RADIOCESIUM DISTRIBUTION IN RICE, WILD PLANTS, AND SOIL FOLLOWING THE FUKUSHIMA-DAIICHI NUCLEAR POWER PLANT ACCIDENT

September 2019

Nittaya Wakai

Graduate School of Environmental and Life Science

(Doctor's Course)

OKAYAMA UNIVERSITY

CONTENTS

ABSTRACT

Nittaya Wakai, 2019. Radiocesium distribution in rice, wild plants, and soil following the Fukushima-Daiichi nuclear power accident. Doctoral Thesis, Okayama University, Japan.

The accidental damage of the Fukushima-Daiichi nuclear power plant in March 2011 had released a large amount of radionuclides to the environment. One year after the accident, Yamashita et al. (2014) investigated concentrations of ^{137}Cs (half-life: 30 years) in hundred species of wild plants in agricultural lands in Iitate-village, indicating that wild plants were contaminated with ^{137}Cs. However, they did not identify factors affecting those contamination. On the other hand, removal of the soil surface seems to be an effective method for minimizing ^{137}Cs in agricultural land, but this reduces soil fertility. There are no studies available on soil improvement by cattle manure application. The aims of this research were to i) determine the ^{137}Cs contamination degree of soil and wild plants in Iitate and the neighboring villages with different land use types, ii) identify factors affecting ^{137}Cs concentration in soil and wild plants and transfer from soil to wild plants, and iii) evaluate effects of cattle manure compost application at various rates on rice yields and ^{137}Cs distribution in rice parts.

For the first and second aims, we collected soil and wild plant samples from three different land use types, including agricultural land, roadside, and a mountain in Iitate-village and the neighboring areas in 2014. Concentrations of ^{137}Cs in agricultural soil ranged between 11 and 14 kBq kg^{-1}, while that in the roadside soil with high clay content was the highest (261 kBq kg^{-1}) and that in the mountain soil with relatively high organic matter showed the lowest (6 kBq kg^{-1}). Concentrations and transfer factors of ^{137}Cs were not significantly different among wild plant species grown in the same soil, but that differed for the same species grown in different soils. Our result suggested that the potential of ^{137}Cs uptake of plants was controlled by soil factors regardless of plant species. Exchangeable K reduced ^{137}Cs uptake by wild plants. Although this is well known for crops, we found that the same mechanism would occur for wild plants.

For the third aim, an experimental field consisted of 4 plots receiving different rates of cattle manure compost (0, 10, 20, and 40 t ha^{-1}) and rice (Akitakomachi) was grown in 2015. As a result, rice yields increased with increasing rates of compost. Compost application increased exchangeable K, resulting in reduced ^{137}Cs concentration in rice. In the treatment with compost application rate of 20 t ha^{-1}, ^{137}Cs concentration and their transfer factors in rice parts were the lowest level. In this plot, exchangeable K in soil was the greatest. Concentrations of ^{137}Cs in stems were greater than in leaves and those in panicles were below the detectable level. In plots with 10 and 20 t ha^{-1} of compost, ratios of ^{137}Cs/K were greater in stems than in leaves of rice and those were related to concentrations of Ca and Mg in the soil. Those results indicated that exchangeable K, Ca and Mg derived from cattle manure compost affected the uptake and distribution of ^{137}Cs in rice.

In summary, we found that i) ^{137}Cs concentration was higher in the roadside soil than those in agriculture and mountain soils, ii) ^{137}Cs uptake by wild plants was affected by soil factors rather than plant species, and iii) decrease of ^{137}Cs in rice parts resulted from inputs of K, Mg and Ca derived from cattle manure. Information on ^{137}Cs contamination in soil and wild plants with different land use types will be useful for recovering ^{137}Cs contaminated soil in non-agricultural areas. Cattle manure compost application is effective for reducing ^{137}Cs uptake by rice in addition to soil fertility improvement.

Key words**:** cattle manure compost, ^{137}Cs, land use, rice, soil properties, wild plants

ACKNOWLEDGEMENTS

This thesis was made with support by many people. I would like to express my deepest appreciate for all. First my supervisor, Prof. Morihiro Maeda, who spent much hard time guiding the research and writing throughout my Ph.D. course. Beside my supervisor, I am so lucky to have been a student under the guidance and help of the best research team, Prof. Yoko Yamamoto, Dr. Jun Yamashita, and Dr. Takashi Enomoto, Institute of Plant Science and Resources, Okayama University, Prof. Toshiro Ono and Prof. Tadashi Hanafusa, Neutron Therapy Research Center, Okayama University, and Prof. Kuniyuki Saitoh, Graduate School of Environmental and Life Science Okayama University.

I am grateful to Prof. Mori Yasushi and Prof. Moroizumi Toshitsugu for invaluable advises to improve the thesis. Thank you to all students of Maeda Sensei`s laboratory; because of their help I had good experiences at Okayama University.

It would have been impossible to complete this thesis without the encouragement and support as all the members of my family both in Thailand and Japan. My husband Hirozo Wakai and younger sister Prakobkit Dangthaisong gave their all heart and energy to supporting me mentally and financially. My father and mother Thoey and Noknoi Dangthaisong who gave me the best life and education. My lovely children, Hirobumi, Mio, and Hiromichi, are my motivation for study and living.

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CHAPTER 1. INTRODUCTION

1.1 Background

Immediately after the Fukushima-Daiichi nuclear power plant (FD1N 1 PP) accident in March 2011, some areas of Fukushima prefecture were evacuated due to radioactive cesium (^{137}Cs) contamination. Since then the agricultural fields have been left fallow. One year later, to repair this situation and allow the people to return, minimizing ^{137}Cs in the fields by removing the contaminated soil and plants from the fields was recommended (Fukushima Environmental Safety Center, 2015); however, it was difficult to do for all fields. Information on the distribution of ^{137}Cs in soils and plants in the fields for different uses might be useful for managing the decontamination work. Previously, the ^{137}Cs contamination of almost 100 species of wild plants in agricultural fields in Iitate-Village, Fukushima, had been reported (Yamashita et al., 2014). However, the information on the factors affecting this contamination had not been defined.

In addition, the strategies for recovering the agricultural land (paddy field) for rice cropping have been recommended (MAFF, 2012). Soil surface removal seems to be an effective method to reduce ^{137}Cs in paddy fields, but this causes the loss of soil fertility. Application of organic matter effectively improved soil fertility (Nishiwaki et al., 2017), but the effect on ^{137}Cs distribution in rice plants has not been reported by previous studies.

1.2 Aims and outline of the thesis

The contamination of ^{137}Cs in land resources is a critical issue of concern after the FD1NPP accident. Wide areas of northeastern region of Japan had been found to have ^{137}Cs contamination in soil and plants. Information about the degrees of contamination in soil and plants for different types of land use, as well as the factors affecting the ^{137}Cs distribution in soil wild plant ecosystems, is necessary for future decontamination works and research.

On the other hand, studies on strategies for maintaining soil productivity and prevention of ^{137}Cs contamination of food crops are crucial.

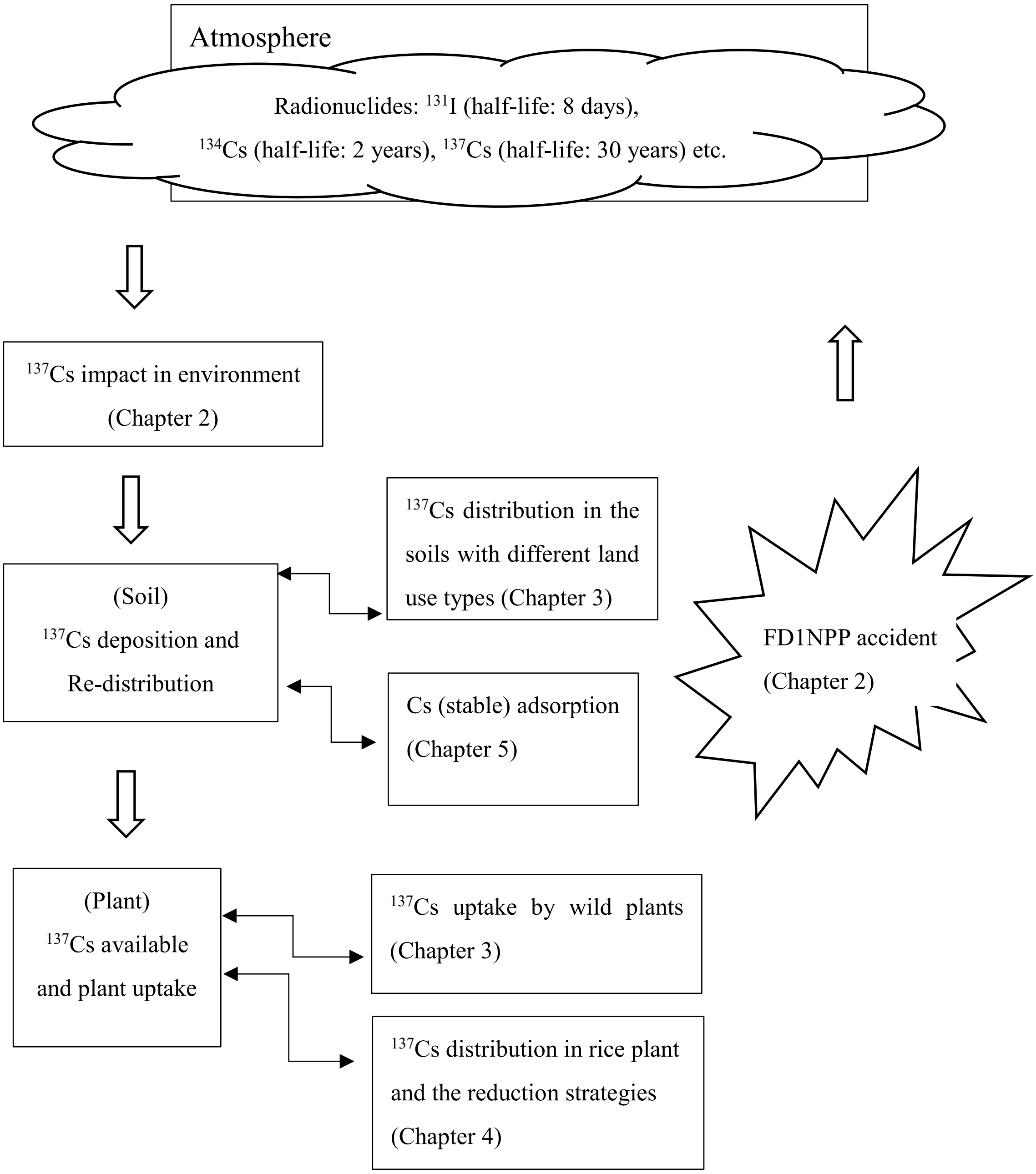

Figure 1.1 Organization of the thesis

The objectives of the present research were to:

i) estimate the degree of ^{137}Cs contamination in soil and wild plants in the different land use types and to identify the factors affecting ^{137}Cs distribution in soil and wild plant ecosystems (Chapter 3);

ii) reduction of ^{137}Cs in soil and rice through soil management (Chapter 4); and

iii) examine the factors affecting cesium adsorption in the soils from agricultural fields of Iitate-Village (Chapter 5).

Chapter 2 reviews the release of ^{137}Cs into the environment and its distribution in the fields. In this chapter, we introduced literature, that is related to our study. After the FD1NPP accident, the distribution of ^{137}Cs in the environment was extensively reviewed, mainly in the agricultural fields. However, contamination in other types of land uses have not been reported frequently. After ^{137}Cs is deposited, it can be fixed in soil layers or adsorbed onto organic matter. Re-distribution of these ^{137}Cs has been controlled by a number of factors. Mechanisms of ^{137}Cs uptake by plants have been studied widely, but the information about the effects of combined factors is limited. Decontamination of the ^{137}Cs contaminating agricultural fields for rice cultivation has been promoted, but the secondary impact has not been extensively studied.

Chapter 3 presents the results of the field survey. We found that ^{137}Cs distributions in the soil of different land uses were significantly different. The distribution of ^{137}Cs in these lands had been affected by a number of factors. Plants grown in contaminated soils play an important role as indicators of ^{137}Cs level in their respective soils. We found that different wild plants grown in same soil showed the same ^{137}Cs concentration and transfer factor (TF) level, in contrast those of the same species grown in different soils, suggesting that there is a potential for ^{137}Cs uptake by wild plants to be affected by soil factors regardless of plant species.

Chapter 4 presents the strategies for reducing ^{137}Cs in rice plants grown in paddy fields after the removal of surface soil. Cattle manure compost was used to improve soil fertility after removal of the surface soil. We found that application of cattle manure compost increased in rice yields and successfully reduced ^{137}Cs in rice plant parts. ^{137}Cs distribution in rice plants has been monitored and expressed as concentrations in rice plant parts. ^{137}Cs is transported in rice plants via the K transporter; as a result, we found the same trend for ^{137}Cs and K concentration in stems > leaves > panicles. The ^{137}Cs/K ratio in rice parts was used as an indicator of their translocation. Different ^{137}Cs/K ratios indicated the different translocation rates between ^{137}Cs and K inside rice plants. We found that the ^{137}Cs/K ratio in stems was higher than in leaves of rice grown in soil with relatively high Ca and Mg; in contrast, it was higher in leaves than in stems in soil with lower Ca and Mg, indicating that ^{137}Cs transport to upper parts of rice plants is affected by Ca and Mg in the soil.

Chapter 5 presents the results of extraction experiments that we conducted in the laboratory using stable cesium instead of radioactive cesium to examine the effects of contact times, concentrations, and competitive ions on cesium adsorption in the mineral soils. In addition, we examined the effects of soil amendment on stable cesium adsorption.

Chapter 6 discusses the factors affecting ^{137}Cs transfer in soil and plant ecosystems. After ^{137}Cs is deposited on the ground, re-distribution occurred naturally. The potentials of ^{137}Cs transportation in plants differed depending on soil properties and concentration of ^{137}Cs. In rice, ^{137}Cs transport between organs is controlled by both soil cation conditions and plant physiology.

CHAPTER 2. LITERATURE REVIEW

2.1 The FD1NPP accident

The accidental damage to the FD1NPP in March 2011 released a large amount of radionuclides including; ^{131}I, ^{132}I, ^{132}Te, ^{134}Cs, ^{137}Cs, ^{140}Ba, ^{110m}Ag, and ^{129m}Te into the environment (Chino et al., 2011, WHO, 2013). The total release of radionuclides from the FD1NPP accident was estimated at 770,000 teraBq. That was approximately 15% of those from the Chernobyl accident in 1986 (Nakashima, 2018). The radionuclides were estimated for ^{131}I (half-life: 8 days) 21%, for ^{134}Cs (half-life: 2 years) 2.3%, for ^{137}Cs (half-life: 30 years) 1.9 % (Nakashima, 2018, MEXT, 2011). Radionuclides entering the environment via direct deposition of fine particles (dry deposition) and fallout with rainfall (wet deposition) (IRSN, 2012). After the accident, the impact of ^{137}Cs in the land resources is a critical issue because its half-life is long and it can enter human foods. After the accident, Japanese authorities prohibited rice cultivation in the paddy soil contaminated with ^{137}Cs higher than 5,000 Bq kg^{-1} (30 km radius zone from FD1NPP), because a transfer factor (TF) level of 0.1 can produce a concentration of ^{137}Cs in rice products of 500 Bq kg^{-1}, the maximum level permited for Japan foods (Saito, 2012). The Ministry of Health, Labor and Welfare (MHLW) established a provisional level of 500 Bq kg^{-1} for radiocesium in cereal, vegetables, meat, and fishery products on 17 March 2011. However, on 1 April 2012, a new maximum limit of 100 Bq kg^{-1} in general foods, excluding infant food, milk, water and beverages was established (Nihei et al., 2015).

2.2 ^{137}Cs distribution in soil ecosystems

Due to the several explosions of FD1NPP, radionuclides including ^{137}Cs were dispersed into the atmosphere before falling to the ground. ^{137}Cs was deposited on the ground through both dry particles and wet fallout with rain. Patterns and rates of deposition differ depend on meteorological conditions (Chino et al., 2011). Davis (1963) reported that ^{137}Cs fallout with rainfall is more widespread than dry deposition. After ^{137}Cs is deposited, it is strongly adsorbed on soil particles, limiting its movement by chemical and biological processes. Most ^{137}Cs movement in the environment is a physical process (Ritchie and McHenry, 1990). This property of ^{137}Cs can be used as a tracer for measuring soil erosion. Soil erosion and deposition of eroded soil particles in fields, flood-plains, and water bodies are major environmental concerns around the world. Although soil erosion is a natural process, many human activities have increased the rates of soil erosion. Soil erosion reduces soil productivity, scars the landscape, and causes downstream damage (Ritchie and McHenry, 1990). Matsuda et al. (2015) reported that ^{137}Cs migration downward into the soil layer after the FD1NPP accident was estimated at 3 cm per year.

