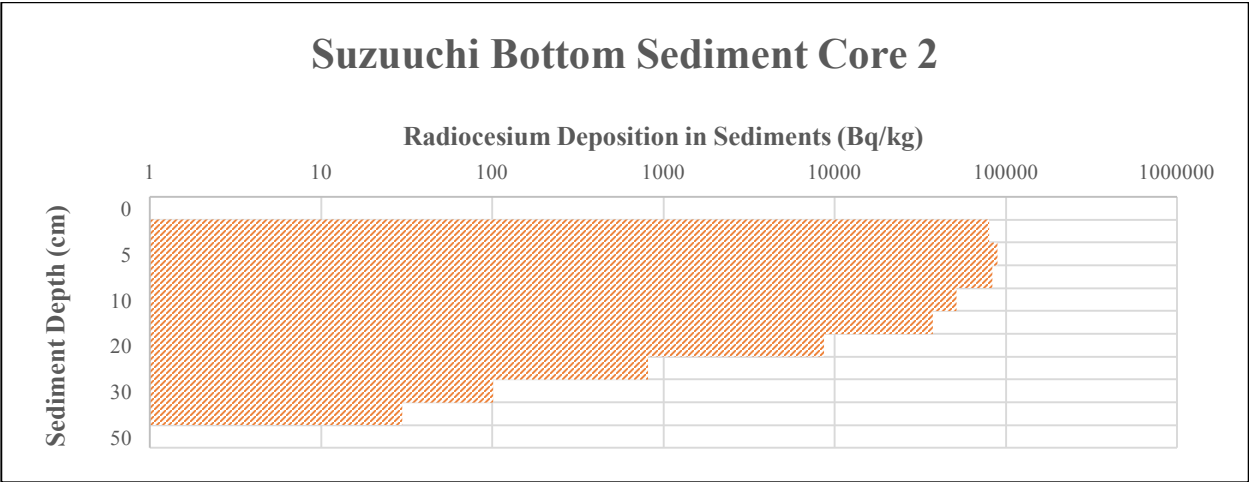
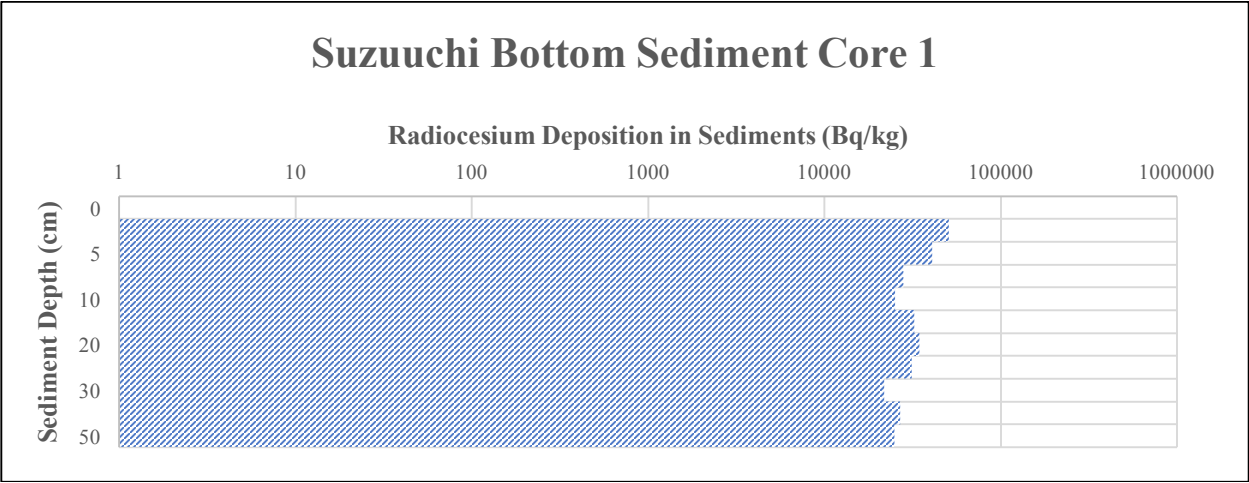


Figure 12: Core Profiles Extracted from Suzuuchi

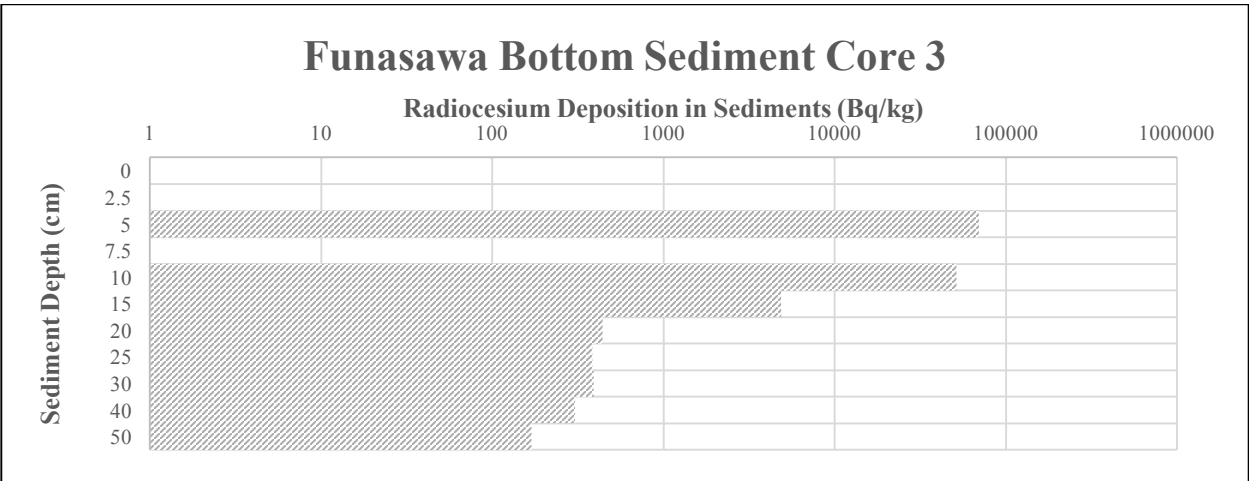
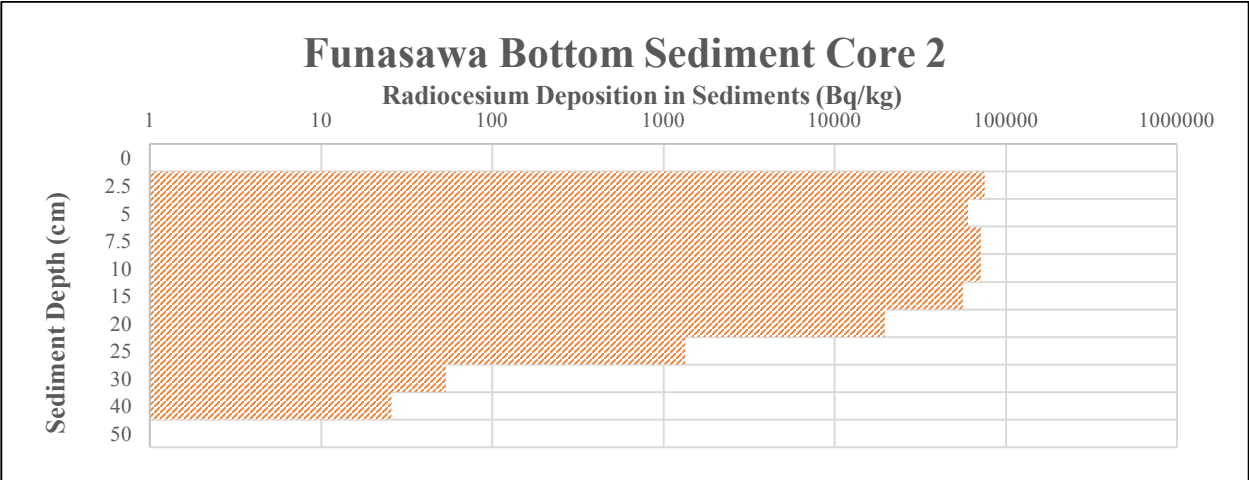
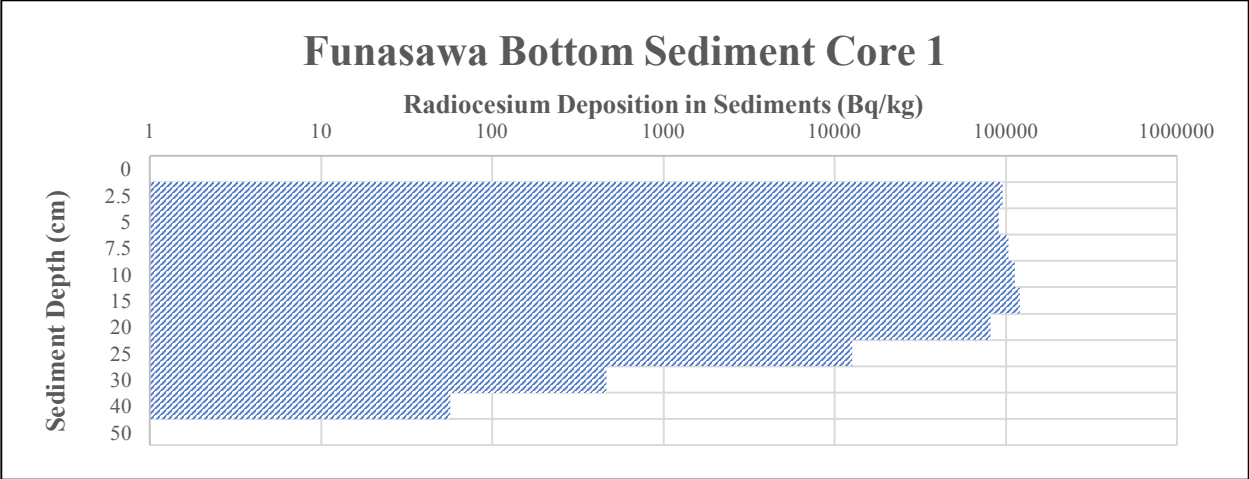