2.3 ^{137}Cs cycle in the landscape

Once ^{137}Cs is deposited on the land, its remains for a long time. ^{137}Cs is deposited in the soil through direct deposition from the atmosphere, wash-off from vegetation, turnover from vegetation, and redeposition of eroded soil particles. Movement of ^{137}Cs in chemical and biological processes is limited, and the major factors of ^{137}Cs movement in the soil are physical processes such as water and erosion (Ritchie and McHenry, 1990).

Transport of ^{137}Cs across the landscape is due to vegetation, soil, and water. ^{137}Cs is distributed on vegetation via adsorption and absorption. The adsorbed ^{137}Cs on vegetation can be washed into the soil (Davis, 1963; Dahlman et al., 1975). The ^{137}Cs in plants is returned to the soil through vegetation decomposition. Uptake of ^{137}Cs by vegetation from soils (Davis, 1963; Dahlman et al., 1975) or water (Eyman and Kevern, 1975; Garten et al., 1975) is less important than direct absorption from direct deposition on foliage (Dahlman et al., 1975). Removal of ^{137}Cs from the landscape with harvested vegetation has a small effect (Brown et al., 1981).

2.4 Decontamination of ^{137}Cs-contaminated land following the FD1NPP accident

Decontamination of the ^{137}Cs contaminated agricultural land was started in 2012. Three methods were recommended: i) removal of the surface soil, ii) separation of the fine particles, and iii) reverse tillage (MAFF, 2012). Removal of the surface soil seems to be the most effective method. According to the study of Shiozawa et al. (2011) who reported that 99% of the radiocesium was adsorbed onto a 0-7 cm layer of paddy soil in Fukushima on 24 May 2011. Similarly, Kato et al. (2012) had found that 96% of total radiocesium was adsorbed onto a 0-5 cm layer. The bases of these studies emphasized that removal of the top 15 cm layer could remove more than 80% of the radiocesium from the land (MAFF, 2013).

2.5 ^{137}C uptake by plants

Plant uptake of ^{137}Cs from soil is commonly expressed as “soil to plant transfer factor, TF” and calculated as:

$$\text{TF} = \frac{\text{Concentration of } ^{137}\text{Cs in plant (Bq kg}^{-1}\text{ weight)}}{\text{Concentration of } ^{137}\text{Cs in soil (Bq kg}^{-1}\text{ weight)}}$$

It is generally assumed that the concentration of ^{137}Cs in a plant linearly related to its concentration in the rooting zone of the soil (Ehlken and Kirchner, 2002).

Because the chemical properties of radiocesium (^{137}Cs) are similar to those of stable cesium (Cs) (Tsukada, 2002), the mechanism of stable Cs transport in soil and plant system could be explained by that for ^{137}Cs. It is well known that the mechanism of cesium uptake by plant roots is similar to that of potassium (K) (White and Broadley, 2000). Cesium is not an essential element for plants, but plants uptake cesium when the soil is deficient in potassium, through the K transporter (Avery, 1995). Distribution of Cs in plants differs among the parts (root>stem>leave>grain) and is similar with that of K (Tsukada et al., 2002a, b). An inhibiting effect of K on Cs adsorption has been reported (Smolder and Tsukada, 2011).

2.6 Cations in the soil affecting ^{137}Cs distribution in rice plant parts

Although cesium (Cs) and potassium (K) ions are similar chemically, but they have different physical characteristics. The ionic radius of Cs is 1.69 nm, which fits well with the size of the clay layer surfaces; therefore, adsorption of Cs in soil is stronger than that of K (ionic radius 1.33 nm) (Johanson et al., 2004).

After the FD1NPP accident, studies of soil management methods for reducing ^{137}Cs uptake by rice have been widely performed (Sato et al., 2012, Kato et al., 2015, Saito et al., 2015). Saito et al. (2012) reported that the radiocesium concentration of brown rice was decreased by increasing exchangeable K in the soil. Kato et al. (2015) recommended that soil with an exchangeable K concentration higher than 0.50 $\mathrm{cmol_c\ kg^{-1}}$ is the most effective for reducing ^{137}Cs uptake by lowland rice. However, the effect of other cations on ^{137}Cs uptake and distribution in rice plant parts is still unknown. Previously, some studies had reported that K concentration regimes affect the activity of the K transporter (Zhu et al. 2000). Sodium (Na) and hydrogen (H) ions inhibit K adsorption and transport mechanisms (Van et al., 1981; White, 1999). Calcium (Ca) blocks the H and Na adsorption sites in the plasma membrane of root cells, resulting in enhanced K uptake by plants (Epstein, 1961; Viets, 1999). Magnesium (Mg) plays a similar role as Ca (Epstein, 1961). According to Heredia et al. (2002), growing plants with sufficient K but low Ca enhance Cs uptake.

Overstreet et al. (1952) found that Ca plays a role in both competition and stimulation effects for K adsorption. The depressing effect of Ca on K adsorption occurs through the competition between Ca and K for binding with the metabolic compounds. The stimulation effect of Ca occurs when the K in medium is very low. This finding should be related to the transportation and accumulation of ^{137}Cs in plant parts. In addition, Smolder et al. (1997) found that only K in medium-solution had a poor effect on reducing Cs uptake by plants (spinach), but significantly reduced Cs uptake when the interaction was with Ca and Mg.

Based on these reviews, not only K but also other cations affect ^{137}Cs uptake and distribution in rice plant parts.

2.7 Rice plant physiology controlling ^{137}Cs accumulation

The ^{137}Cs in soil solution enters the root cells and translocate in plant parts via the K transport systems, it delivers to xylems through a symplastic pathway (White and Broadley, 2000). The relationship between Cs and K fluxes into the root cells is controlled by the abundance and activity of transport proteins catalyzing K, which is present in the plasma membrane of root cells. After the Cs and/or K loading in xylems they are transport upward, which K is easier loading in the vacuoles than Cs (Van et al., 1981). Plant shoot vacuoles limits for Cs accumulation, the surpluses have been loading into the phloem for recirculation for root cells (Buysse et al., 1995).

The function of vacuoles is different in the different types of tissues. The physiologically relevant functions, such as turgor control, ion selectivity, and cellular signaling have been associated with the lytic vacuoles (LV), which generally present in the vegetative tissues. The biosynthetic functions, such as storage of nutrients occur in the protein storage vacuoles (PSVs) (Marty, 1999).

According to Isayenkov et al. (2011) the K-selective cation channels present at the tonoplast of the vacuolar membrane of all plants, however different in its size for different organs. Rice (*Oryza sativa*) has been demonstrated that different types of K-selective cation channels, so call K-transporter encoding proteins (TPK). Different TPK (TPKa and TPKb) is present in different types of vacuole. There is different size for TPKa and TPKb, TPKa is larger than TPKb. The TPKa presents in the tonoplast of the lytic vacuole (LV), and the TPKb is in the tonoplast of the protein storage vacuoles (PSVs). There is no studied about the effect of these K-transporter proteins on the ^{137}Cs selective of rice parts, but this rice physiological characteristic might be involved in the different of accumulation rate between K and Cs in different parts of rice.

2.8 Radiocesium properties

Cesium (or Caesium) means blue in Latin. The symbol is Cs, atomic number: 55, atomic weight: 132.905, melting point: 28.44°C, boiling point: 671°C, and density: 1.93 g cm^{-3}. Phase at room temperature: solid, element classification: metal, period number: 6, group number: 1, group name: alkali metals.

Cesium was discovered by Robert Wilhelm Bunsen and Gustav Robert Kirchhoff, German chemists, in 1860 through the spectroscopic analysis of Durkheim mineral water. Cesium has a number of isotopes from ^{112}Cs to ^{151}Cs, but among these isotopes only ^{133}Cs is stable in nature. Cesium is chemically similar to potassium and rubidium. ^{134}Cs and ^{137}Cs are not found in nature, they are emitted during the uranium fission process. They are dangerous to living cells as emissions of β and γ radiation and have a relatively long half-life (half-life: 2.06 years for ^{134}Cs and 30 years for ^{137}Cs).

2.9 Radiocesium enters the environment

The primary source of radionuclides in the environment was the testing of nuclear weapons. The public has been exposed to these radionuclides for the past three decades or longer, but the exposure has substantially declined in the past two decades. In recent years, the major source of radionuclides was from nuclear accidents. On 28 March 1979, a very serious accident of the Three Mile Island nuclear power plant in the USA caused the release of approximately 370 PBq of ^{133}Xe (half-life: 5.3 days) and 550 GBq of ^{131}I (half-life: 8 days). Health damage due to radiation exposure has not been reported. On 26 April 1986, the most serious nuclear accident in history occurred at the Chernobyl nuclear power plant of Ukraine, and this accident released the largest amounts of long-lived-radionuclides, including ^{131}I (1,760 PBq), ^{134}Cs (47 PBq), and ^{137}Cs (85 PBq) into the environment. On 11 March 2011, an accident at the Fukushima-Daiichi nuclear power plant of Japan occurred. The accidental damage to nuclear power plants released large amounts of radionuclides into the environment (Yamada, 2012).

2.10 Effects of radiocesium to human health

Cesium (^{137}Cs) can be stored in the flesh of fish and animals. The emitted radiation (γ and β) was found to stop cell division, and this could also be used therapeutically to stop cancer. On the other hand, radiation applied locally was found to cause wounds, which are difficult to heal, and to induce cancer. The biological effect of very large whole-body doses is radiation sickness and early death, while large organ doses lead to local cell destruction and, possibly, organ death. The effects at lower doses are cell changes (decreased surviving fraction, decreased rate of division, chromosomal aberrations, etc.). The induction of cancer may take years to observe, and genetic changes may not be discovered until after several generations.

Once ^{137}Cs is distributed in the environment, it will remain hazardous for many decades.

2.11 Summary and knowledge gaps

After the Chernobyl nuclear accident, the mechanism of ^{137}Cs uptake by plants has been widely studied. In plants, ^{137}Cs uptake and transport between plant organs through the K transporter (Avery, 1995). Potassium (K) has an inhibitory effect on ^{137}Cs uptake by plant

(Zhu and Smolders, 2000; Smolders et al., 1997; White, 1999; Hampton et al., 2005). The effect of calcium (Ca) on K transport (Epstien, 1961) and effect of N on ^{137}Cs uptake (Evan and Dekker, 1969) have been reported. However, most previous studies were done on the effect of a single factor. In the present study, we found that ^{137}Cs uptake by rice and wild plants was affected by a combination of factors. On the other hand, ^{137}Cs distribution in fields with different land use types has not been studied frequently. We found that the ^{137}Cs concentration of soil from roadside was higher than those in agricultural fields and mountain soil. Re-distribution of ^{137}Cs occurs through the movement of ^{137}Cs-fixed soil particles, which are affected by rain and wind. Factors affecting ^{137}Cs distribution in the soil system had not been studied yet.

Since the FD1NPP accident, methods of reducing ^{137}Cs in rice have been widely studied. Minimizing ^{137}Cs in paddy fields through removing the surface soil was recommended. However, that caused a loss of soil fertility. Using a potassium fertilizer has successfully reduced ^{137}Cs uptake by rice (Saito et al., 2012; Kato et al., 2015; Saito et al., 2015). However, there are no available studies on soil improvement by application of cattle manure compost. The present study found that application of cattle manure compost increased soil nutrients including exchangeable K, Ca, and Mg, resulting in increased rice production and reduced ^{137}Cs concentration in rice plants.

CHAPTER 3. FACTORS AFFECTING ^{137}Cs CONCENTRATION IN WILD PLANTS AND SOILS OF DIFFERENT LAND USE IN IITATE-VILLAGE AFTER THE FUKUSHIMA NUCLEAR POWER ACCIDENT

3.1 Introduction

On 11 March 2011, a major earthquake following with a huge tsunami hit a wide area on the northeast coast of Japan. The event caused the FD1NPP to lose its reactor cooling systems, resulting in several explosions in four units of the plant. Due to this accident, a plume containing large amounts of radionuclides, including radioactive cesium (^{137}Cs), dispersed into the atmosphere (Morino et al., 2011) and then deposited on the grounds. ^{137}Cs has a long half-life (30 years) and is highly soluble in water. Cesium can enter the human body through air, soil, water, and foods (Avci, 2010).

Deposition of ^{137}Cs on the grounds was spatially heterogenous (Tanaka et al., 2013), which was caused by several factors including the length of emission time (Ohno et al., 2012), meteorological conditions, and the topography of the lands (Kinoshita et al., 2011). The ^{137}Cs can be fixed in soil layers after deposition and it can then be redistributed through the movement of soil water (Sutherland,1994). The ^{137}Cs can be taken up by plants through soil water solution. The potential of ^{137}Cs uptake for plants differ, depending on the plant species and the amount of ^{137}Cs available in the soil. The index of ^{137}Cs transferred from the soil to the plants is referred to as the transfer factor (TF) and is calculated as the ratio of the ^{137}Cs concentration in a plant to ^{137}Cs concentration in soil (Frissel et al., 2002). Higher TF values indicate greater ^{137}Cs uptake from soil by the plant.

Plants can be both indicators (Koranda and Robison, 2002) and extractors (Lasat et al., 1997) of ^{137}Cs in contaminated land. Wild plants growing in agricultural areas during ^{137}Cs emission can be both a sink and a source of ^{137}Cs within an ecosystems (Murakami et al., 2014; Ishii et al., 2017). After the FD1NPP accident, Yamashita et al. (2014) reported ^{137}Cs accumulation in wild plants and reported on the ^{137}Cs uptake of 99 species of wild plants in Iitate-village and found that the ^{137}Cs transfer factor of wild plants differed depending on the species. However, they did not mention the effect of soil conditions. In general, ^{137}Cs uptake by plants is affected by both soil conditions and plant physiology (Johanson, et al., 2004). Understanding the transportation of ^{137}Cs in soil and plant systems is necessary for future management of contaminated land. The objectives of the present study were to examine the degree of contamination in the soils under different land use types after the FD1NPP accident and to identify the factors affecting ^{137}Cs transfer in soils and wild plants.

3.2 Materials and method

3.2.1 Sampling of soils and wild plants

Six different wild plant species were collected. They expressed as their scientific name followed by their abbreviation and Japanese name in parentheses. Picture of selected plants are shown in Figure 1.

1) *Artemisia indica* Willd. var. *maximowiczii* (Nakai) H. Hara. (*Ar. indica*; Yomogi)
2) *Athyrium deltoidofrons* Makino. (*At. deltoidofrons*; Satomeshida)
3) *Athyrium yokoscense* (Franch. Et Sav. (Christ). (*At. yokoscense*; Hebinonegoza)
4) *Dryopteris tokyoensis* (Makino) C.Chr. (*D. tokyoensis*; Tanihego)
5) *Sedum sarmentosum* Bunge. (*Se. sarmentosum*; Tsurumannengusa)
6) *Solidago altissima* L. (*So. altissima*; Seitaka-awadachiso)

Five of them showed high ^{137}Cs uptake values as reported by Yamashita et al. (2014). The plant samples were randomly selected and collected by severing the aboveground parts ($n = 3$). Soil samples close to the selected plants were collected using a soil sampling scoop to a depth of 5 cm.

Figure 3.1 Picture of the selected plants

3.2.2 Locations and description of sampling points

Figure 3.2 shows the study locations. The sampling points are numbered within each location followed by an alphabetical code for the land use types. The plant name is given as an abbreviation of the scientific name.

Location 1: *Ar. indica* and *So. altissima* were sampled in a paddy field (1P) and upland field (1U). Three soil samples from each field were collected. In addition, we collected *Se. sarmentosum* and three soil samples at the roadside (1R), which is located approximately 1 km away from 1P and 1U.

Location 2: *At. deltoidofrons, D. tokyoensis*, and *So. altissima* were collected in a paddy field (2P). *At. yokoscense* in two areas along a canal ditch (2C). Three soil samples were collected at each plant sampling point.

Location 3: *At. yokoscense* and three soil samples were collected at the base of a mountain (3M).

For sites 1P and 1U, soils and plants were collected in August 2014. For sites 1R, 2P, 2C, and 3M, the samples were collected in October 2014. The fresh soil and plant samples were kept separately in polyethylene bags for transport to the laboratory.

3.2.3 Samples preparation

From each soil sample, the moisture soil was used for inorganic nitrogen analyses. The rest was air dried at room temperature for one week, and then sieved through a 2 mm mesh before the chemical and physical property analyses and radioactivity measurements. The results are given on the oven dried (105 °C for 48 hrs) weight basis.

Plant samples were oven dried at 60°C for five days and then kept in a desiccator until use. The dried samples were ground to the powder with a Waring blender (J-spec Blender

7011JBB, Waring Commercial, Torrington, USA) using a 250 mL jar set (BC250) before radioactivity measurements.

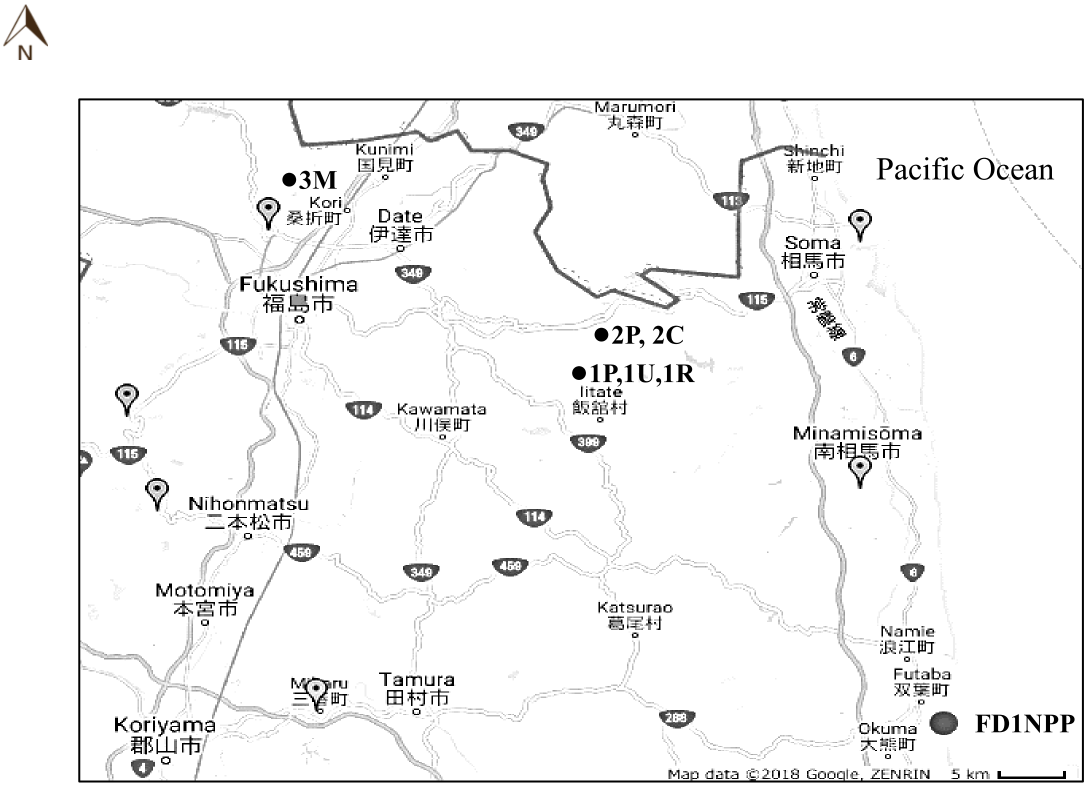

Figure 3.2 Study locations and sampling points

(Source: google map (2018))

3.2.4 Soil analysis

Soil texture fractionation was done by the pipet method (Nakai 1997). Inorganic nitrogen (NO_3^- and NH_4^+) in the moist soils was extracted with 2 *M* KCl at the ratio of 1:10 (soil: solution). We filtered the supernatant through a 0.2 µm membrane before analyzing it with the continuous flow analyzer (QuAAtro-HR, BL-TEC Autoanalyzer, Osaka, Japan). We determined the soil cation exchange capacity (CEC) and exchangeable potassium (K) using Kamewada's method (Kamewada, 1997), and measured the total carbon (TC) and total nitrogen (TN) with a CN coder (MT-700 Mark II, Yanaco, Kyoto, Japan). Soil organic matter (SOM) was calculated with the formula TC × 1.724 (Périé et al., 2008; Jiménez et al., 1992).

3.2.5 Radioactivity measurement for soil and wild plant

To measure ^{137}Cs radioactivity in the soil and plant samples, we used a Ge semiconductor detector (GMX 15200P, Seiko EG&G, Tokyo Japan). Samples were prepared for radioactivity measurement by adding a dried sample of each to a plastic container (U8 container, 48 mm in diameter and 58 mm high, Umano Kagaku Youki Co. Ltd., Osaka, Japan) to more than 10 mm deep. Each sample container was wrapped in a polyethylene bag before placing it into the Ge semiconductor. Measuring time was 30 minutes for each sample. Data analysis was performed using Gamma Studio Software (Seiko EG & G). The detection efficiency of the Ge detector was determined using the nine nuclides mixed activity standard including ^{137}Cs (Japan Radioisotope Association, Tokyo, Japan). The soil to plant transfer factor (TF) was calculated with the following equation.

$$\mathrm{TF} = \frac{^{137}\mathrm{Cs\ concentration\ in\ plant\ (Bq\ kg^{-1})}}{^{137}\mathrm{Cs\ concentration\ in\ soil\ (Bq\ kg^{-1})}}$$

3.3 Results and discussion

3.3.1 ^{137}Cs concentration in soils with different land use types

Table 3.1 shows that ^{137}Cs concentration in soil was highest in 1R and lowest in 3M, whereas that in agricultural lands was in the range of 10 to 14 kBq kg^{-1}. Different ^{137}Cs concentration in these soils was presumably because the effect of combination factors involving soil properties and the slope of the landscape.

The highest level of ^{137}Cs in the soil of 1R was possibly due to the high ^{137}Cs-fixed clay particles accumulating on the roadside. Tanaka et al. (2013) reported that ^{137}Cs was emitted from the FD1NPP as water-soluble fractions. After deposition, the ^{137}Cs became fixed in clay particles. Sutherland (1994) reported that ^{137}Cs fixation in soil particles was irreversible, and redistribution of ^{137}Cs fixed particles occurred naturally through soil erosion. Similarly, Richie and McHenry (1990) reviewed that ^{137}Cs adsorbed on soil particle is strong and limiting its movement by chemical and biological, and this property was used as the tracer for measuring soil erosion and sedimentation. Our result can be explained that the ^{137}Cs-eroded particles had deposited at the roadside.

In contrast, soil from 3M had a high clay but exhibited the lowest concentration of ^{137}Cs. This was presumably due to the soil containing high SOM and was located on the slope landscape. According to Rigol et al. (2002), ^{137}Cs adsorbs to SOM as a weak interaction, resulting in enhanced removal through leaching. Koarashi et al. (2016) found that after the FD1NPP accident, the major portions of ^{137}Cs were retained in the organic layers of forest soil. The lowering of ^{137}Cs in this was affected by the combination of a weak adsorption of ^{137}Cs on SOM and leaching. The ^{137}Cs concentration in agricultural lands (1P, 1U, 2P, and 2C) should reflect the initial amount of deposition, as these lands were less affected by leaching through rain water.

3.3.2 Soil properties affecting ^{137}Cs uptake by wild plants

The ^{137}Cs concentration in the plants was positively correlated with that in the soil. In contrast, the transfer factor (TF) value of the plants did not follow the same trend (Figure 3.3), indicating that the availability of ^{137}Cs to plants was affected by other soil factors. In the present study, however, the TF of wild plants grown in soil with high exchangeable K tended to have low TF values (Table 3.2 and Figure 3.4). Table 3.2(a) shows that ^{137}Cs was not detected in either *Ar. indica* or *So. altissima*. from 1P and 1U, in which soils contained 1.3 and 2.0 cmol$_c$ kg^{-1} of exchangeable K, respectively. It is well known that exchangeable K strongly affects ^{137}Cs uptake by plants (Smolders et al., 1997; Zhu and Smolders, 2000).

According to Frissel et al. (2002) soil media with exchangeable K lower than 0.5 cmol$_c$ kg^{-1} allow high ^{137}Cs uptake by plants. However, the present study results showed that plant did not uptake ^{137}Cs in soil containing high K content.

Table 3.1 ^{137}Cs concentration and property of soils of different land use types.

Sampling points	Land use types	Number of plant species (*n*)	Number of soil samples	^{137}Cs $^{\#}$ kBq kg^{-1}	SOM‡ %	CEC‡ cmol$_c$ kg^{-1}	Clay$^{≠}$ %
1P	Paddy	2	3	12.06 ± 6.76	10.8 ± 0.4* a	17.5 ± 0.8 a	4.7
1R	Roadside	1	3	260.58	4.9 ± 0.7 c	6.1 ± 0.2 c	10.5
1U	Upland	2	3	10.68 ± 0.02	7.7 ± 0.3 b	16.0 ± 0.4 b	1.3
2C	Canal ditch	1 (2 points)	6	14.64 ± 1.33	6.4 ± 2.0 bc	7.7 ± 0.9 d	4.5
2P	Paddy	3	9	13.82 ± 4.62	7.0 ± 2.81 bc	8.7 ± 2.0 d	3.1
3M	Mountain	1	3	6.35	8.5 ± 1.37 b	11.1 ± 0.7 c	10.8

* Mean ± SD , $^{\#}$ Measured with *n* = (*n*), ‡ Measured with *n* = number of soil samples, $^{≠}$ Measured with *n* = 1, Values in the same column followed by the same alphabet are not significantly different (*P*>0.05) by Tukey's test.

3.3.3 The relationship between plant species and ^{137}Cs uptake

(a) ^{137}Cs uptake by the same plant species in different soils

Table 3.2(a) and (b) show that *So. altissima* and *At. yokoscense* grown in different soils exhibited different ^{137}Cs concentrations and TF values.

For instance, *So. altissima*. in 2P exhibited a high ^{137}Cs concentration and TF value, differing from those for 1P and 1U. This is possibly because soil from 2P has relatively lower exchangeable K and higher NH_4^+ than soil from 1P and 1U. According to Evans and Dekker (1969), applying NH_4^+ to the soil with low exchangeable K resulted in high ^{137}Cs uptake by plants. Similarly, Lasat et al. (1997) reported that applying of NH_4^+ to the ^{137}Cs contaminated soils caused the desorption of ^{137}Cs fixed to clay minerals, resulting in greater ^{137}Cs bio-availability for plants.

Similarly, *At. yokoscense* from 3M exhibited higher ^{137}Cs concentrations and TF values by 1.5 - 2.7 times and 3 - 6 times, respectively, than those from 2C. This is probably because the soil from 3M has relatively higher inorganic nitrogen (NO_3^-, NH_4^+) and SOM content. Mineralization of nitrogen through SOM decomposition causes desorption of ^{137}Cs fixed to clay minerals, resulting in greater ^{137}Cs bio-availability for plants (Sanchez et al., 1999). In addition, among two 2C collection sites for *At. yokoscense*, we found that the ^{137}Cs concentration and TF value were apparently higher in *At. yokoscense* grown in soil with a higher SOM content.

Table 3.2 ^{137}Cs concentration in wild plants and their respective soils, and the TF values.

Wild plants	Locations	Land use types	^{137}Cs in soils [#] kBq kg^{-1}	^{137}Cs in plants [##] kBq kg^{-1}	TF	K^{+}⁑ cmol$_c$ kg^{-1}	NH_4^{+}⁑ mg kg^{-1}	SOM⁑ %
(a)								
Ar. indica	1P	Paddy	16.84	ND[‡]	0	1.3 ±0.1*b[†]	1.8 ±1.1 ab	10.8 ±0.5 a
"	1U	Upland	10.67	0.04±0.04	0.004	2.0 ±0.1 a	2.0 ±0.1 ab	7.6 ±0.4 ab
So. altissima	1P	Paddy	7.29	ND	0	1.3 ±0.1 b	1.8 ±1.1 ab	10.8 ±0.5 a
"	1U	Upland	10.70	ND	0	2.0 ±0.1 a	2.0 ±0.1 ab	7.6 ±0.4 ab
(b)								
At. yokoscense	2C	Canal	13.70	2.51 ±0.20 d	0.18	0.7 ±0.2 cd	0.7 ±0.1 bc	7.3 ±3.1 ab
"	2C	Canal	15.58	1.37 ±0.31 d	0.09	0.9 ±0.1 c	0.5 ±0.1 bc	5.4 ±0.6 b
"	3M	Mountain	6.35	3.71 ±2.98 c	0.58	0.7 ±0.0 cd	3.4 ±0.1 ab	8.5 ±1.6 a
So. altissima⁑	2P	Paddy	12.92	2.76 ±0.32 d	0.21	0.8 cd	5.9 a	4.9 b
"	2P	Paddy	7.29	2.16 ±0.18 d	0.30	1.0 c	3.8 a	4.4 b
"	2P	Paddy	16.18	3.97 ±0.31 c	0.25	0.7 cd	1.7 b	4.4 b
(c)								
At. deltoidofrons	2P	Paddy	12.93	2.90 ±0.74 c	0.22	1.2 ±0.3 bc	1.1 ±0.4 bc	10.6 ±0.7 a
D. tokyoensis	2P	Paddy	19.80	5.63 ±0.26 b	0.28	0.7 ±0.1 cd	0.6 ±0.2bc	5.8 ±0.4 b
(d)								
Se. sarmentosum	1R	Roadside	260.58	21.90 ±1.76 a	0.08	0.3 ±0.0 d	0.8 ±0.2 c	4.9 ±0.8 b

[#] Measured with $n = 1$, [##] Measured with $n = 3$, * Mean ±SD, [‡] Not detected)less than 3 Bq kg^{-1}), ⁑ Measured with $n = 3$, excepted for *So. altissima* in 2P ($n = 1$), [†] Values in the same column followed by the same alphabet are not significantly different (P>0.05) by Tukey's test.

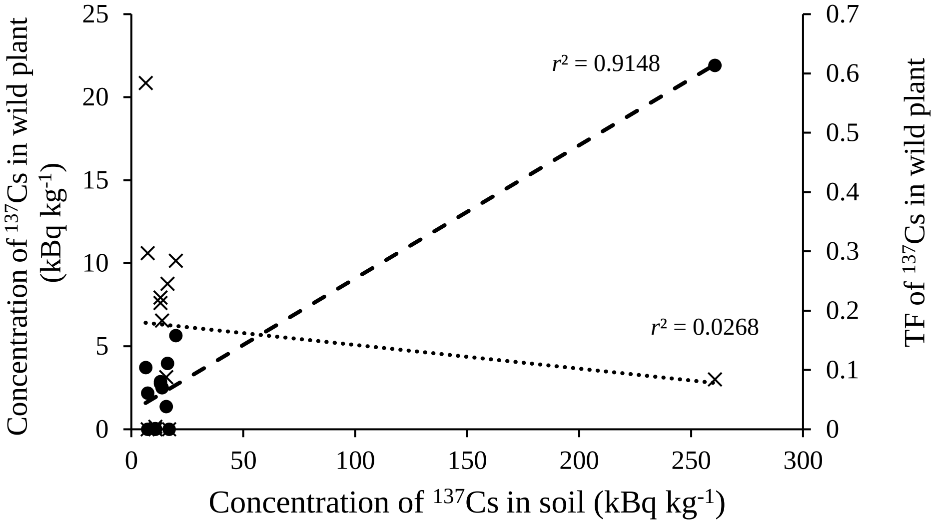

Figure 3.3 Relationships between ^{137}Cs concentration in wild plants and their respective soil)circles(and between TF values of wild plants and ^{137}Cs concentration in the soil)cross(.

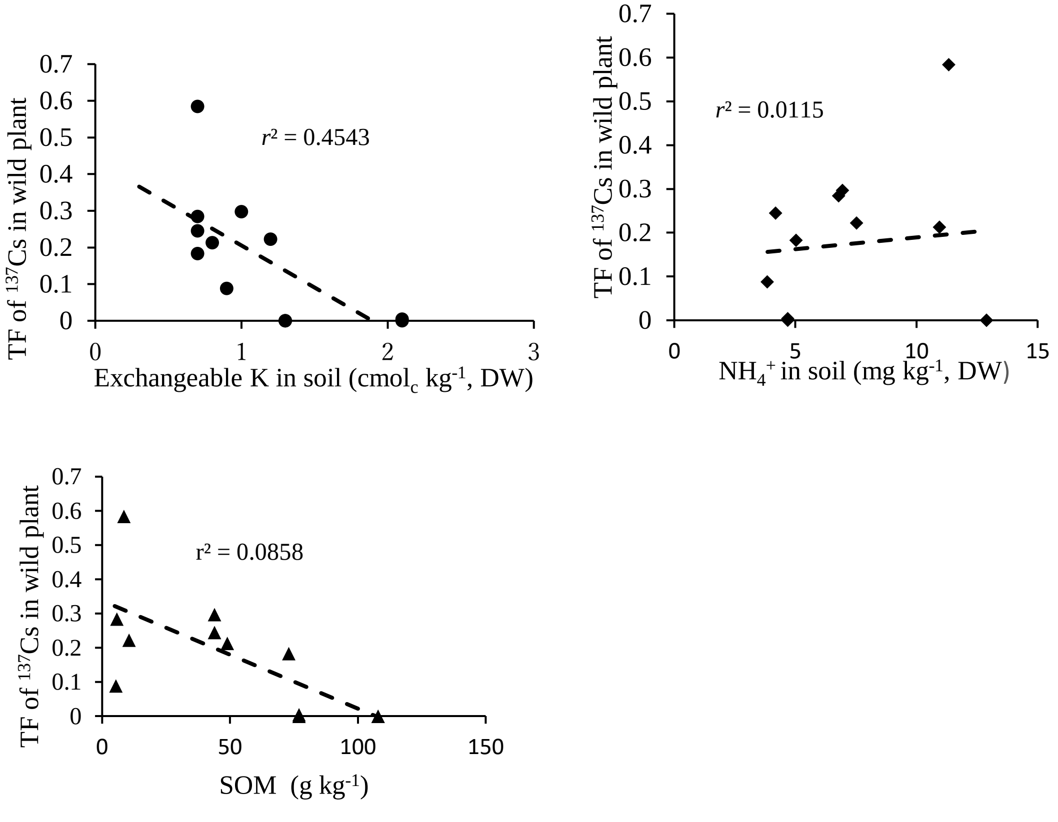

Figure 3.4 Relationships between TF of wild plants and exchangeable-K, NH_4^+, and SOM in the soil.