Figure 13: Core Profiles Extracted from Funasawa

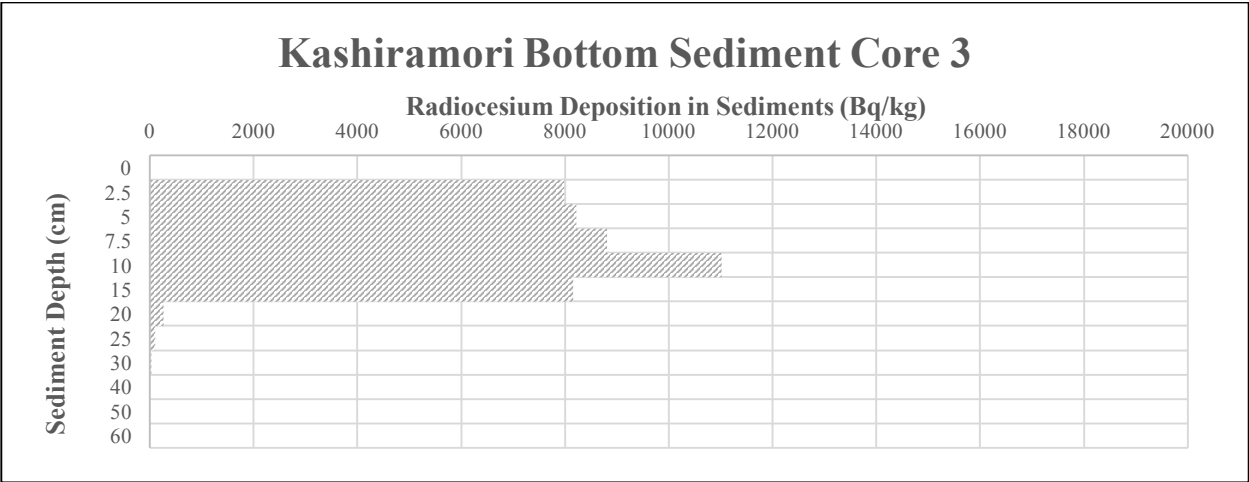
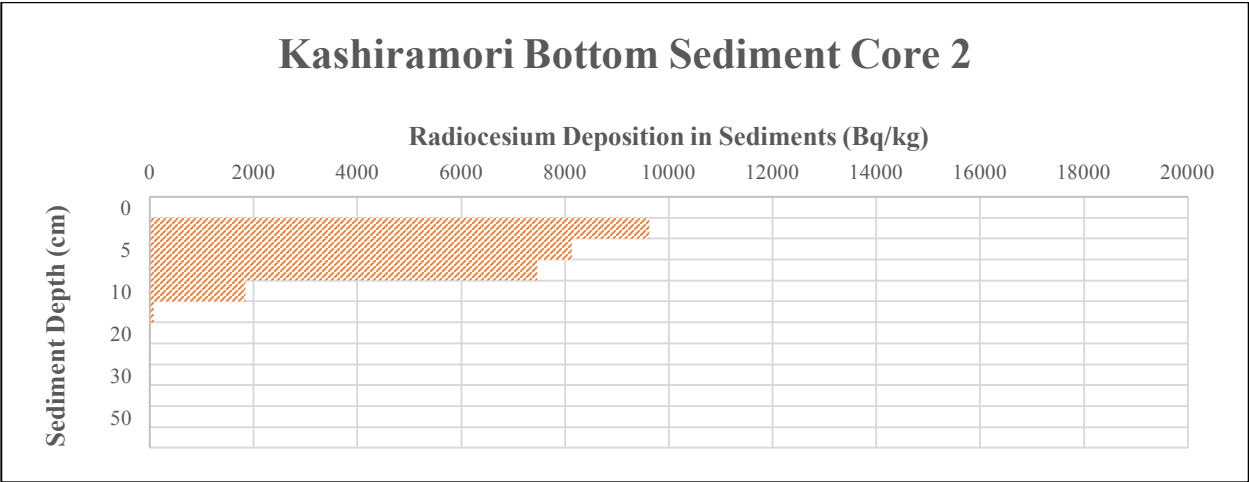
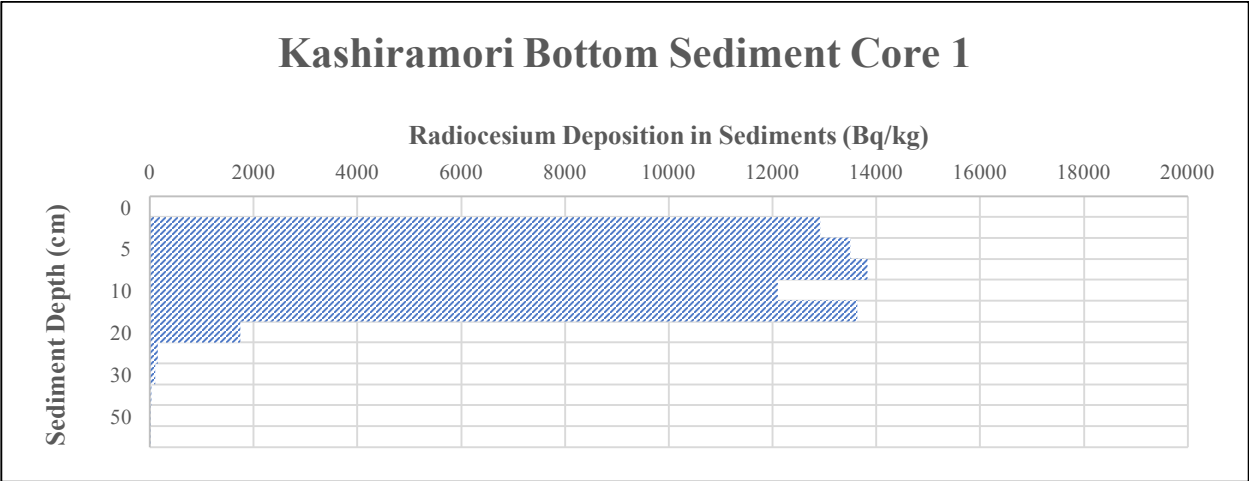