(b) ^{137}Cs uptake by different plant species in the same soil

At location 2 (2P and 2C), we collected four species of wild plants, one species of weed (*So. altissima*) and three species of ferns (*At. yokoscense*, *At. deltoidofrons*, and *D. tokyoensis*). We found that these plants exhibited similar ^{137}Cs concentrations and TF values (Table 3.2(b) and (c)). Our results, however, differed from Lasat et al. (1997) and Yamashita et al. (2014), who reported that potentially ^{137}Cs uptake of plants depends on their species. Lasat et al. (1997) qualified their results across monocot and dicot species, found that ^{137}Cs accumulation in dicot species was two to four times greater than that in monocots (grasses). In the present study, the investigated wild plants are weeds and ferns, this similarity in their respective physiologies might explain the lower variation in ^{137}Cs concentrations and TF values among the species.

3.3.4 ^{137}Cs concentration and TF of wild plant in an extremely contaminated soil

We collected *Se. sarmentosum* in 1R, where the soil has an extremely high ^{137}Cs concentration (260 kBq kg^{-1}). Our results in Table 3.2(d) shows that the ^{137}Cs concentration in this plant was extremely high (21 kBq kg^{-1}), while the TF value was relatively low (0.085). This result differed from Yamashita et al. (2014), who reported that *Se. sarmentosum* displayed a high potential uptake of ^{137}Cs (TF = 0.4).

The present result suggested that the TF value may not be enough for interpreting the potential for ^{137}Cs uptake by the plants in an extremely contaminated soil.

3.4 Conclusions

The ^{137}Cs concentration in the roadside soil was significantly higher than in the agricultural and mountain soil samples. These differences were affected by a combination of factors, which can be summarized as: (1) land use type and/or the topography affected ^{137}Cs transportation through the leaching process; and (2) soil properties involve clay content and soil organic matter (SOM) affected ^{137}Cs adsorption or desorption.

The ^{137}Cs concentrations and TF values for the same species of plant grown in different soils were significantly different; in contrast, they were similar in different plant species grown in the same soil. These results indicate that a plant's ^{137}Cs uptake potential is affected by soil factors, regardless of the plant species. The soil factors involved were: (1) increasing and decreasing ^{137}Cs bio-availability, which is affected by the soil's clay and SOM content; and (2) the presence of competitive or cooperative ions, where exchangeable K effectively reduces ^{137}Cs uptake by the plant and NH_4^+ increases it. In the present study, however, the soil-to-plant transfer factor value may not enough for interpreting the potential for transferring ^{137}Cs in the extremely contaminated soil.

CHAPTER 4. RADIOCESIUM CONCENTRATION IN STEMS, LEAVES, AND PANICLES OF RICE GROWN IN A SANDY SOIL REPLACEMENT PADDY FIELD TREATED WITH DIFFERENT RATES OF CATTLE MANURE COMPOST IN KAWAMATA, FUKUSHIMA

4.1 Introduction

On 11 March 2011, a huge tsunami triggered by great earthquake hit wide areas of northeast Japan. Due to this event, the FD1NPP was damaged, causing large amounts of radionuclides including ^{137}Cs (half-life: 30 years, emission of β and γ rays during its decay) to be released to the atmosphere subsequently deposited on the ground (MEXT, 2011). The ^{137}Cs can enter the human body through contaminated foods (Avci, 2010).

Food crops produced in ^{137}Cs contaminated land are the main pathway for ^{137}Cs ingestion in human (Zhu and Smolders, 2010). After the FD1NPP accident, the Ministry of Agriculture, Forestry, and Fisheries (MAFF) proposed three methods for recovering the contaminated agricultural lands (MAFF, 2012) : (1) removal of top soil, (2) mixing the soil with water and separating fine particles, and (3) reverse tillage. Removal of top soil seems to be an effective method, as the major portion of ^{137}Cs is fixed in the top soil layer (Nakanishi et al., 2013; Shiozawa et al., 2011; Tanaka et al., 2012). In our study area, Kawamata-town, Yamanichi et al. (2015) reported that ^{137}Cs was fixed in the top 1 cm of the soil.

Based on the above articles, removing the top 0-15 cm layer is estimated to remove more than 80% of the ^{137}Cs from the land (MAFF, 2013). However, it is impossible to remove 100% of ^{137}Cs from the soil, since some small amount may have been absorbed in the deeper layers. According to Ohse et al. (2015). after removal of the 0-10 cm soil layer, approximately 25% of the original cesium remained in the deeper layers. Therefore, reduction of ^{137}Cs uptake by crops through soil management is necessary.

For agricultural lands, removing surface soil is an effective method to reduce ^{137}Cs content in the soil, but this may result in a loss of soil fertility. Improving the soil after the removal is thus necessary. Application of organic matter such as rice straw and cattle manure compost are possible options because of their low cost and high potential for soil improvement. According to Nishiwaki et al. (2017) using rice straw and cattle manure compost to recover soil productivity after removal of surface soil, successfully increased rice production. However, the authors found that application of manure compost caused radiocesium increases in the soil, because the manure compost was also contaminated by radiocesium (Nishiwaki et al., 2017). Cattle manure compost is low cost fertilizer with high plant nutrients, particularly potassium (K) (Eghball et al., 2002; Qian et al., 2005). It is well known that potassium effectively reduces ^{137}Cs uptake by plants (Smolders et al., 1997). To obtain more information concerning the effects of cattle manure compost application on ^{137}Cs in soil and rice, we conducted a field experiment. The experimental paddy field had been decontaminated by removing the soil surface followed by replacement with sandy soil. Cattle manure compost was subsequently applied before rice planting. The objectives of this study were to determine the effects of various rates of cattle manure compost application on rice yields and on ^{137}Cs distribution in stems, leaves, and panicles of rice plants.

4.2 Materials and method

4.2.1 Experimental field and treatments

The experimental field located in Kawamata-town, approximately 50 km northwest of the FD1NPP. The field is bordered by a water way on the north side and a water drainage canal on the south side. The field has an area of 0.48 hectare (ha) and is divided into four plots, each measuring 15 x 80 m. Before the experiments, surface soil had been removed and then replaced with sandy soil.

In each plot, cattle manure compost was applied at 0 (control), 10, 20 and 40 t ha^{-1}, and tillage was done before planting rice (*Oryza sativa* L., cv. Akitakomachi). Twenty-three days old rice seedlings were transplanted with a spacing of 18 x 25 cm on 27 May and harvested in October 2015.

4.2.2 The chemical fertilizer and irrigation

The chemical fertilizers, KCl (K = 40 kg ha^{-1}) and N-P-K (N = 65, P = 87, K= 65 (kg ha^{-1})) were added for all plots. The field was irrigated with an irrigation system. After the transplant of rice, the field had been water logged until harvest.

4.2.3 Cattle manure compost and ^{137}Cs

The cattle manure compost was made from the milk-cow manure mixed with rice husk and fermented over 6 months. The ^{137}Cs concentration of compost was less than 400 Bq kg^{-1}. Chemical components of compost are shown in Table 4.1.

Table 4.1 Chemical components of cattle manure compost.

Analyzed factors	Dried weight bases
Total-N (%)	1.2
Total-C (%)	39.7
P_2O_5 (%)	0.86
CaO (%)	1.3
MgO (%)	0.6
K_2O (%)	3.13
Ash (%)	24.6

Data was provided by Fukushima Prefecture.

4.2.4 Soil and rice sampling

We collected soil and rice samples at harvest on 5 October 2015. In each plot, we collected soil samples at three points with two layers (0-5 and 5-10 cm) using soil core. Rice samples were collected from three points in each plot, and at each sampling point we cut the aboveground parts of rice plants in a 1 m^2 sector. Rice samples were separated into three parts: stem, leave, and panicle. All samples were oven dried at 75°C for three days. The dried rice was ground into powder before the chemical analyses and ^{137}Cs measurement.

4.2.5 Rice yield component measurement

At the harvest, 12 hills (in average) at each sampling point were collected. Grain yield was measured by brown rice weight. The ripened grains were selected by sieving through 1.8 mm mesh. Thousand-grams weights were measured for brown rice at 14.5% moisture content.

4.2.6 Soil analysis

Fresh soil samples were used for inorganic nitrogen analyses. The remaining soils were air dried at room temperature for one week before being sieved through a 2 mm mesh. The dried soils were used for physical and chemical analyses and ^{137}Cs measurement.

Inorganic nitrogen (NO_3^- and NH_4^+) in fresh soil was extracted with 2 *M* KCl at a ratio of 1:10 (soil: solution) and their concentrations were measured with a continuous flow analyzer (BL-TEC Autoanalyzer, QuAAtro-HR). Exchangeable K, Mg, Ca, and Na in dried soil were extracted with 1 *M* CH_3COONH_4 at a ratio of 1:20 (soil: solution) and their concentrations were measured with an atomic absorption spectrophotometer (Z-5300 Polarized Zeeman Atomic Absorption Spectrophotometer). Cation exchange capacity (CEC) measurement was based on the semi-micro Schollenberger method (Kamewada, 1997). Total carbon (TC) and nitrogen (TN) were measured with a CN coder (MT-700 Mark II, Yanaco, Kyoto, Japan). Soil pH and EC were measured at a ratio of 1:5 (soil: water) with pH/ion meter (Horiba F-23, Japan) and conductivity meter (Horiba DS-14, Japan), respectively. Soil texture classification was determined by the pipet method (Nakai, 1997).

4.2.7 Rice analysis

Rice samples were digested by the Kjeldahl method (Horneck et al., 1998). After digestion, K concentration was measured with an atomic absorption spectrophotometer (Shimadzu AA-6800F, Japan).

4.2.8 Radioactivity measurement

Concentrations of ^{137}Cs in soil and rice were measured with a Ge semiconductor (GMX 15200P, Seiko EG&G, Tokyo Japan). Soil samples for the radioactivity measurement were prepared by adding 70 g of dried soil into plastic containers (U8 containers 47 mm in diameter and 60 mm in height); the samples measured 35 mm deep and 1.1 g cm^{-3} in density. Rice samples (approximately 14 to 42 g DW) of each part were added to the containers. Each U8 container was sealed in a polyethylene bag before being placed into a Ge semiconductor. The times of measurement were 30 minutes for soil and 24 hours for rice samples.

4.2.9 Data calculations

The data were used to calculate as the following quantities:

(1) Harvest index (HI) (Bueno et al., 2009; Yang et al., 2010):

$$\mathrm{HI} = \frac{\text{Grain weight (DW)}}{\text{Total rice weight (DW)}}$$

(2) The ^{137}Cs transfer factor (TF) from soil to rice part (Smolders et al., 1997):

$$\mathrm{TF} = \frac{^{137}\mathrm{Cs\ in\ rice\ part\ (Bq\ kg^{-1})}}{^{137}\mathrm{Cs\ in\ soil\ (Bq\ kg^{-1})}\ *}$$

where, * an average of 0-5 and 5-10 cm soil layers.

(3) The TF to aboveground parts:

$$\mathrm{TF\ to\ aboveground} = \frac{\sum_{i=1}^{n} \quad m_i \times c_i}{\sum_{i=1}^{n} \quad m_i} \Big/ \ ^{137}\mathrm{Cs\ in\ soil}$$

where n = the number of rice parts: stems, leaves, and panicles

m = mass weight of each part (g m^{-2}, DW)

c = concentration of ^{137}Cs in each part (Bq kg^{-1})

(4) The ^{137}Cs/K ratio in each rice part:

$$^{137}\mathrm{Cs/K\ ratio} = \frac{^{137}\mathrm{Cs\ (Bq\ kg^{-1})} \times 2.35 \times 10^{-13}\ (\mathrm{cmol_c\ Bq^{-1}})\ *}{\mathrm{K\ (cmol_c\ kg^{-1})}}$$

*(Kondo et al., 2015; Staunton et al., 2002)

4.2.10 Statistical analysis

The effect of compost application on soil nutrients was tested by Tukey multiple comparisons test using the R Statistical Software package (R i386 3.2.0). One-way analysis of variance of rice yields and rice components was done using Microsoft Excel.

4.3 Results and discussion

4.3.1 Effect of cattle manure compost application on soil properties

Soil properties in the experimental plots at harvest are shown in Table 4.2. Soil pH, EC, and bulk density did not change due to compost application. In contrast, clay content was significantly increased by the compost application ($P<0.05$). Total carbon (TC) and total nitrogen (TN) were significantly increased by applying 20 t ha^{-1} of compost ($P<0.05$).

Ammonium (NH_4^+) in soils did not differ among plots and accounted for 0.02 to 0.0.06 % of TN, while nitrate (NO_3^-) was not detected in the majority soils.

Table 4.2 Effect of compost application rates on soil properties at harvest.

Treatment t ha^{-1}	Bulk density Mg m^{-3}	Clay %	pH	EC µS cm^{-1}	NH_4^+ mg kg^{-1}	NO_3^- mg kg^{-1}	T-C g kg^{-1}	T-N g kg^{-1}	SOM %
				top soil (0-5 cm)					
0	1.3 a	5 b	5.3 a	29.5 a	0.56 a	ND	10.8 b	0.98 b	1.86 b
10	1.3 a	11 a	5.4 a	31.2 a	0.63 a	0.26	17.0 ab	1.38 ab	2.92 ab
20	1.2 a	10 a	5.4 a	26.6 a	0.68 a	0.57	22.7 a	1.74 a	3.90 a
40	1.3 a	9 a	5.7 a	29.4 a	0.39 a	ND	19.7 a	1.49 ab	3.39 a
				sub soil (5-10 cm)					
0	1.4 a	4 b	5.6 a	33.9 a	0.41 a	ND	10.6 b	0.93 b	1.81 b
10	1.3 a	10 a	5.5 a	36.6 a	0.42 a	ND	14.1 ab	1.19 ab	2.43 ab
20	1.2 a	9 a	5.6 a	34.8 a	0.36 a	ND	21.7 a	1.61 a	3.72 a
40	1.3 a	9 a	5.7 a	22.4 a	0.25 a	ND	17.5 a	1.32 ab	3.01 a

SOM = Soil organic matter, ND = Not detected, Values in a column followed by the same letter are not significantly different at ($P>0.05$) based on Tukey's test.

Soil cation exchange capacity (CEC) did not significantly differ among plots and layers ($P>0.05$) (Table 4.3). Exchangeable K, Mg, and Ca were significantly increased by applying 20 t ha^{-1} of compost ($P<0.05$). However, Na did not differ between plots ($P>0.05$).

Somewhat unexpectedly, soil nutrients in the 40 t ha^{-1} plot seemed to decrease as compared to those of the 20 t ha^{-1} plot. Among three sampling points in the 40 t ha^{-1} plot, the south side of the plot showed a significantly lower concentration of soil nutrients; for example, TC and TN were lower than those in the center and the north side ($P<0.05$). This may possibly have been caused by soil erosion.

Table 4.3 Effect of compost application rates on CEC, exchangeable K, Mg, Ca, and Na in soil at harvest.

Treatment t ha^{-1}	CEC	K	Mg	Ca	Na
	cmol$_c$ kg^{-1}				
			top soil (0-5 cm)		
0	16.24 a	0.15 b	0.82 b	6.35 b	0.28 a
10	15.78 a	0.39 ab	0.95 a	7.48 ab	0.32 a
20	19.02 a	0.47 a	0.94 ab	7.86 a	0.28 a
40	14.60 a	0.40 ab	0.88 ab	6.81 ab	0.28 a
			sub soil (5-10 cm)		
0	14.24 a	0.18 b	0.80 b	6.47 b	0.34 a
10	23.33 a	0.42 ab	0.97 ab	7.46 ab	0.32 a
20	15.39 a	0.55 a	1.09 a	8.31 a	0.31 a
40	21.49 a	0.43 ab	0.88 ab	6.65 ab	0.27 a

Values in a column followed by the different letters are significantly different at (*P*<0.05) based on Tukey's test.

4.3.2 Effect of compost application on ^{137}Cs concentration in soils

In all plots, ^{137}Cs remained in the soil after removing the soil surface (Figure 4.1). The concentration of ^{137}Cs varied widely within each plot, with a coefficient of variation of 14 to 50%, and did not differ between the top and sub layers.

The concentration of ^{137}Cs in soils tended to increase with increasing composting rates and was positively correlated with soil exchangeable K ($r^2 = 0.79$) (Figure 4.2). The ^{137}Cs concentration in the 20 t ha^{-1} plot was higher than in other plots, and a similar trend was observed with soil nutrients. This was probably due to ^{137}Cs remaining in the soil after removal of the soil surface and combination with the compost being contaminated by ^{137}Cs (< 400 Bq kg^{-1}).