Figure 14: Core Profiles Extracted from Kashiramori

3.2 Solid-Liquid Distribution Coefficient

The solid-liquid distribution coefficient (K_d) is determined using the activity concentration in the suspended material counted on the filter (in Bq/kg) and the activity concentration of the dissolved ^{137}Cs in the filtered water (in Bq/L). The calculation to find the K_d values was completed using equation 1 (NCRP 154, 2006). Figure 15 show the K_d values for each pond.

$$K_d \left(\frac{\text{L}}{\text{kg}} \right) = \frac{\text{Activity Concentration in Suspended Solids} \left(\frac{\text{Bq}}{\text{kg}} \right)}{\text{Activity Concentration in Water} \left(\frac{\text{Bq}}{\text{L}} \right)} \quad (1)$$

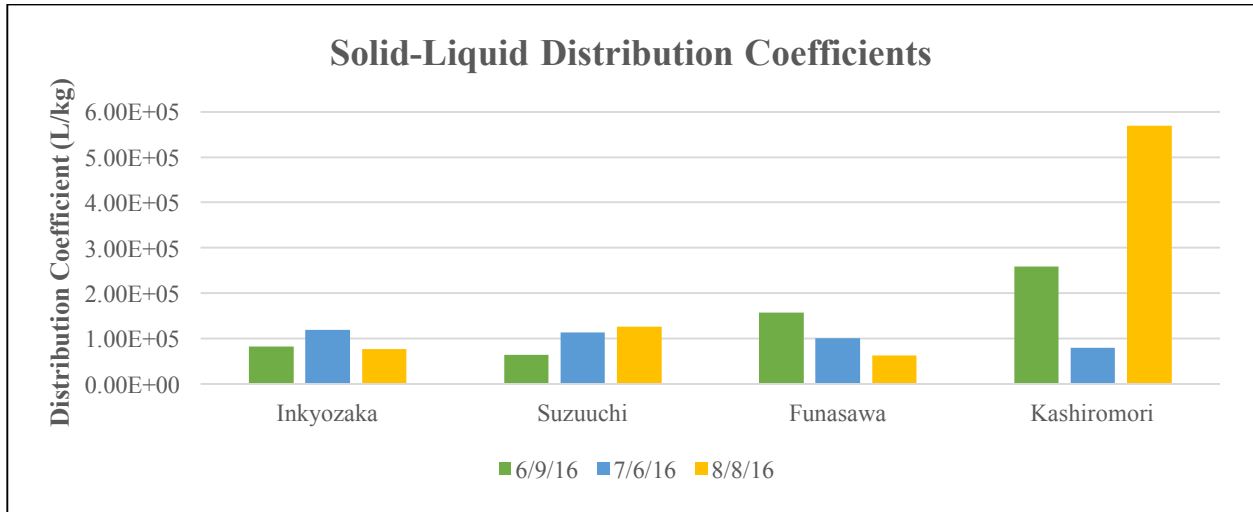


Figure 15: Calculated K_d Values for each of the four ponds in Okuma

3.3 Ion Chromatography and ICP/MS

Pond water was measured for dissolved K^+ , NH_4^+ , Na^+ , Ca^{2+} , and Mg^{2+} . Concentrations of $^{133}\text{Cs}^+$ were measured with ICP/MS. Samples are tested within 24 hours of extraction from the pond. Table 2 shows the results.

Table 2: Results from Ion Chromatography and ICP/MS

Hydrochemistry Results								
Pond	Sampling Date	pH	K ⁺ mg/L	NH ₄ ⁺ mg/L	Na ⁺ mg/L	Ca ₂ ⁺ mg/L	Mg ₂ ⁺ mg/L	¹³³ Cs ⁺ ng/L
Inkyozaka	6/9/16	8.13	0.79	ND	5.5776	2.06	0.8958	42
Inkyozaka	7/6/16	7.57	0.7928	ND	5.1449	2.38	1.0014	27
Inkyozaka	8/8/16	8.46	0.8155	ND	5.1036	2.11	0.9026	27
Suzuuchi	6/9/16	7.7	2.8677	0.0978	7.7579	15.35	5.6852	38
Suzuuchi	7/6/16	7.569	2.5847	0.0783	6.8971	14.09	5.1535	16
Suzuuchi	8/8/16	7.934	2.4028	ND	5.1421	10.52	3.6952	12
Funasawa	6/9/16	7.701	1.9075	ND	6.2805	23.41	3.7379	37
Funasawa	7/6/16	8.133	1.8365	ND	5.8549	21.87	3.3938	20
Funasawa	8/8/16	8.516	2.2325	ND	6.847	25.56	3.7603	27
Kashiramori	6/9/16	8.143	1.1109	ND	6.2953	14.61	3.0228	42
Kashiramori	7/6/16	8.008	1.1055	ND	5.8763	14.60	2.8769	17
Kashiramori	8/8/16	8.348	1.1277	ND	6.0752	14.67	2.8892	15

3.4 Sequential Extractions

The results of the two completed extractions are shown in Figures 16 and 17. The first extraction determined the ratio of ¹³⁷Cs bound to organic materials in the sediment. The second extraction shows the fraction of ¹³⁷Cs bound to inorganic materials, after destruction of the organic components by hydrogen peroxide.