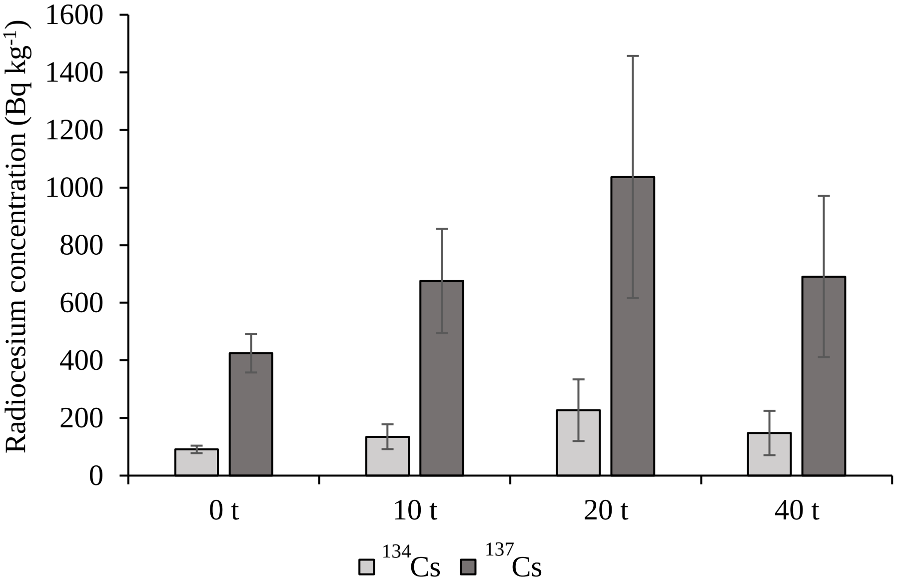

Figure 4.1 Concentrations of ^{134}Cs and ^{137}Cs in the soil (an average of the 0-5 and 5-10 cm layers) that received different rates of compost (t ha^{-1}). (Error bars are SD; n = 3).

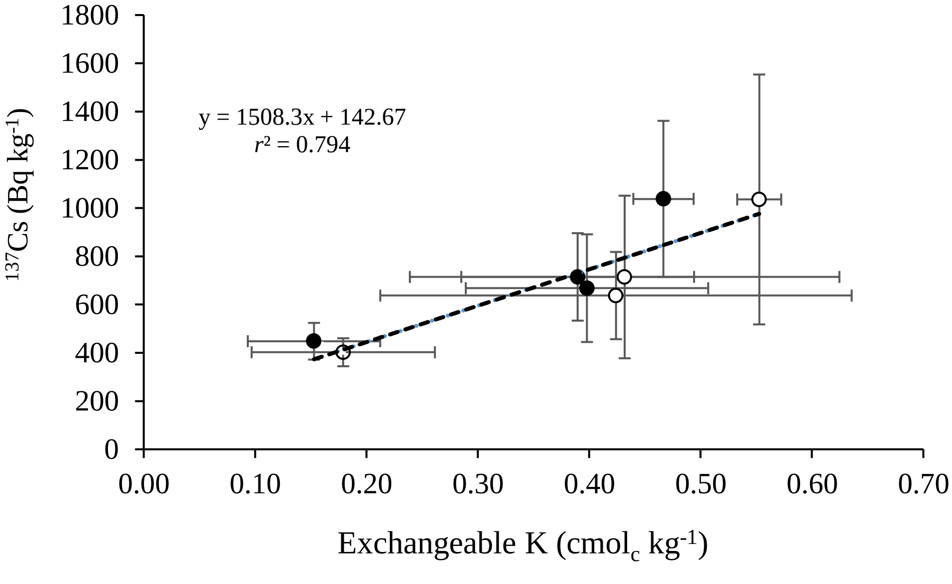

Figure 4.2 The Relationship between ^{137}Cs and exchangeable K in top soil, 0-5 cm (solid circles) and sub soil, 5-10 cm (open circles) (Error bars are SD; n = 3).

4.3.3 Effect of compost application rates on rice yields

Rice yield related traits are shown in Table 4.4. Total mass of rice increased by 2, 22, and 34 %, and unhulled grains weight increased by 1.6, 19, and 31 % by applied 10, 20 and 40 t ha^{-1} of compost, respectively. Harvest index (HI) and grain to straw ratios were not significantly different between plots but tended to decrease in the 40 t ha^{-1} plot (P>0.05).

Table 4.4 Rice yield related traits.

Treatment t ha^{-1}	Total dry mass g m^{-2}	Unhulled grain g m^{-2}	Straws g m^{-2}	HI	Grain/Straw ratio
0	1116 ± 156 a	568 ± 86 a	547 ± 71 a	0.509 ± 0.007 a	1.04 ± 0.029 a
10	1139 ± 89 a	577 ± 34 a	561 ± 60 a	0.508 ± 0.021 a	1.04 ± 0.084 a
20	1359 ± 343 a	681 ± 139 a	678 ± 205 a	0.505 ± 0.026 a	1.03 ± 0.107 a
40	1499 ± 355 a	745 ± 185 a	753 ± 185 a	0.497 ± 0.038 a	0.99 ± 0.154 a

Mean ± SD; (n = 3).
Values in a column followed by the same letter are not significantly different ($P > 0.05$) based on one-way analysis of variance.

Table 4.5 Grain yield and components.

Treatments t ha^{-1}	No. of panicles m^{-2}	No. of spiklets panicle^{-1}	Ripened grains %	1000 gains weight g	Grain yield g m^{-2}
0	371 ± 57 a	61 ± 4 a	86.7 ± 2.3 a	23.1 ± 0.3 bc	453 ± 64 a
10	366 ± 24 a	64 ± 3 a	85.0 ± 2.7 a	22.9 ± 0.2 c	458 ± 39 a
20	417 ± 42 a	64 ± 8 a	85.7 ± 1.4 a	23.4 ± 0.2 ab	593 ± 36 a
40	438 ± 13 a	67 ± 4 a	86.9 ± 1.0 a	23.7 ± 0.2 a	601 ± 149 a

Mean ± SD; (n = 3).
Values in a column followed by the same letter are not significantly different based on one-way analysis of variance.

Rice grain yields and components are listed in Table 4.5 Panicle number, spikelet per panicle, and percentage of ripened grains were not significantly different between plots (P>0.05). However, the 1000-grains weight of rice in the 40 t ha^{-1} was significantly higher than those in other plots (P<0.05).

4.3.4 Effect of compost application rates on ^{137}Cs in rice plant parts

The concentration and transfer factor (TF) of ^{137}Cs in rice parts are listed in Table 4.6. The concentration of ^{137}Cs in all rice parts was much lower than the Japanese standard (100 Bq kg^{-1}) (Ministry of Health, Labour and Welfare, 2013).

Table 4.6 Concentration of ^{137}Cs in rice plant parts and transfer factor (TF).

Treatments t ha^{-1}	Stems Bq kg^{-1}	Leaves Bq kg^{-1}	Panicles Bq kg^{-1}	TF of stems	TF of leaves	TF of aboveground parts
0	21	14	ND	0.050	0.034	0.022
10	35	6	ND	0.051	0.009	0.020
20	24	5	ND	0.024	0.005	0.009
40	18	9	ND	0.026	0.013	0.011

The ^{137}Cs concentration in rice parts was measured with n = 1, ND = Not detected.

Transfer factor (TF) of ^{137}Cs from soil to aboveground parts was decreased by cattle manure compost application. The TF value was lowest in the plot with 20 t ha^{-1} of compost (TF = 0.009), this was 2.4 times lower than that of the control plot. We found that TF values in aboveground parts of rice plants were negatively correlated with exchangeable K in the soil (r^2 = 0.64) (Figure 4.3).

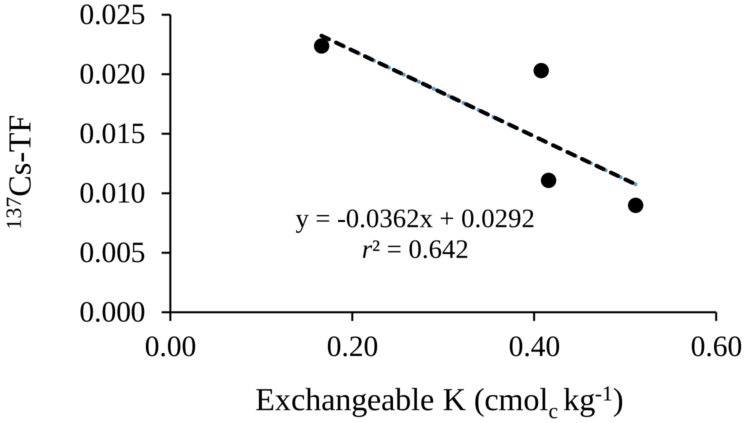

Figure 4.3 The relationship between ^{137}Cs transfer factor to aboveground parts of rice plants and exchangeable K in the soils (an average of 0-5 and 5-10 cm soil layers), (n =1).

The ^{137}Cs transfer factor (TF) value of stems was higher than that of leaves, indicating that ^{137}Cs accumulation in stems was greater than that in leaves. In the present study, ^{137}Cs was not detected in rice panicles. This result differed from those of other researchers. Endo et al. (2013), who studied about a heavily contaminated paddy field in Minami-Soma town in 2011, found that TF of ^{137}Cs of rice panicles ranged from 0.019 to 0.026. Uchida et al (2006). reported that TF of ^{137}Cs of brown rice in Japan before the FD1NPP accident was in the range of 0.001- 0.03.

Application of potassium (K) effectively reduces ^{137}Cs uptake by rice roots and accumulation in rice grains (Saito et al., 2012; Kato et al., 2015; Saito et al., 2015). Saito et al. (2012) reported that radiocesium concentration of brown rice decreased by the increasing of exchangeable K in soil. Kato et al.(2015) recommended that exchangeable K in soil should be higher than 0.50 cmol kg^{-1}. Saito et al. (2015) found that application of potassium fertilizer on the early growth stage effectively reduced ^{137}Cs concentration in rice grain. According to Nobori et al. (2014), rice grown in K-deficient culture solution showed 6.7 times more ^{137}Cs translocation from leaves to the panicle than those in the K-sufficient solution. Appropriate soil K content during the growth stage of rice reduced ^{137}Cs transportation and accumulation in rice panicle (Nobori et al., 2014).

Our results could be explained that exchangeable K derived from the cattle manure compost effectively reduced ^{137}Cs uptake and accumulation in leave and panicle of the rice plants.

4.3.5 Distributions of ^{137}Cs, K, and N in rice plant parts

Concentrations of ^{137}Cs, K, and N in each rice plant part are shown in Figure 4.4 Distributions of ^{137}Cs and K in the rice plant parts were similar and differed from those for N. Both ^{137}Cs and K in stems was higher than in leaves and panicles, suggesting that ^{137}Cs transport in the rice plants was associated with K transporter.

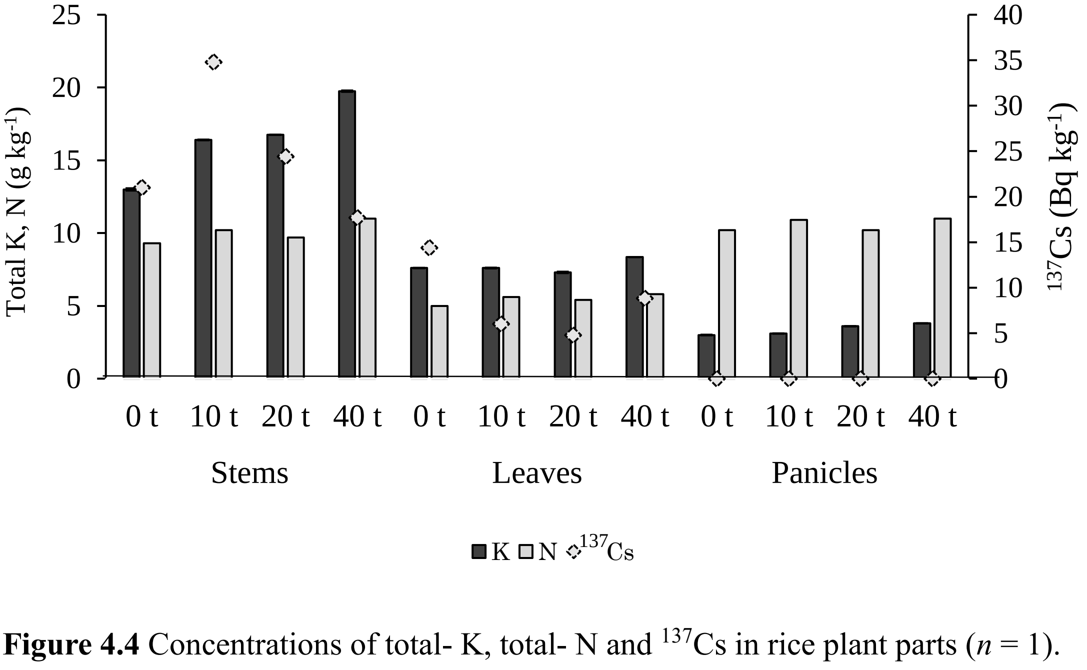

Figure 4.4 Concentrations of total- K, total- N and ^{137}Cs in rice plant parts (n = 1).

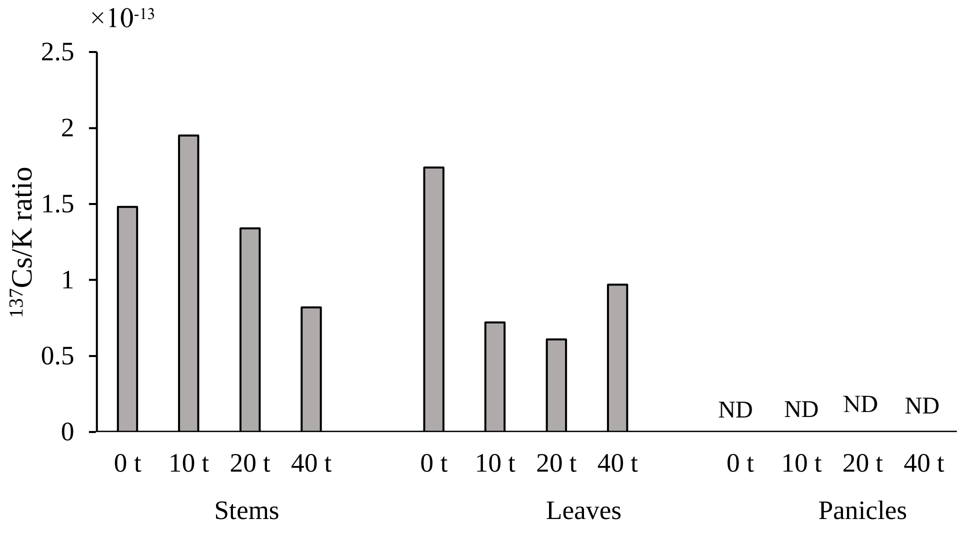

Figure 4.5 The ^{137}Cs/K ratios of stems, leaves, and panicles of rice ($n = 1$), ND = Not detected.

The mechanism regulating ^{137}Cs distribution in plant organs is closely related to the K transport (White et al., 2004; Maathuis et al., 1996; Cline et al., 1960). However, the translocation and accumulation of ^{137}Cs and K in plant parts were not exactly the same in our experiment. This can be explained by their different physical characteristic such as the ionic radiuses (1.69 nm for Cs and 1.33 nm for K) (Johanson et al., 2004). Different of ^{137}Cs and K accumulation in rice parts has been previusly reported (Kondo et al., 2015; Nobori et al., 2014). Rice parts with higher ^{137}Cs/K indicate greater ^{137}Cs accumulation or greater K transportation. In the present study, the ^{137}Cs/K ratio was higher in stems than in leaves in 10 and 20 t ha^{-1} plots (Figure 4.5). Our results agreed with those of Kondo et al. (2015), who reported that the ^{133}Cs/K ratio in roots was higher than in aboveground parts of rice. Similarly, Tsukada et al. (2002) reported that the stable-Cs/K ratio decreased in the younger leaf blade positions of rice plant, indicated that Cs was lower translocated to younger leaves than K.

In contrast, the ^{137}Cs/K ratios of rice plants in the 0 and 40 t ha^{-1} plots were higher in leaves than that in stems; these plots were low in Mg and Ca compared to the 10 and 20 t ha^{-1} plots. This result suggests that soil with low Mg and Ca might affect K uptake, resulting in a permision of ^{137}Cs translocation between rice organs. The K transport mechanism in plant cells is regulated by cations including K, Ca, Mg, Na in soil solution(Smolders et al., 1997; Epstein, 1961; Viets, 1994; Heredia et al., 2002; Overstreet et al., 1952).

In the present study, the ^{137}Cs/K ratio of leaves was negatively correlated with exchangeable K, Mg, and Ca in the soil (Figure 4.6), suggesting that K, Mg and Ca affected ^{137}Cs translocation inside the rice plants. However, those correlations were not apparented in stems (Figure 4.7).