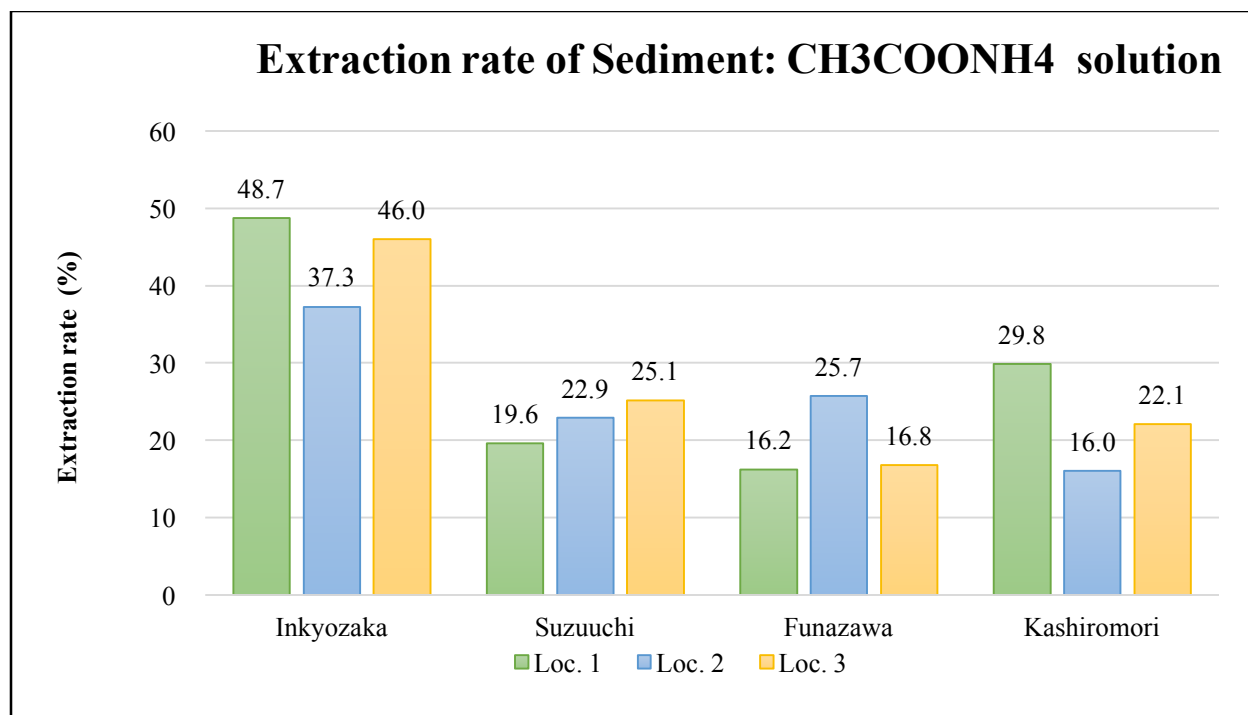


Figure 16: Extraction Percentage from Organic Materials

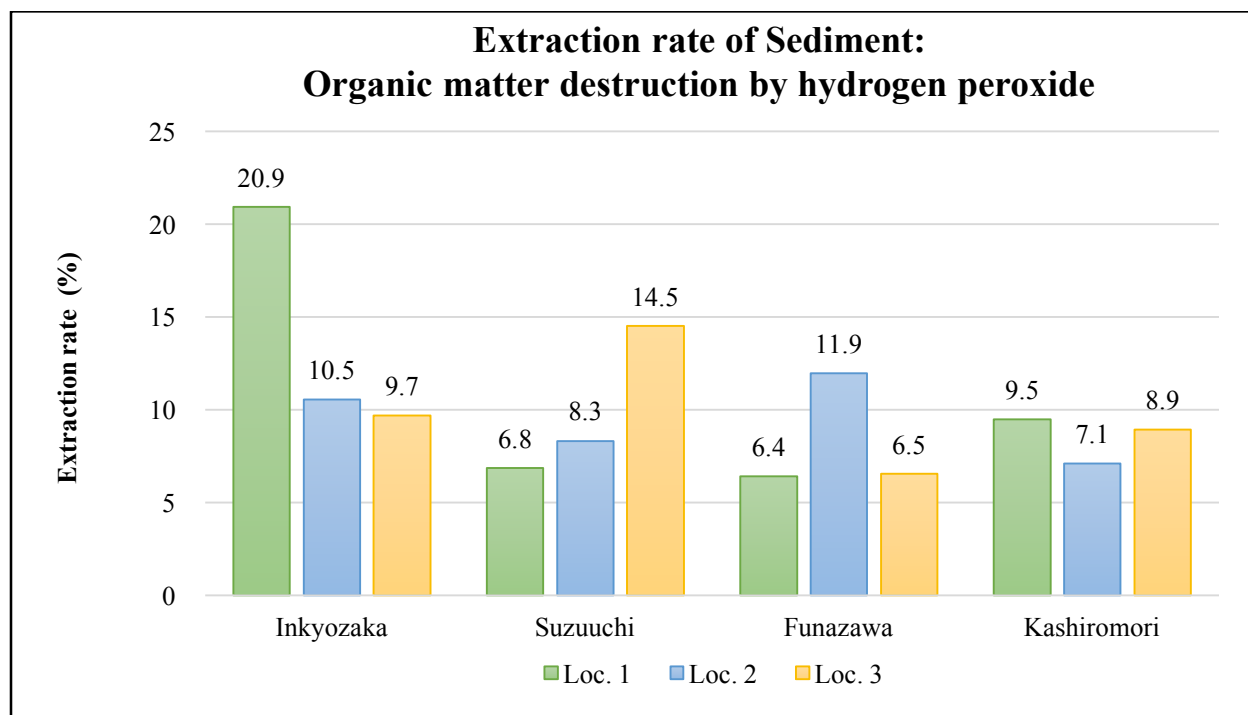


Figure 17: Extraction Percentage from Inorganic Materials

DISCUSSION

4.1 Sediment Core Profiles

The target ponds, Inkyozaka, Suzuuchi, Funasawa, and Kashiramori, are representative of the numerous ponds in Fukushima Prefecture (Ministry of Agriculture, Forestry, and Fisheries, 2015). Average ^{137}Cs activity concentrations in the surface sediments were on the order of $1.0 \times 10^{+04}$ to $1.0 \times 10^{+05}$ Bq/kg for all four ponds. The samples were extracted 5 years after the original deposition in March 2011. All cores showed higher activity concentrations in the upper third of the profile. Table 3 shows the percent of ^{137}Cs found in the top 10 cm of each core.

Table 3: The Percentage of ^{137}Cs that was found in the top 10 cm of each core and the average in each pond

Inkyozaka 1	Inkyozaka 2	Inkyozaka 3	Average
99%	98%	99%	99%
Suzuuchi 1	Suzuuchi 2	Suzuuchi 3	Average
46%	87%	55%	63%
Funasawa 1	Funasawa 2	Funasawa 3	Average
65%	78%	95%	79%
Kashiramori 1	Kashiramori 2	Kashiramori 3	Average
77%	100%	81%	86%

Previous research has shown that radiocesium takes some time to move out of the top sediment layers in water bodies. Studies of White Oak Lake at Oak Ridge National Laboratory, Tennessee, U.S.A. in the 1960's showed that 65% of the ^{137}Cs was concentrated in the top 6 cm of sampled sediment cores (Lomenick & Gardiner, 1965). Liquid waste discharges to this lake were started in 1944 and 250 core samples were extracted from 1950-1954; 6 to 10 years later. A similar study was conducted at Pond B at the Savannah River Site in South Carolina, U.S.A. (Pinder III, Bowling, Lide, & Beatty, 1995). Discharges from a former reactor were directed to this pond from 1961-1964. Sediment cores were extracted 23 years later in 1987 and found, on

average, that 75% of the ^{137}Cs was found in the top 4 cm. To make a more apt comparison, Table 4 shows the percent of ^{137}Cs that is found in the top 5 cm of the ponds in Okuma, Japan.

Table 4: The percentage of ^{137}Cs that was found in the top 5 cm of each core and the average in each pond

Inkyozaka 1	Inkyozaka 2	Inkyozaka 3	Average
63%	68%	97%	76%
Suzuuchi 1	Suzuuchi 2	Suzuuchi 3	Average
29%	48%	28%	35%
Funasawa 1	Funasawa 2	Funasawa 3	Average
30%	38%	54%	41%
Kashiramori 1	Kashiramori 2	Kashiramori 3	Average
39%	65%	36%	47%

The cores in this study were extracted 5 years after the release from FDNPP; a release that was not continuous. Similar to the White Oak Lake and Pond B studies, Inkyozaka had an average of 76% of the ^{137}Cs in the cores concentrated in the top 5 cm. Each core location decreased in an expected fashion with the highest activity concentration at the surface of the sediments, as seen in Figure 11. The surface sediments contained around $5.0 \times 10^{+04}$ Bq/kg and reached the lower end around 20 cm containing 10 Bq/kg. Inkyozaka is also the closest pond to FDNPP. However, this was not the case for Suzuuchi, Funasawa, and Kashiramori; all of which had less than 50% of the total ^{137}Cs from their cores in the top 5 cm. A possible reason for the discrepancy may be that the higher concentrations of ^{137}Cs is migrating deeper into the sediment layers. Research on Chernobyl-sourced ^{137}Cs in Finnish lakes showed that, generally, the peak concentration of ^{137}Cs will migrate further into the sediment layers over time (Ilus & Saxen, 2005). Sediment cores of 30 cm depth were removed from lakes in 2000 and 2003. Given that it was 14 and 17 years after the Chernobyl accident, the goal was to determine the sedimentation rate of ^{137}Cs . The cores were separated into 1 or 2 cm segments and the peak concentration of ^{137}Cs occurred in the top 10 cm and the sedimentation rates ranged between 0.6 – 6.1 mm per

year. However, as shown in the Figures 11-14, some of the cores from Okuma had an even distribution of ^{137}Cs rather than a peak that is migrating deeper into the sediment layer.

The sediment core profiles from Suzuuchi (Figure 12) were the most even of the ^{137}Cs distributions. Overall, Suzuuchi was the most contaminated pond with surface contamination levels at $1.0 \times 10^{+05}$ Bq/kg. In the core profiles, the top 20 cm showed a nearly even distribution of radiocesium, especially in the first core location. Suzuuchi core 1 was taken from a platform located at the mouth of the stream that feeds the pond, shown in Figure 3. Rather than a cesium peak, as described in Ilus and Saxon, the profiles from Suzuuchi suggest either a mixing of the sediment layer or a steady reintroduction of new contaminated sediments from the external environment. The Finnish lake study had cores, lake Keurusselka, with one unusually broad cesium peak, indicating the concentration of ^{137}Cs was roughly even across the top 10 cm (Ilus & Saxen, 2005). The authors attributed the unusual data to a mixing of the sedimentation layers or potentially a failed sample extraction. Mixing can occur from possible bioturbation from benthic creatures or misuse of the coring tool. Another lake, Kuerevesi, had a core with a cesium peak at 23 cm; significantly further down than the other lakes. Lake Kuerevesi contained a pump mill that regularly discharged fresh material to the pond, thereby providing a new source of clean sediment. The sedimentation rate for lake Kuerevesi was 16 mm per year (Ilus & Saxen, 2005). In that Finnish lake, the new material that was introduced was not contaminated with ^{137}Cs . However, new material from the watershed surrounding the ponds in Okuma, Japan would be contaminated with ^{137}Cs . The reintroduction of new contaminants from the stream entering Suzuuchi is likely responsible for adding new layers of contaminated sediments, thus explaining the even distribution of ^{137}Cs .

Funasawa was the second most contaminated pond after Suzuuchi; located in the center of Okuma. As shown in Figure 13, the sediment core profiles extracted from Funasawa exhibited similar even ^{137}Cs distributions to Suzuuchi. 41% of the radiocesium in Funasawa was concentrated in the top 5 cm, also lower than the 65% or 75% seen in previous studies (Lomenick & Gardiner, 1965) (Pinder III, Bowling, Lide, & Beatty, 1995). While not as dramatic, the cores taken from Funasawa generally had an even distribution of ^{137}Cs in the top 15 cm. Similar to Suzuuchi, there was a drainage flow system seen running in and out of the pond. Funasawa was located in the center of Okuma and was meant to be a recreation and drainage pond for the town. The presence of the drainage system reinforces the assumption that reintroduction of new contaminated materials is the reason that the top layers of the core have an even distribution of ^{137}Cs .

Although there appears to be environmental sources of mixing in the surface sediments of these irrigation ponds, potential bias may occur from the core sampling technique as well and must be discussed. Two areas of bias that may have effected this study are post core extraction mixing and core shortening. Post extraction mixing involves the redistribution, resuspension, or loss of materials of interest in the core (Blomqvist, 1991). The causes of such mixing are diverse. When placing and removing a core sample, the tube must remain vertical. Placing the tube at an angle, such as during removal, may result in redistribution of materials, especially in the uppermost layer of the core where the density of the sediment is lower. Similarly, twisting the core tube during removal, a method almost required when removing by hand as the cores were in this experiment, can result in shear stress on the entrapped sediments. Core shortening is an issue that occurs when the extracted core is shorter than the penetration depth of the tube and may skew results when representing data by core depth, as in this study (Blomqvist, 1991). This

phenomenon occurs when friction between the sediment and the inner wall of the tube cause thinning towards the center of the core. The resultant thinning can lead to compaction of the sediment core.

Mitigating sources of bias in coring can be difficult, especially as core techniques such as the one used in this study are limited by feasibility. Care was taken to ensure that the cores from the target irrigation ponds were placed and driven vertically. However, twisting to remove the core was required when pulling the core out of the sediment. A common technique to mitigate the issues that result from core extraction is freezing the sediments in the core (Blomqvist, 1991). However, core freezing was not feasible in this study due to limitations of technique and materials. There was not enough time to develop a technique to freeze the cores when they are taken from an average depth of 1 m from a boat. In-core mixing may have occurred due to agitating the sediments during transportation, by van, from the field to the laboratory. Agitation may have been avoided by segmenting the cores in the field, directly after extraction. Limitations on the amount of time allowed in the Fukushima deposition zone did not allow for in-field slicing for this study. However, in future studies could allow for time as part of the planned methods. There are analytical techniques to evaluate the extent of post extraction mixing as well, such as dating the sediments by layer (Van Metre, Wilson, Fuller, Callender, & Mahler, 2004). These methods may be useful in future studies to draw conclusions about the degree of in-core mixing.

Overall, the unusual characteristics of the pond core profiles would indicate that the vertical distribution of radiocesium in these ponds is more mobile than expected. The causes of such distribution could be ecological in origin, such as introduction of fresh material from

outside the ponds or environmental mixing. However, to some extent, the core technique also contributed to the results and must be considered.

4.2 Solid-Liquid Distribution Coefficient

Values for the distribution coefficients were generally on the order of 10^4 or 10^5 (L/kg). Inkyozaka contained the lowest average K_d values, while Kashiramori contained the highest. Previous research on rivers in the FDNPP deposition zone found similar results (Konoplev A. , Distribution of Radiocesium of Accidental Origin between the Suspended Alluvium and Solution in Rivers: a Comparison of Fukushima and Chernobyl, 2015). The sediment compositions of the irrigation ponds and rivers in the Fukushima watershed must be very similar. In comparison, large, pre-alpine lakes were studied in Germany after the Chernobyl accident (Konoplev, et al., 2002). Lakes Constance and Lugano, in that study, had a higher K_d than the Fukushima irrigation ponds by roughly one order of magnitude.

4.3 Cation Measurements

Primary cations of interest are K^+ (potassium) and NH_4^+ (ammonium) since they are the primary competitors to ^{137}Cs in soils and sediments. Stable cesium in the form of ^{133}Cs is also of interest because it is 1000 times more competitive than ammonium and 200 times more competitive than potassium. All 3 minerals bind in place of ^{137}Cs and high levels would indicate that radiocesium is not well bound and is bioavailable. The results are shown in Table 2. NH_4^+ was below the detectable limit in almost all samples except the samples taken from Suzuuchi in June and July 2016. In addition, Suzuuchi had the highest measured levels of K^+ , on average .

Collectively, this indicates that the ^{137}Cs present in that pond is more bioavailable than the others. Inkyozaka had the highest measurable levels of ^{133}Cs .

Comparable measurements were made in 2015 (Konoplev A. , et al., 2015). Dissolved K^+ , NH_4^+ , Na^+ , Ca^{2+} , Mg^{2+} in the pond water was measured by ion chromatography and $^{133}\text{Cs}^+$ (stable cesium) was measured by ICP/MS. Table 5 shows the results from data collected in 2015 compared to the average results from the data collected in 2016 (shown in parenthesis).