The differences in ^{137}Cs/K ratio in rice plant parts indicated that their was discrimination in the rate of ^{137}Cs versus K transportation3) and that was affected by both external nutrient conditions and rice plant physiology (Van et al., 1981; White, 1999; White et al., 2003; Hamton et al., 2005; Buysse et al., 1995).

In addition, regardless of soil nutrient condition, it is important to study rice plant physiological mechanism controlling ^{137}Cs distribution and accumulation in different rice parts.

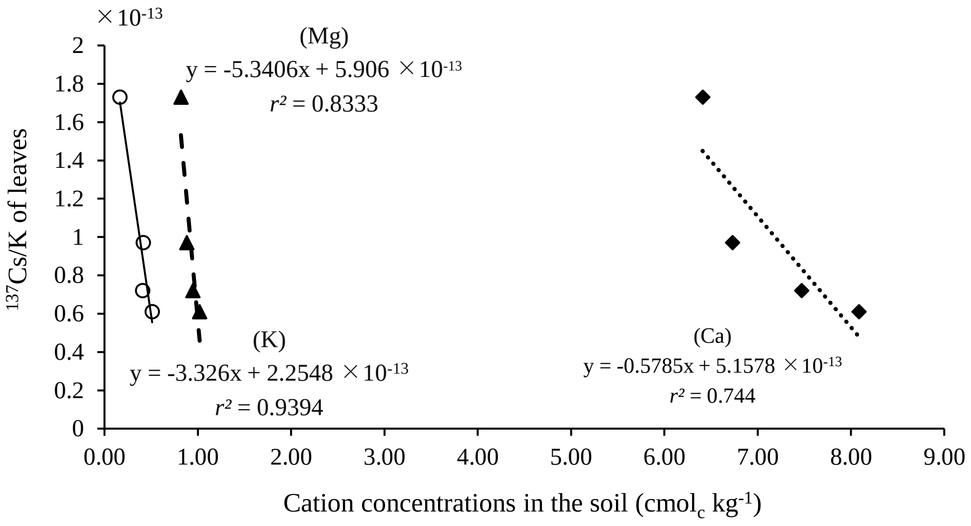

Figure 4.6 The relationship between ^{137}Cs/K ratio of rice leaves and exchangeable (circles), Mg (triangles) and Ca (diamonds) in soil (n = 1).

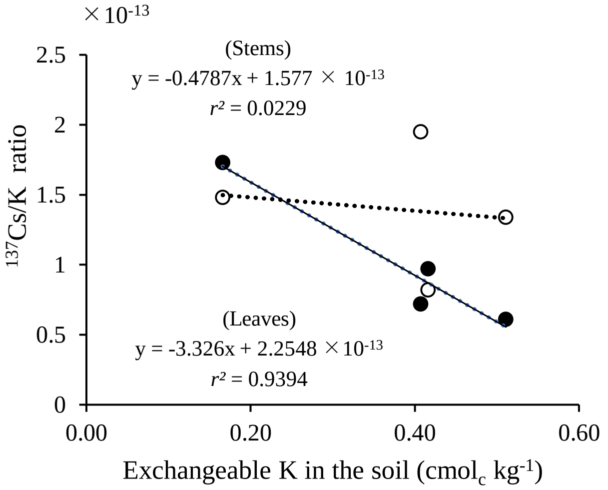

Figure 4.7 The relationship between exchangeable K in the soil (an average of 0-5 and 5-10 cm soil layers) and ^{137}Cs/K ratio of stems (open circles), and leaves (solid circles) (n = 1).

4.4 Conclusion

Application of cattle manure compost induced the increase of rice yields. Concentrations of ^{137}Cs in rice plant parts were decreased by application of cattle manure compost. The lowest ^{137}Cs was found in rice grown in the 20 t ha^{-1} plot (2.4 times lower than control soil) probably because of the increase of soil K (exchangeable K). The ^{137}Cs in rice plant parts was decreased: stems > leaves and was not detected in the panicles of rice plants in all plots. Distribution of ^{137}Cs in rice plant parts was similar to that of K. Different of ^{137}Cs /K ratio in rice plant parts indicated the different accumulation rate of ^{137}Cs and K. The ^{137}Cs/K ratio in stems was higher than that in leaves in the soil with higher Mg and Ca, indicating that Mg and Ca in soil regulated ^{137}Cs translocation between rice plant parts.

For future research, an important goal will be to understand the mechanism of ^{137}Cs transportation and accumulation between rice parts. The effects of Mg and Ca on ^{137}Cs distribution in rice plants should be more researched.

CHAPTER 5. ADSORPTION AND DESORPTION OF STABLE CESIUM IN MINERAL SOILS FROM IITATE-VILLAGE

5.1 Introduction

After the FD1NPP accident, the government designed some areas to be an evacuation zone, Iitate-Village was included. The population was moved out and the agricultural land had not been cultivated since 2011. To recover this situation, decontamination of agricultural land would be the first step. Information on the distribution of ^{137}Cs in the agricultural land and factors affecting ^{137}Cs adsorption in soil will be useful for further research and decontamination works. In this study, we investigated in agricultural land in Iitate-village. The soil samples were collected from the paddy field and upland field in August 2014. Stable cesium (CsCl) was used for extraction experiment in the laboratory. The objective of this study was to determine factors affecting Cs adsorption and desorption for mineral soils of Iitate-village.

5.2 Materials and method

5.2.1 Soil samples

The soil samples were collected using a scope to the depth of 0-5 cm in paddy field and upland field in Iitate-Village in August 2014. The fields had no cultivated since the accident in 2011. The collected flesh soils were kept in polyethylene bags to carried for laboratory. Soils were dried at room temperature for one week, then ground and sieved through 2 mm mesh screen. The samples were kept dried in polyethylene bags until used. For all experiments, the soils from both paddy and upland fields were used and the extraction experiments were done with two replications.

Table 5.1 Initial properties of soils.

Sample	Clay (%)	Exchangeable K cmol kg^{-1}	CEC cmol kg^{-1}	NH_4^+ mg kg^{-1}
Paddy soil	4.68	1.28	17.5	1.78
Upland soil	1.27	2.07	16.0	1.97

Table 5.2 Chemical components of bamboo fresh powder and charcoal

Materials	NO_3^- (mg kg^{-1})	NH_4^+ (mg kg^{-1})
Fresh bamboo	ND	2.46
Bamboo charcoal	ND	514

ND = Not detected

5.2.2 Extraction experiments

The experiment was conducted to examine the effects of:

a) Contact times.

b) Concentrations.

c) Competitive ion.

d) Organic materials amendment.

5.2.3 Chemical solution preparation

The chemical property of ^{137}Cs similarly to that of stable cesium (Khandaker et al., 2018), stable cesium is general use instead of ^{137}Cs. In our laboratory, stable Cs solution was prepared by using cesium chloride (CsCl, FW= 168.36 g). Preparing 1000mg-Cs l^{-1} stock solution was performed by dissolved dried 1.266 g of CsCl in pure water and then adjusted to 1000 ml with pure water. Cesium adsorption experiments were used 5 mg l^{-1} (ppm) and 10 mg l^{-1}, which were prepared by using stock solution diluted with pure water.

5.2.4 Measurement of Cs concentration

For all experiments, adding the soil samples and solution to 45 ml centrifuge tube the shook and centrifuged by 5000 rpm for 5 minutes, and then filtered through 45μ membrane.

The supernatant was used to measure Cs and K concentrations by an atomic absorption spectrophotometer (Z-5300 Polarized Zeeman Atomic Absorption Spectrophotometer). The equilibrium of Cs adsorbed in the soil was calculated as follows.

$$(\triangle Cs \times \text{Cs solution's volume}) / \text{Soil}$$

where:

$\triangle Cs = Cs_0 - Cs_1$

Cs_0 = initial concentration

Cs_1 = Cs concentration of supernatant

5.2.5 The Cs adsorption experiments

a) Effect of contact times

Soils 4 g (DW) and 40 ml 5 mg l^{-1} of Cs solution were added to the 45 ml centrifuge tube, shook for 1, 6, 12, 24, and 48 hrs. After shaking, centrifuged by 5000 rpm for 5 minutes and then filtered through 45μ membrane using the plastic syringe. The supernatant was used for Cs concentration measurement. Both paddy and upland soils were used and done with two replications.

b) Effect of Cs concentrations

Soils, 4 g (DW) and 40 ml of various concentrations; 0, 5,10, and 20 mg l^{-1} of Cs were added to 45 ml centrifuge tube, shook 2 hours. After shaking, centrifuged by 5000 rpm for 5 minutes and then filtered through 45μ membrane using the plastic syringe. The supernatant was used for Cs concentration measurement.

c) Effect of competitive ion

The competitive ion was used potassium (K). Preparation of solution was done by mixing solutions 10 mM of Cs (0.0752 mg l^{-1}) and 10 mM of K (0.256 mg l^{-1}), expressed as Cs (mM) /K(mM) ratio. For each sample 4 g of dried soil and 40 ml of solutions at the ratios of 1:0, 1:0.5, 1:1, and 1:2 were added to the 45 ml centrifuge tube, the extraction procedure was same as expressed above.

d) The effect of soil amendment with various material

Preparing of soil with organic materials by added fresh bamboo powder at rates of 10% and 20% of soil weight (DW) into soil and mixed. This method was the same for bamboo charcoal. For each sample, added 4 g of mixed material and 40 ml of 10 mg l^{-1} of Cs to the centrifuge tube, the extraction procedure was the same as above. The following content, treatments will be expressed as soil paddy or upland10 and 20 is equal to paddy or upland soil + 10% and 20% amendment materials, respectively.

5.3 Results and discussion

5.3.1 Effect of the contact-time

Figure 5.1 showed that Cs can be adsorbed suddenly after contacted with the soil. Cesium adsorbed in soil was countered to over 90 % of the initial concentration for both soils. Different of adsorption rate among two soils probably because different in soil properties such as clay and potassium (exchangeable K) contents. Our soil samples were contained 4.68 and 1.27 % of clay, and 1.28 and 2.07 cmol/kg of exchangeable K, paddy soil and upland soil respectively. Our result suggested that Cs can be fixed immediately after its contact with the soils.

According to Nakashima (2018) radioactive cesium ^{134}Cs and ^{137}Cs remained at the initial points of deposition with not moving. The resulted of our study may explain that Cs be fixed strongly in mineral soil after its deposition. However, Cs can be re-distributed through soil erosion Sutherland (1994).

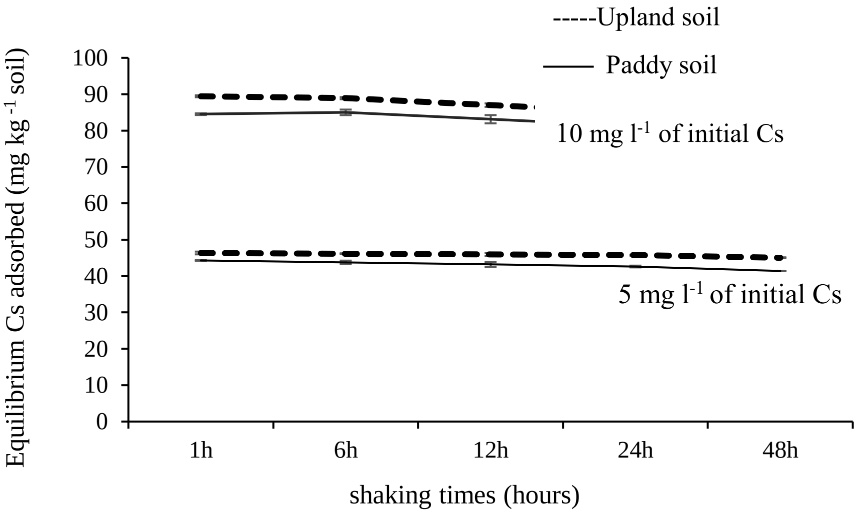

Figure 5.1 The effect of shaking time on Cs adsorption for soils.

5.3.2 Effect of the concentration

Figure 5.2 showed that Cs adsorption increased by the increase of Cs concentration. This result may be explained that capacity for Cs adsorption of the soils should be higher than our tested concentration. The different adsorption lines for two soils was not observed, indicated that soil properties were not affected the capacity of adsorption for Cs.

Our result was agreed with the report of Nakanishi (2018), the adsorption for ^{137}Cs in mineral soil was had not showed the saturation. However, in our experiment, the highest concentration may be much lower than the capacity of adsorption of the soils.

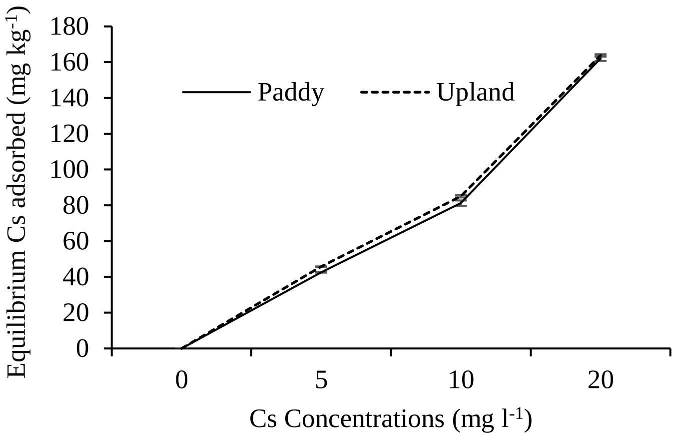

Figure 5.2 The effect of concentration of Cs on Cs adsorption for soils.

5.3.3 Effect of the competitive ion

Figure 5.3 showed that Cs adsorption was increased when the solution containing the ratio of Cs/K in solution from 1/0 to 1/0.5, but these was gradually decreased when rates of K increased. This result suggests that increasing of K from 0 to 0.5 caused Cs adsorption, but when K equaled to Cs the adsorption will not be affected. The different for Cs and K to be fixed in clay mineral may be explained by their difference in ionic radius (1.33 nm for K and 1.69nm for Cs) (Johanson et al.1999). When K presence in solution it competed with Cs to be fixed in clay mineral.

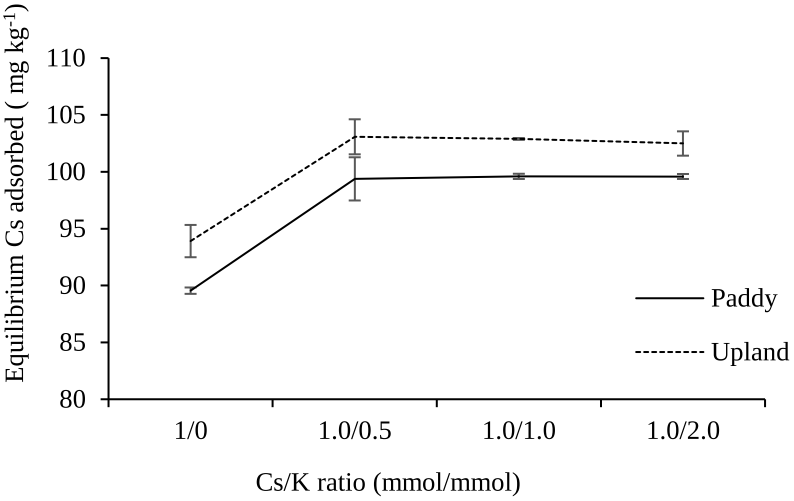

Figure 5.3 The effect of K concentration in solution on Cs adsorption for soil.

5.3.4 Effect of organic materials amendment on Cs adsorption

Figure 5.4 showed that when the soils received fresh bamboo powder and charcoal powder the adsorption of Cs were reduced as compared with control soils. Among two rates of both fresh powder and charcoal, bamboo charcoal showed the lowest Cs adsorption. Our result was similar to the report of Khandaker et al. (2018) who studied using bamboo charcoal as an adsorbent for Cs from aqueous solution. The results suggested that the addition of organic material caused the reduction of Cs adsorption for soils. This explained that the soil with high organic matter resulted in more Cs desorption and enhance Cs available for plant uptake. Dumat and Staunton (1999) reported that clay-humic substance complexes caused less Cs adsorption than the uncoated clay mineral.

Both paddy and upland soil showed that adding fresh bamboo and charcoal resulted in reduce Cs adsorption. Among fresh bamboo and charcoal, charcoal was more effectively reduced Cs adsorption.