Table 5: Hydrochemical Data from Okuma Ponds collected in 2015 with 2016 data shown in parenthesis

Parameter	Inkyozaka	Suzuuchi	Funasawa	Kashiramori
$^{133}\text{Cs}^+$ (ng/L)	12.5 (32)	10 (20)	13 (28)	13 (25)
K^+ (mg/L)	0.66 (0.79)	2.1 (2.6)	1.3 (2.0)	1.1 (1.1)
NH_4^+ (mg/L)	N/D (N/D)	0.17 (0.08)	0.9 (N/D)	1.4 (N/D)
pH	7.1 (8.05)	7.2 (7.73)	7.8 (8.12)	8.3 (8.17)

Of note, the highest potassium concentrations were found in Suzuuchi (2.1 mg/L) and the highest ammonium concentrations were observed in Kashiramori (1.4 mg/L). The highest concentrations for ^{133}Cs were identified in Funasawa and Kashiramori, however, the concentrations are very low across all four ponds. Therefore, the impact of the low concentrations of $^{133}\text{Cs}^+$ is minimal on this study.

4.4 Sequential Extractions

Sequential extractions are a method for examining the binding potential of contaminants in soils and sediments. By using a solvent, materials in a sample are dissolved and the remaining fluid can be measured. The goal of the extraction is to observe what is liberated from the sample materials by the solvent. In this study, ammonium acetate ($\text{CH}_3\text{COONH}_4$) is used to dissolve sediments to find the fraction of bound ^{137}Cs . A two-extraction method was used to determine the fractions of bound radiocesium to inorganic materials such as clays and organic materials like

surface debris (Tsukada, Takeda, Hisamatsu, & Inaba, 2007). The first extraction was completed by an ammonium acetate solution that would break down inorganic materials. The result is a fraction of the measured ^{137}Cs in the remaining residue against the total from the sample seen in Figure 16. The highest extraction ratios, an average of 40%, were observed in Inkyozaka. Extraction ratios for the other ponds were $\sim 21\%$ in contrast.

The second extraction was completed by suspending the remaining sediment from the first extraction in pure water and adding hydrogen peroxide (H_2O_2) and nitric acid (HNO_3) to break down any organic materials. After cooling, the sediments were extracted again with an ammonium acetate solution with 20% nitric acid. The remaining fluid was measured by HPGe to show the ratio of ^{137}Cs that was bound in the organic materials versus the total in the original sample. Overall, the extracted fractions from organic materials were much lower than the fractions from inorganic matter. The difference shows that ^{137}Cs is generally more bound to the inorganic clays than it is to the debris that has settled at the bottom of the pond. Again, the fraction of extracted ^{137}Cs was highest in Inkyozaka, on average of 13.7%. The extracted average from the other ponds was roughly 9%. Overall, extraction rates were low; averaging a total of 30% - 40%. The indication is that there must be significant quantities of ^{137}Cs either remaining in the sediment sample or lost in the extraction process. The two-extraction technique was developed to examine fallout ^{137}Cs in agricultural soils prior to the accident at FDNPP and had a total extraction rate of roughly 90% (Tsukada, Takeda, Hisamatsu, & Inaba, 2007). The results from the irrigation pond sediments suggest that the technique may not be completely effective for these types of soils. Future studies may use higher stage extraction techniques to attempt to remove a greater percentage of ^{137}Cs .

CONCLUSION

The objective of this study was to assess the extent of ^{137}Cs contamination in irrigation ponds effected by the release of radioactive materials from FDNPP. From examining the activity concentration in sediment cores and associated water chemistry, ^{137}Cs is appears to be very mobile in the top layers of pond sediments. In particular, Suzuuchi and Funasawa showed significant concentrations of ^{137}Cs in the top 20 cm of sediments. The migration of ^{137}Cs beyond the top 5 cm is likely due to agitation from external sources; either biological or ecological. However, sources of bias due to post extraction mixing are considered to have an impact on the results. Determination of the extent of post extraction mixing and mitigation methods should be strongly considered for future sediment studies. The observation of streams drawing water in and out of the ponds suggested that they are more open than closed water bodies, which is supported by the measured K_d values. When compared to previous research, the K_d values for the ponds in this study were consistent to the values found in Fukushima rivers and somewhat lower than values found in large lakes contaminated by the Chernobyl accident. Comparing to the K_d values of Fukushima rivers suggests the ponds in Okuma have comparable suspended sediment types and water chemistry. Rivers are open water bodies, by definition, and the comparison to the irrigation ponds of this study suggests they are open to some degree as well.

The exchange of radiocesium in the ponds examined in this study poses a challenge to regional agriculture and obstructs contamination clean-up efforts. Japan has a limit on contamination in rice of 100 Bq/kg (Fujiwara, et al., 2012). The irrigation ponds were designed to support the widespread land use for rice cultivation. The significant mobilization of radiocesium to and from these ponds ensures continuous contamination of the rice paddies as

well. Similarly, the ponds in this study do not adhere to the strict definition of an environmental “sink” for contaminants as outlined by Ward Whicker (Whicker & Schultz, 1982). While radiocesium from the ecosystem may accumulate in these ponds, some fraction might migrate out of the ponds and, potentially, to rice paddies. Radiocesium migration in this way can be very obstructive to clean up efforts, potentially re-contaminating rice paddies after clean-up efforts have been attempted. Future work would involve mathematical modelling of the wider watershed. Of particular interest to the Japanese is the ratio of radiocesium that is exchanged from the irrigation ponds to the rice paddies. Successful modeling of radioactive material exchange in the watershed might also be applied to other ponds, which are numerous in the affected areas of Fukushima Prefecture.

REFERENCES

1. Baba, M. (2013). Fukushima Accident: What Happened. *Radiation Measurements*(55), 17-21.
2. Baum, E., Ernesti, M., Knox, H., Miller, T., & Watson, A. (2010). *Nuclides and Isotopes: Chart of the Nuclides* (7th Edition ed.). Schenectady, NY, USA: Knolls Atomic Power Laboratory.
3. Blomqvist, S. (1991). Quantitative Methods of Soft-Bottom Sediments: Problems and Solutions. *Marine Ecology Progress Series*, 72, 295-304.
4. Brookhaven National Laboratory. (2017, February 06). *National Nuclide Data Center*. Retrieved from National Nuclide Data Center:

<http://www.nndc.bnl.gov/nudat2/decaysearchdirect.jsp?nuc=137CS&unc=nds>
5. Bulgakov, A., Konoplev, A., Smith, J., Hilton, J., Comans, R., Laptev, G., & Christyuk, B. (2002). Modeling the Longterm Dynamics of Radiocesium in Closed Lakes. *Journal of Environmental Radioactivity*(61), 41-53.
6. Cremers, A., Elsen, A., De Preter, P., & Maes, A. (1988). Qunatitative analysis of radiocesium retention in soils. *Nature*, 355(15), 247-249.
7. Fujiwara, T., Saito, T., Muroya, Y., Sawahata, H., Yamashita, Y., Nagasaki, S., . . . Tanaka, S. (2012). Isotopic Ratio and Vertical Distribution of Radionuclides in Soil affected by the Accident of Fukushima Dai-ich Nuclear Power Plant. *Journal of Environmental Radioactivity*(113), 37-44.
8. Ilus, E., & Saxen, R. (2005). Accumulation of Chernobyl-derived ¹³⁷Cs in bottom sediments of some Finnish lakes. *Journal of Environmental Radioactivity*(82), 199-221.

9. International Atomic Energy Agency. (2017, 01 28). *Fission Product Yields*. Retrieved from IAEA.org: <https://www-nds.iaea.org/sgnucdat/c3.htm>
10. Konoplev, A. (2015). Distribution of Radiocesium of Accidental Origin between the Suspended Alluvium and Solution in Rivers: a Comparison of Fukushima and Chernobyl. *Radiochemistry*, 57(5), 552-556.
11. Konoplev, A., Golosov, V., Laptev, G., Nanba, K., Onda, Y., Takase, T., . . . Yoshimura, K. (2016). Behavior of accidentally released radiocesium in soil-water environment: Looking at Fukushima from a Chernobyl perspective. *Journal of Environmental Radioactivity*(151), 568-578.
12. Konoplev, A., Golosov, V., Nanba, K., Omine, K., Onda, Y., Takase, T., . . . Kivva, S. (2015). Radiocesium solid-liquid distribution and migration in contaminated areas after the accident at the Fukushima Dai-ichi Nuclear Power Plant. *Proceedings on the Environment*, (pp. 21-25). Thessaloniki.
13. Konoplev, A., Kaminski, S., Klemt, E., Konopleva, I., Miller, R., & Zibold, G. (2002). Comparative Study of ^{137}Cs partitioning between solid and liquid phases in Lake Constance, Lugano, and Vorse. *Journal of Environmental Radioactivity*(55), 1-11.
14. Kortov, V., & Ustyantsev, Y. (2013). Chernobyl Accident: Causes, Consequences and Problems of Radiation Measurement. *Radiation Measurements*, 12-16.
15. Lomenick, T., & Gardiner, D. (1965). The Occurance and Retention of Radionuclides in the Sediments of White Oak Lake. *Health Physics*, 11, 567-577.
16. Ministry of Agriculture, Forestry, and Fisheries. (2015). *About Actual Situation of Radionuclides in Ponds etc: an Overview of Investigation Results obtained by a Demonstration Project for Countermeasures against Expanding Contamination in Ponds*

etc. (in Japanese). Retrieved from

http://www.maff.go.jp/j/kanbo/joho/saigai/pdf/tameike_tyousa26.pdf

17. NCRP 154. (2006). *Cesium-137 in the Environment: Radioecology and Approaches to Assessment and Management*. Bethesda: National Council on Radiation Protection and Measurements.
18. Pinder III, J., Bowling, J., Lide, R., & Beatty, L. (1995). The Distribution of ¹³⁷Cs in Sediments in the Littoral Zone of a former Reactor Cooling Pond. *Journal of Environmental Radioactivity*, 28(1), 57-71.
19. Steinhäuser, G., Brandl, A., & Johnson, T. (2014). Comparison of the Chernobyl and Fukushima nuclear accident: A review of the environmental impacts. (470-471), 800-817.
20. Tsukada, H., Takeda, A., Hisamatsu, S., & Inaba, J. (2007). Concentration and specific activity of fallout ¹³⁷Cs in extracted and particle-size fractions of cultivated soils. *Journal of Environmental Radioactivity*(99), 875-881.
21. Van Metre, P., Wilson, J., Fuller, C., Callender, E., & Mahler, B. (2004). *Collection, Analysis, and Age-Dating of Sediment Cores From 56 U.S. Lakes and Reservoirs Sampled by the U.S. Geological Survey, 1992–2001*. U.S. Geological Survey. Reston: U.S. Geological Survey.
22. Whicker, F. W., & Schultz, V. (1982). *Radioecology: Nuclear Energy and the Environment*. Boca Raton, Florida: CRC Press Inc.

APPENDIX 1: Raw Data from Sediment Cores

5.1 Inkyozaka

Core ID	Depth	Activity (Bq/kg)
Inkyozaka Core 1	0-2.5 cm	3.51E+04
Inkyozaka Core 1	2.5-5 cm	1.56E+04
Inkyozaka Core 1	5-7.5cm	1.98E+04
Inkyozaka Core 1	7.5-10cm	9.62E+03
Inkyozaka Core 1	10-15 cm	7.52E+02
Inkyozaka Core 1	15-20 cm	1.42E+01
Inkyozaka Core 1	20-30 cm	3.38E+00

Core ID	Depth	Activity (Bq/kg)
Inkyozaka Core 2	0- 2.5 cm	1.99E+04
Inkyozaka Core 2	2.5-5 cm	1.25E+04
Inkyozaka Core 2	5-7.5 cm	9.48E+03
Inkyozaka Core 2	7.5-10 cm	4.97E+03
Inkyozaka Core 2	10-15 cm	8.99E+02
Inkyozaka Core 2	15-20 cm	3.39E+01
Inkyozaka Core 2	20-27 cm	6.33E+01

Core ID	Depth	Activity (Bq/kg)
Inkyozaka Core 3	0- 2.5 cm	2.14E+04
Inkyozaka Core 3	2.5-5 cm	1.25E+04
Inkyozaka Core 3	5-7.5 cm	6.65E+02
Inkyozaka Core 3	7.5-10 cm	9.92E+01
Inkyozaka Core 3	10-15 cm	7.75E+01
Inkyozaka Core 3	15-20 cm	5.01E+01
Inkyozaka Core 3	20-25 cm	2.27E+01
Inkyozaka Core 3	25-30 cm	9.97E+00
Inkyozaka Core 3	30-40 cm-A	2.11E+00
Inkyozaka Core 3	40-47 cm	1.21E+01

5.2 Suzuuchi

Core ID	Depth	Activity (Bq/kg)
Suzuuchi Core 1	0- 2.5 cm	5.07E+04
Suzuuchi Core 1	2.5-5 cm	4.08E+04
Suzuuchi Core 1	5-7.5 cm	2.80E+04
Suzuuchi Core 1	7.5-10 cm	2.51E+04
Suzuuchi Core 1	10-15 cm	3.24E+04
Suzuuchi Core 1	15-20 cm	3.45E+04
Suzuuchi Core 1	20-25 cm	3.12E+04
Suzuuchi Core 1	25-30 cm	2.18E+04
Suzuuchi Core 1	30-40 Cm	2.69E+04
Suzuuchi Core 1	40-50 cm	2.50E+04

Core ID	Depth	Activity (Bq/kg)
Suzuuchi Core 2	0- 2.5 cm	8.01E+04
Suzuuchi Core 2	2.5-5 cm	8.95E+04
Suzuuchi Core 2	5-7.5 cm	8.36E+04
Suzuuchi Core 2	7.5-10 cm	5.16E+04
Suzuuchi Core 2	10-15 cm	3.75E+04
Suzuuchi Core 2	15-20 cm	8.64E+03
Suzuuchi Core 2	20-25 cm	8.11E+02
Suzuuchi Core 2	25-30 cm	1.01E+02
Suzuuchi Core 2	30-35 cm	1.91E+02
Suzuuchi Core 2	35-40 cm	2.99E+01

Core ID	Depth	Activity (Bq/kg)
Suzuuchi Core 3	0- 2.5 cm	1.45E+05
Suzuuchi Core 3	2.5-5 cm	1.57E+05
Suzuuchi Core 3	5-7.5 cm	1.53E+05
Suzuuchi Core 3	7.5-10 cm	1.53E+05
Suzuuchi Core 3	10-15 cm	1.71E+05
Suzuuchi Core 3	15-20 cm	1.92E+05
Suzuuchi Core 3	20-25 cm	1.17E+05
Suzuuchi Core 3	25-30 cm	8.95E+03
Suzuuchi Core 3	30-40 cm	4.24E+02
Suzuuchi Core 3	40-50 cm	5.46E+01