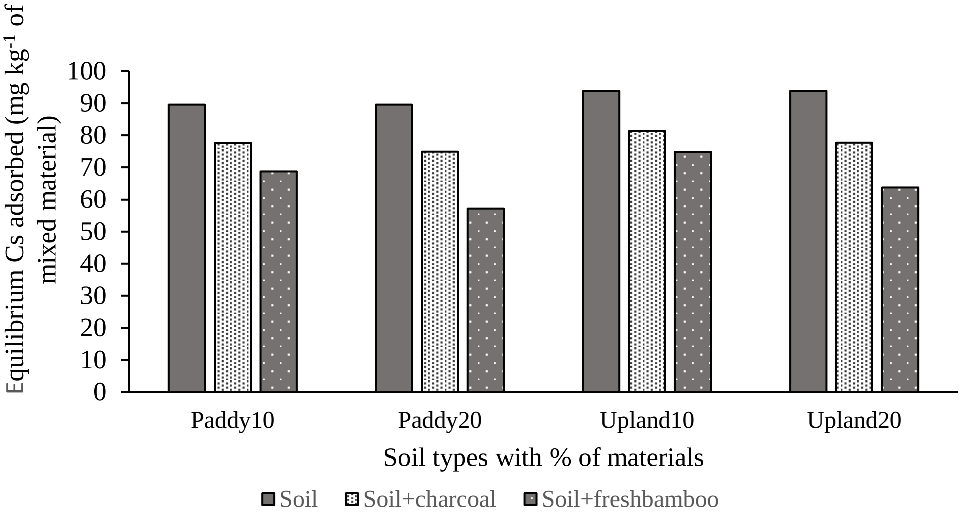

Figure 5.4 Effect of organic materials amendment in soil on Cs adsorption.

5.4 Conclusion

The results showed that Cs adsorption capacity of mineral soils from Iitate-village did not show the saturation stage but increased with increasing of Cs concentration. This can explain that the soil can adsorbed the fallout of ^{137}Cs with unlimited. The Cs or ^{137}Cs can be adsorbed immediately after its contact with soils. The present of potassium (K) in solution had influenced to Cs adsorption up to the ratio of 1/0.5, suggesting that the Cs adsorption was less when the solution absent of K, but the present of K ion caused Cs adsorption. This result explained that K ion in soil solution expanded the adsorption sites for Cs. On the other hand, soil organic matter plays a role as reduces Cs adsorption as its block the adsorption sites in soil particles, resulted in less adsorption.

CHAPTER 6. GENERAL DISCUSSIONS AND CONCLUSION

6.1 Factors affecting ^{137}Cs adsorption in soil

Our results from Chapter 5 revealed that stable Cs can be adsorbed by mineral soils immediately after its deposition, and that the capacity for adsorption by soils from agricultural fields in Iitate-Village did not show the saturation stage and unlimited Cs concentration. The presence of K in solution seems to have stimulated Cs adsorption. The presence of organic matter in solution can interfere with the Cs to be adsorbed onto the soils. These results explained the finding of the investigation of the degree of ^{137}Cs contamination in areas of different land use types, as represented in Chapter 3.

The roadside soil with high clay content showed the highest ^{137}Cs (260 kBq kg^{-1}), and this can be explained by the deposition of eroded particles at the roadside. The soils from agricultural fields showed the ^{137}Cs range of 11-14 kBq kg^{-1}, which should be the initial ^{137}Cs deposition. The mountain soil with relatively high organic matter showed the lowest level (6 kBq kg^{-1}).

In Chapter 3, we showed that in natural conditions, distribution of ^{137}Cs across the landscape was affected by a number of factors involved in both the sloping of the fields and soil property. After ^{137}Cs is deposited on the ground, it can be fixed strongly to soil minerals, but re-distribution occurs through physical process (soil erosion).

6.2 ^{137}Cs uptake by plants

Plants grown in the ^{137}Cs contaminated fields play important roles as indicator, extractant, and distributor of ^{137}Cs in ecosystems. In Chapter 3, revealed that concentrations of ^{137}Cs in wild plants directly reflected that of their respective soil. Different plant species grown in the same soil showed similar ^{137}Cs levels, but the same plant species grown in different soils showed different ^{137}Cs concentration levels, and these finding suggested that capacity for ^{137}Cs uptake by plants depend on the ^{137}Cs concentration in the soil regardless of the plant species. In general, the ^{137}Cs transfer factor (TF) used to explain the relationship of ^{137}Cs concentrations between plants and their corresponding soil, but we found that plants grown in the extremely high ^{137}Cs concentration showed low TF values. This result suggested that the TF value may not adequate to interpret the levels of ^{137}Cs concentration in extremely contaminated soil.

6.3 Reduction of ^{137}Cs in soils and rice

After the FD1NPP accident, the Japanese government recommended effectively methods to reduce the ^{137}Cs concentration in the contaminated fields. Removal of the soil surface seems to be the most effective. Chapter 4 presented the strategy of reduction ^{137}Cs in soil through soil surface removal. However, after soil surface had been removed, some ^{137}Cs still remained. Protection of food crops from ^{137}Cs contamination is important. It is well known that sufficient K in soil can reduce the ^{137}Cs uptake by rice. Decontamination of agricultural fields through removal of the surface soil caused a loss of soil fertility. Application of cattle manure compost is a useful option for soil improvement, and derived cations (K, Ca, and Mg) can reduce ^{137}Cs uptake by rice plants.

In addition, as reviewed in Chapter 2, possibly factors involved in our results can be sumarized as, 1) the physical characteristic of ^{137}Cs, ^{137}Cs is larger ionic radius than K, resulting in its lower transport than K. 2) the shoot vacuoles of rice limits for accumulating ^{137}Cs. And 3) rice grain vacuoles have the TPKb which is small and limits for ^{137}Cs permeability in rice grain. These factors involved in the decrease of the ^{137}Cs/K ratio in the aboveground parts of rice plant and the ^{137}Cs was not detected in rice panicles.

6.4 Conclusion

From our research, we found that ^{137}Cs concentrations in fields of different uses were significantly different, and these differences involved a number of factors. After ^{137}Cs is deposited on the ground, re-distribution occurred through ground water (erosion). The slope of the lands, further soil properties affect the movement of ^{137}Cs across the landscape. As our results showed, roadside soil with a high clay content was highest in ^{137}Cs concentration, a middle level was found in agricultural fields, and soil from the base of the mountain relatively high in organic matter showed the lowest ^{137}Cs.

On the other hand, the concentration of ^{137}Cs in rice was successfully reduced by input of cations such as potassium (K) into the soil. Application of cattle manure compost increased the soil fertility after soil surface removal, and it provided cations such as potassium (K), calcium (Ca), and magnesium (Mg), which effectively reduced ^{137}Cs uptake by rice plants and ^{137}Cs distribution between rice parts.

For future research it is important to study further the mechanism of ^{137}Cs uptake and accumulation in rice plant parts because it would be controlled by both external soil conditions and rice plant physiology.

Additionally, our study results were based on investigations in natural conditions and outdoor fields, so the variations of data were wide. However, we believe that our research will be valuable for future research on the ^{137}Cs distribution in soil and plant systems as well as the decontamination of the ^{137}Cs contaminated lands.

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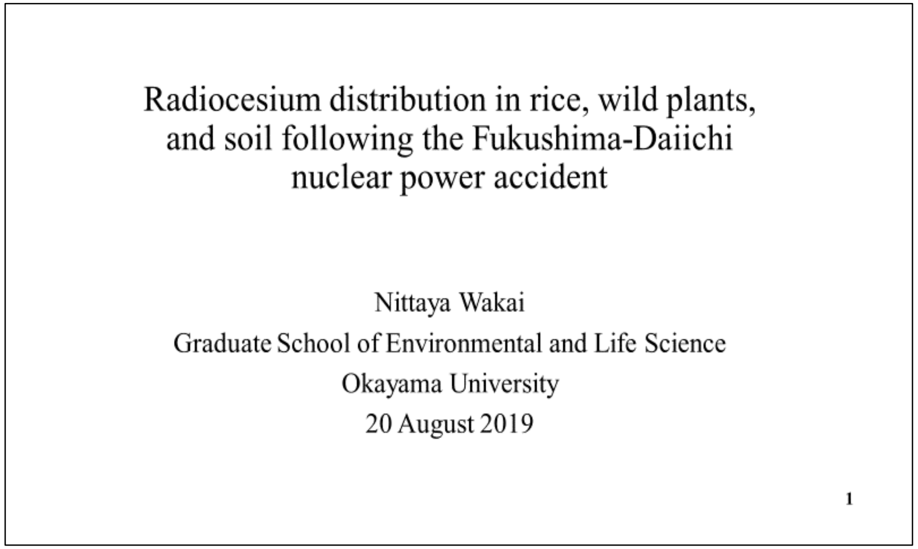
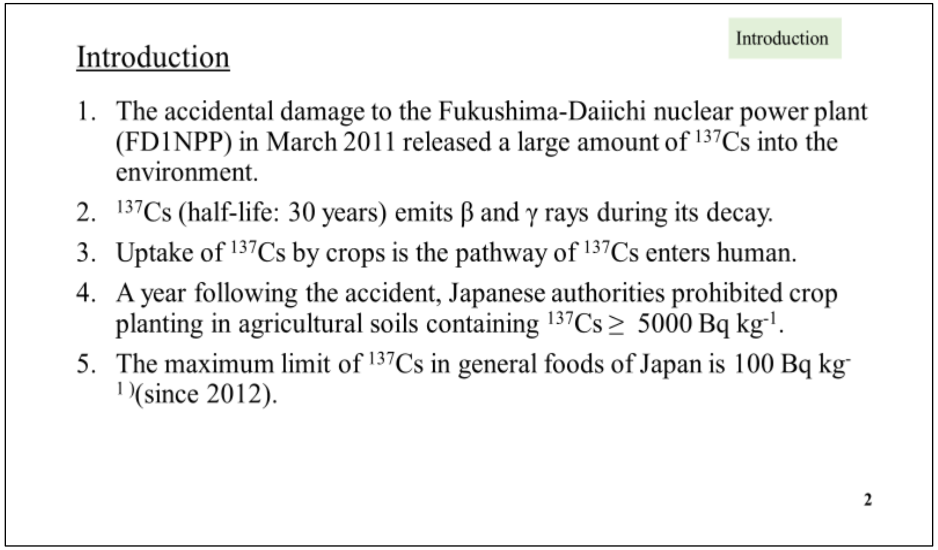
Presentation slides



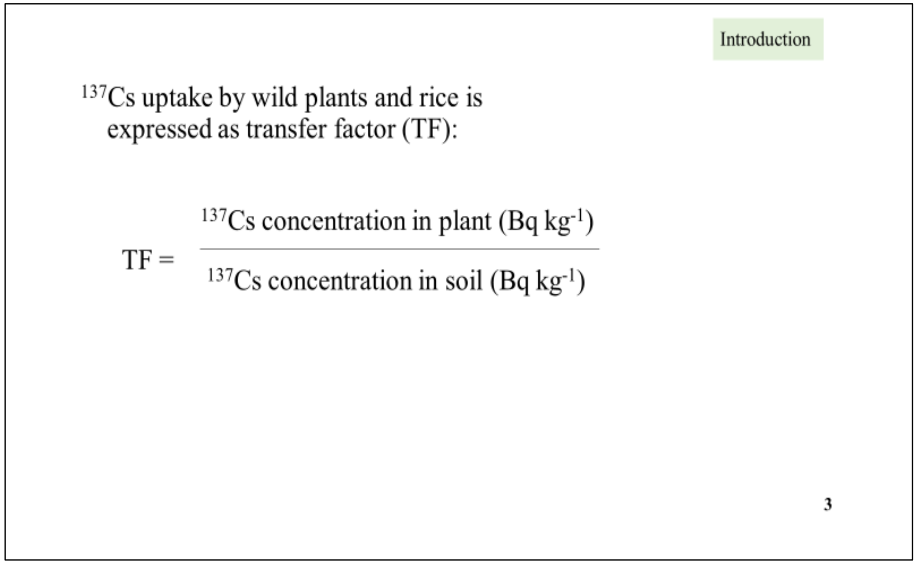
Radiocesium distribution in rice, wild plants, and soil following the Fukushima-Daiichi nuclear power accident

Nittaya Wakai
Graduate School of Environmental and Life Science
Okayama University
20 August 2019



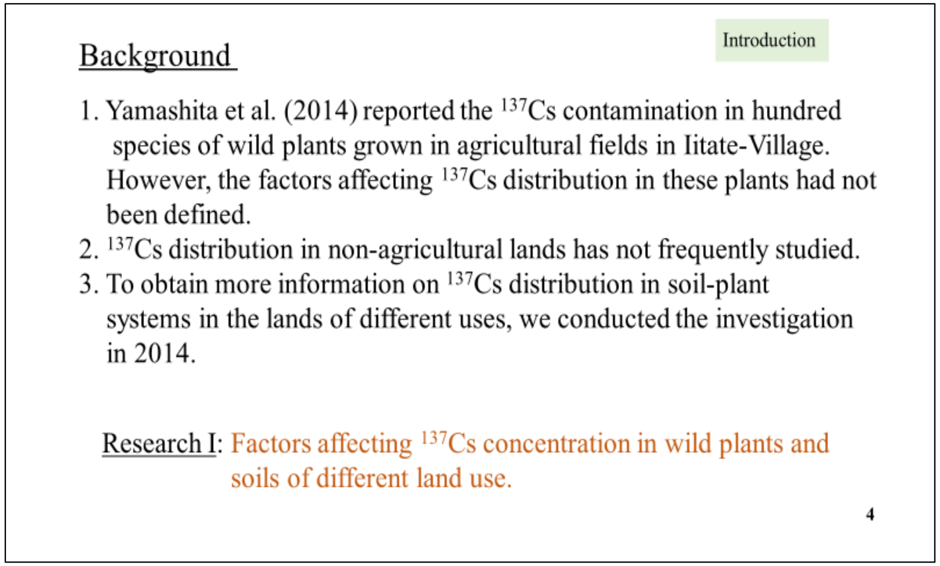
Introduction

1. The accidental damage to the Fukushima-Daiichi nuclear power plant (FD1NPP) in March 2011 released a large amount of ^{137}Cs into the environment.
2. ^{137}Cs (half-life: 30 years) emits β and γ rays during its decay.
3. Uptake of ^{137}Cs by crops is the pathway of ^{137}Cs enters human.
4. A year following the accident, Japanese authorities prohibited crop planting in agricultural soils containing ^{137}Cs $\geq$ 5000 Bq kg^{-1}.
5. The maximum limit of ^{137}Cs in general foods of Japan is 100 Bq kg^{-1})(since 2012).



^{137}Cs uptake by wild plants and rice is expressed as transfer factor (TF):

$$\mathrm{TF} = \frac{^{137}\mathrm{Cs\ concentration\ in\ plant\ (Bq\ kg^{-1})}}{^{137}\mathrm{Cs\ concentration\ in\ soil\ (Bq\ kg^{-1})}}$$



Background

1. Yamashita et al. (2014) reported the ^{137}Cs contamination in hundred species of wild plants grown in agricultural fields in Iitate-Village. However, the factors affecting ^{137}Cs distribution in these plants had not been defined.
2. ^{137}Cs distribution in non-agricultural lands has not frequently studied.
3. To obtain more information on ^{137}Cs distribution in soil-plant systems in the lands of different uses, we conducted the investigation in 2014.

Research I: Factors affecting ^{137}Cs concentration in wild plants and soils of different land use.

Background (continued)

3. Experimental paddy field in Kawamata-Town, the surface soil was removed and cattle manure compost was used to improve soil fertility.
4. Reducing ^{137}Cs in crop plants by applying K fertilizer has been well researched, but effects of compost has not been studied yet.
5. To obtain the information about the effects of cattle manure compost application at various rates on soil productivity and on ^{137}Cs distribution in rice plants, the study was conducted in 2015.

Research II: ^{137}Cs concentration in rice plant parts affected by cattle manure compost application

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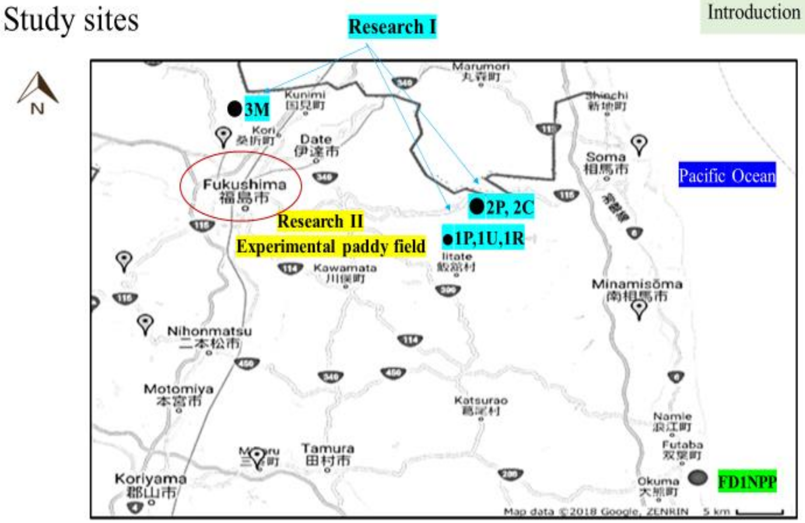
Study sites



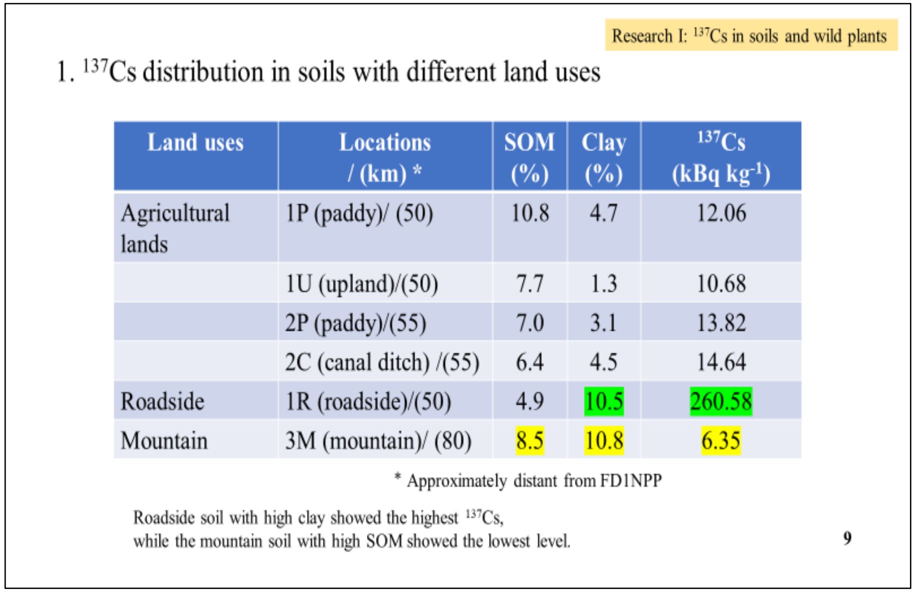
Research I: Factors affecting ^{137}Cs concentration in wild plants and soils with different land uses.