5.3 Funasawa

Core ID	Depth	Activity (Bq/kg)
Funasawa Core 1	0- 2.5 cm	9.60E+04
Funasawa Core 1	2.5-5 cm	9.13E+04
Funasawa Core 1	5-7.5 cm	1.03E+05
Funasawa Core 1	7.5-10 cm	1.13E+05
Funasawa Core 1	10-15 cm	1.21E+05
Funasawa Core 1	15-20 cm	8.16E+04
Funasawa Core 1	20-25 cm	1.27E+04
Funasawa Core 1	25-30 cm	4.66E+02
Funasawa Core 1	30-36 cm	5.70E+01

Core ID	Depth	Activity (Bq/kg)
Funasawa Core 2	0- 2.5 cm	7.52E+04
Funasawa Core 2	2.5-5 cm	6.03E+04
Funasawa Core 2	5-7.5 cm	7.15E+04
Funasawa Core 2	7.5-10 cm	7.14E+04
Funasawa Core 2	10-15 cm	5.62E+04
Funasawa Core 2	15-20 cm	1.97E+04
Funasawa Core 2	20-25 cm	1.34E+03
Funasawa Core 2	25-30 cm	5.36E+01
Funasawa Core 2	30-40 cm	2.57E+01
Funasawa Core 2	40-50 cm	2.88E+01
Funasawa Core 2	50-56 cm	4.73E+02

Core ID	Depth	Activity (Bq/kg)
Funasawa Core 3	0- 5 cm	6.94E+04
Funasawa Core 3	5-10 cm	5.15E+04
Funasawa Core 3	10-15 cm	4.89E+03
Funasawa Core 3	15-20 cm	4.40E+02
Funasawa Core 3	20-25 cm	3.85E+02
Funasawa Core 3	25-30 cm	3.92E+02
Funasawa Core 3	30-40 cm	3.06E+02
Funasawa Core 3	40-50 cm	1.70E+02

5.4 Kashiramori

Core ID	Depth	Activity (Bq/kg)
Kashiramori Core 1	0- 2.5 cm	1.29E+04
Kashiramori Core 1	2.5-5 cm	1.35E+04
Kashiramori Core 1	5-7.5 cm	1.38E+04
Kashiramori Core 1	7.5-10 cm	1.21E+04
Kashiramori Core 1	10-15 cm	1.36E+04
Kashiramori Core 1	15-20 cm	1.74E+03
Kashiramori Core 1	20-25 cm	1.57E+02
Kashiramori Core 1	25-30 cm	1.05E+02
Kashiramori Core 1	30-40 cm	2.89E+01
Kashiramori Core 1	40-50 cm-A	2.88E+00
Kashiramori Core 1	50-63 cm	2.27E+00

Core ID	Depth	Activity (Bq/kg)
Kashiramori Core 2	0- 2.5 cm	9.62E+03
Kashiramori Core 2	2.5-5 cm	8.14E+03
Kashiramori Core 2	5-7.5 cm	7.47E+03
Kashiramori Core 2	7.5-10 cm	1.85E+03
Kashiramori Core 2	10-15 cm	8.05E+01
Kashiramori Core 2	15-20 cm	7.13E+00
Kashiramori Core 2	20-25 cm	7.56E+00
Kashiramori Core 2	25-33 cm	7.17E+00

Core ID	Depth	Activity (Bq/kg)
Kashiramori Core 3	0- 2.5 cm	7.99E+03
Kashiramori Core 3	2.5-5 cm	8.22E+03
Kashiramori Core 3	5-7.5 cm	8.81E+03
Kashiramori Core 3	7.5-10 cm	1.10E+04
Kashiramori Core 3	10-15 cm	8.16E+03
Kashiramori Core 3	15-20 cm	2.64E+02
Kashiramori Core 3	20-25 cm	1.04E+02
Kashiramori Core 3	25-30 cm	2.55E+01

APPENDIX 2: Sediment Densities by Depth

Inkyozaka Core Densities (g/cm ³)			
Depth	Inkyozaka 1	Inkyozaka 2	Inkyozaka 3
2.5	0.621	0.888	0.969
5	1.493	1.204	1.419
7.5	1.570	1.445	1.379
10	1.513	1.463	1.465
15	1.675	1.522	1.469
20	1.563	1.576	1.574
25		1.472	1.558
30	1.235		1.537
40			1.369

Suzuuchi Core Densities (g/cm ³)			
Depth	Suzuuchi 1	Suzuuchi 2	Suzuuchi 3
2.5	1.184	0.354	0.345
5	1.223	0.466	0.319
7.5	1.581	0.540	0.292
10	1.571	0.685	0.288
15	1.405	0.823	0.313
20	1.300	0.690	0.377
25	1.385	0.549	0.477
30	1.471	0.848	0.679
40	1.217	1.257	0.515
50	1.042		0.508

Funasawa Core Densities (g/cm³)			
Depth	Funasawa 1	Funasawa 2	Funasawa 3
2.5	0.628	0.422	
5	0.653	0.493	0.409
7.5	0.709	0.656	
10	0.719	0.672	0.872
15	0.835	0.719	0.683
20	0.894	0.768	0.626
25	1.465	0.862	0.571
30	1.754	0.943	0.606
40	1.543	0.838	0.617
50		0.797	1.252

Kashiramori Core Densities (g/cm³)			
Depth	Kashiramori 1	Kashiramori 2	Kashiramori 3
2.5	0.576	0.711	0.862
5	0.585	0.894	0.845
7.5	0.589	0.874	0.953
10	0.753	0.983	0.926
15	0.823	0.958	0.916
20	0.877	1.085	0.822
25	0.696	1.162	0.861
30	0.404	1.687	0.979
40	0.856		1.049
50	0.889		0.936
60			1.185

APPENDIX 3: Raw Data for Solid-Liquid Distribution Coefficient Calculation

Inkyozaka	Suspended Sediments (Bq/kg)	Dissolved ¹³⁷ Cs (Bq/L)	Distribution Coefficient (L/kg)
6/9/16	1.72E+05	2.06E+00	8.34E+04
7/6/16	2.31E+05	1.94E+00	1.19E+05
8/8/16	1.78E+05	2.32E+00	7.67E+04

Suzuuchi	Suspended Sediments (Bq/kg)	Dissolved ¹³⁷ Cs (Bq/L)	Distribution Coefficient (L/kg)
6/9/16	3.30E+05	5.06E+00	6.53E+04
7/6/16	4.41E+05	3.85E+00	1.15E+05
8/8/16	2.97E+05	2.35E+00	1.27E+05

Funasawa	Suspended Sediments (Bq/kg)	Dissolved ¹³⁷ Cs (Bq/L)	Distribution Coefficient (L/kg)
6/9/16	3.73E+05	2.37E+00	1.57E+05
7/6/16	1.94E+05	1.91E+00	1.02E+05
8/8/16	2.70E+05	4.24E+00	6.38E+04

Kashiramori	Suspended Sediments (Bq/kg)	Dissolved ¹³⁷ Cs (Bq/L)	Distribution Coefficient (L/kg)
6/9/16	5.87E+04	2.26E-01	2.60E+05
7/6/16	1.97E+04	2.48E-01	7.94E+04
8/8/16	1.66E+05	2.92E-01	5.70E+05

APPENDIX 4: Copyright Information

Figure 1 is a map created using custom location markers in Google Earth. Copyright information can be found in the link below:

<https://www.google.com/permissions/geoguidelines.html>

Per Fair Use Section:

“Apart from any license granted to you by Google, your use of the Content may be acceptable under principles of “fair use.” Fair use is a concept under copyright law in the U.S. that, generally speaking, permits you to use a copyrighted work in certain ways without obtaining a license from the copyright holder.”