Objectives:

1) To estimate the ^{137}Cs contamination degree of soils and wild plants in the lands of different uses
2) To identify the factors affecting ^{137}Cs transfer in soils and wild plants ecosystems.

Material and methods

1. Three land use types; roadside, agricultural fields, and mountain.
2. Six species of wild plants and three soil samples were collected.
3. ^{137}Cs concentration in soil and plant samples was measured using a Ge semiconductor for 30 minutes per one sample.

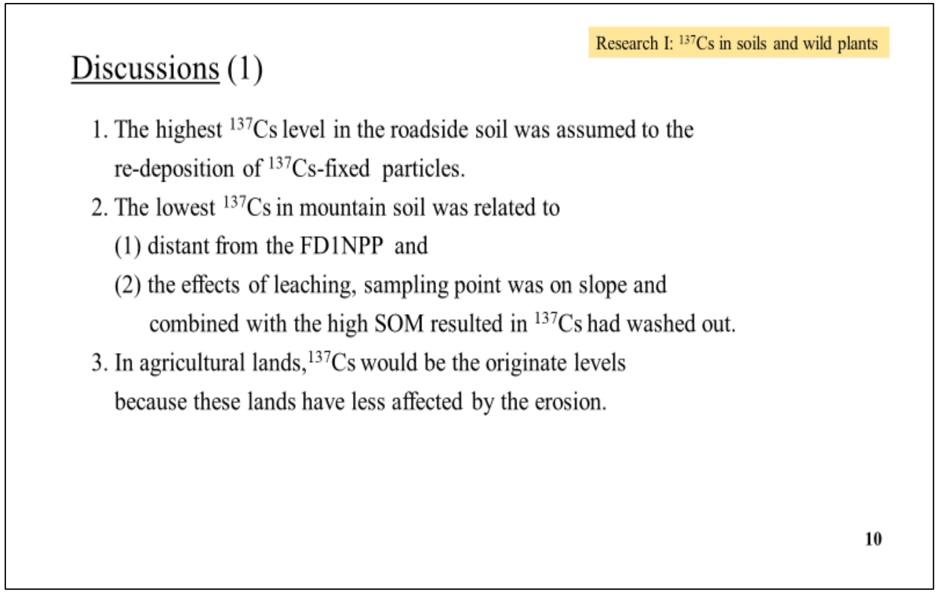


1. ^{137}Cs distribution in soils with different land uses

Land uses	Locations / (km) *	SOM (%)	Clay (%)	^{137}Cs (kBq kg^{-1})
Agricultural lands	1P (paddy)/ (50)	10.8	4.7	12.06
	1U (upland)/(50)	7.7	1.3	10.68
	2P (paddy)/(55)	7.0	3.1	13.82
	2C (canal ditch) /(55)	6.4	4.5	14.64
Roadside	1R (roadside)/(50)	4.9	10.5	260.58
Mountain	3M (mountain)/ (80)	8.5	10.8	6.35

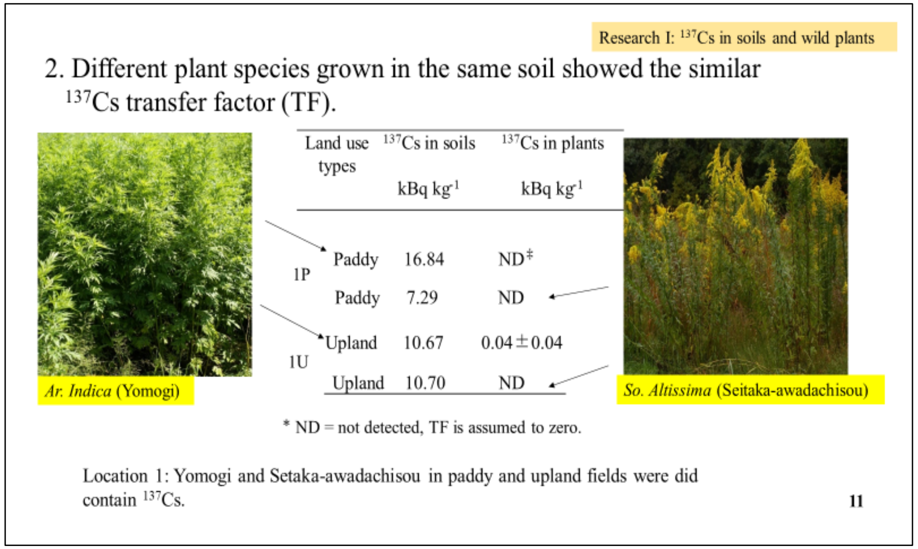
* Approximately distant from FD1NPP

Roadside soil with high clay showed the highest ^{137}Cs,
while the mountain soil with high SOM showed the lowest level.



Discussions (1)

1. The highest ^{137}Cs level in the roadside soil was assumed to the re-deposition of ^{137}Cs-fixed particles.
2. The lowest ^{137}Cs in mountain soil was related to
   (1) distant from the FD1NPP and
   (2) the effects of leaching, sampling point was on slope and combined with the high SOM resulted in ^{137}Cs had washed out.
3. In agricultural lands, ^{137}Cs would be the originate levels because these lands have less affected by the erosion.



2. Different plant species grown in the same soil showed the similar ^{137}Cs transfer factor (TF).

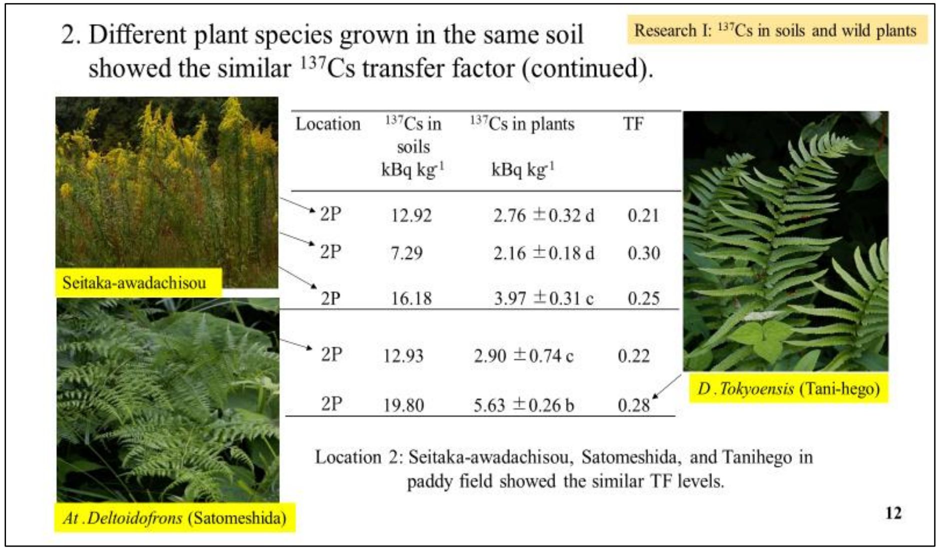
	Land use types	^{137}Cs in soils kBq kg^{-1}	^{137}Cs in plants kBq kg^{-1}
1P	Paddy	16.84	ND‡
	Paddy	7.29	ND
1U	Upland	10.67	0.04±0.04
	Upland	10.70	ND

* ND = not detected, TF is assumed to zero.

Ar. Indica (Yomogi)

So. Altissima (Seitaka-awadachisou)

Location 1: Yomogi and Setaka-awadachisou in paddy and upland fields were did contain ^{137}Cs.



2. Different plant species grown in the same soil showed the similar ^{137}Cs transfer factor (continued).

Location	^{137}Cs in soils kBq kg^{-1}	^{137}Cs in plants kBq kg^{-1}	TF
2P	12.92	2.76 ±0.32 d	0.21
2P	7.29	2.16 ±0.18 d	0.30
2P	16.18	3.97 ±0.31 c	0.25
2P	12.93	2.90 ±0.74 c	0.22
2P	19.80	5.63 ±0.26 b	0.28

Seitaka-awadachisou

At .Deltoidofrons (Satomeshida)

D .Tokyoensis (Tani-hego)

Location 2: Seitaka-awadachisou, Satomeshida, and Tanihego in paddy field showed the similar TF levels.

3. The same plant species grown the different soils showed different ^{137}Cs concentration and TF.

At . Yokoenscense (Hebino negosa)

Sampling points	^{137}Cs in soils kBq kg^{-1}	^{137}Cs in plants kBq kg^{-1}	TF
Canal	13.70	2.51 ±0.20 d	0.18
Canal	15.58	1.37 ±0.31 d	0.09
Mountain	6.35	3.71 ±2.98 c	0.58

Comparison between Hebinonegoza in 2 locations, TF of plant at the mountain (3M) was higher than that at Canal ditch (2C).

3. The same plant species grown in different soils showed different ^{137}Cs concentration and TF. (continued)

Comparison between Seitaka awadachisou at Location 1 (1P and 1U) and in Location 2 (2P). At Location 1 (1P and 1U) there were not ^{137}Cs detected, but that in Location 2 (2P) were found to high TF levels (0.2 to 0.3).

4. Wild plant grown in the soil with high ^{137}Cs concentration showed the high ^{137}Cs content, but TF was relatively low.

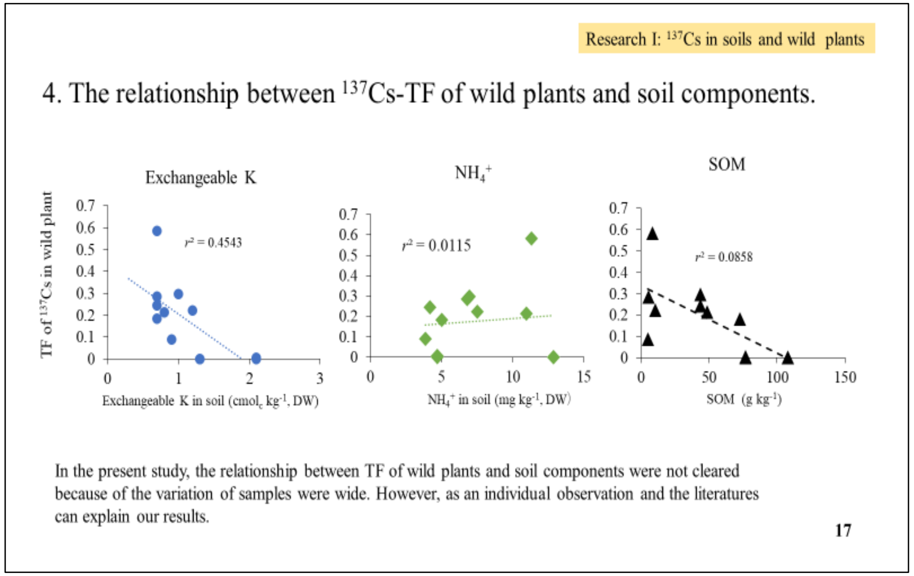
Se. samentosum (Tsuru mannengusa)

Location	^{137}Cs in soils kBq kg^{-1}	^{137}Cs in plants kBq kg^{-1}	TF
Roadside	260.58	21.90 ±1.76	0.08

This suggests that in soil with extremely high ^{137}Cs concentration TF value may not enough for interpret the ^{137}Cs uptake potential of plant.

Discussion (2)

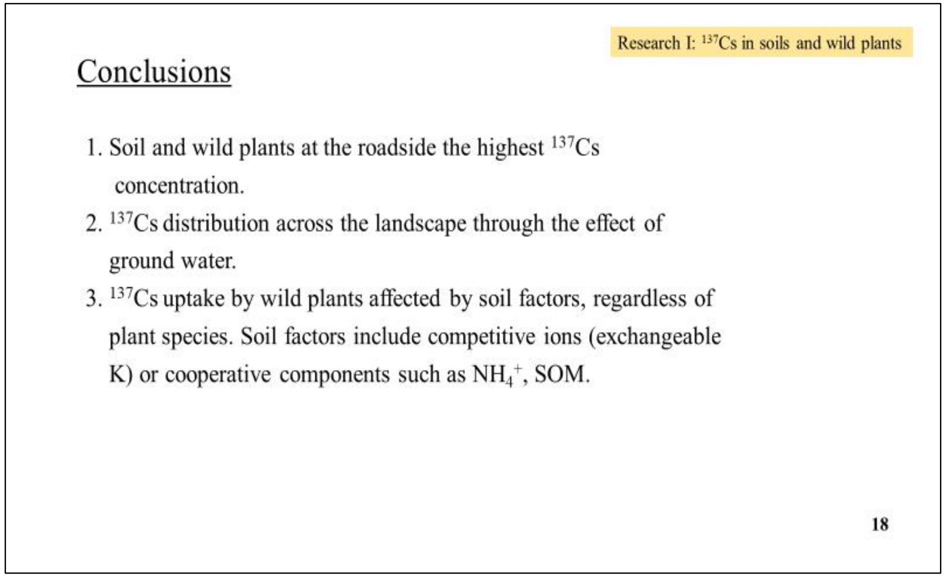
1. The potential of ^{137}Cs uptake of plants depending on soil factors regardless of plant species.
   (1) Seitaka awadachisou in 1P and 1U were not detected ^{137}Cs because these soils had a high ex-K. In contrast, that in 2P showed a high TF because the soil had a lower ex-K and high content of NH_4^+.
   (2) Hebinonegoza at the mountain showed higher TF than that at 2C because the soil had a relatively higher SOM and NH_4^+.
2. TF value may not enough to interpret the potential of ^{137}Cs uptake of plant in the extremely contaminated soil.
3. ^{137}Cs had re-distributed after its deposition.



Research I: ^{137}Cs in soils and wild plants

4. The relationship between ^{137}Cs-TF of wild plants and soil components.

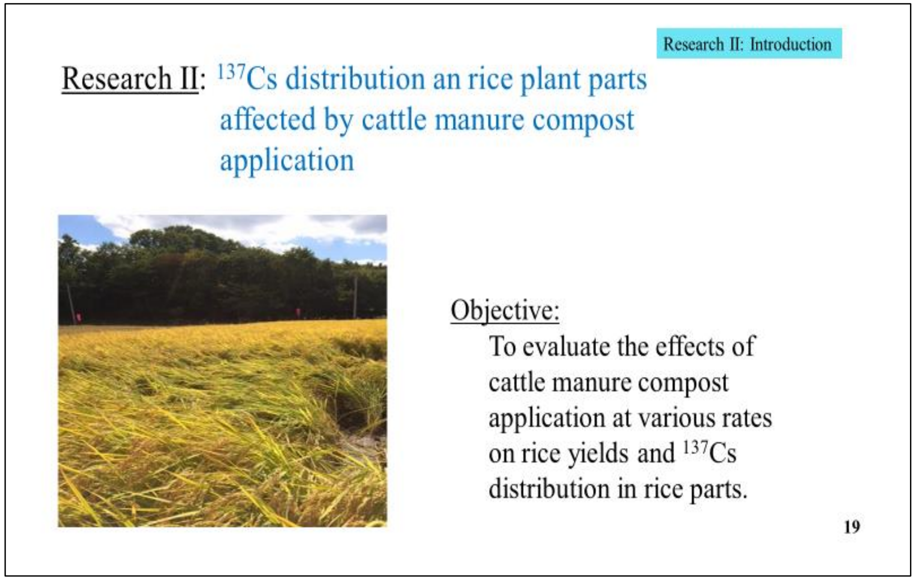
In the present study, the relationship between TF of wild plants and soil components were not cleared because of the variation of samples were wide. However, as an individual observation and the literatures can explain our results.



Research I: ^{137}Cs in soils and wild plants

Conclusions

1. Soil and wild plants at the roadside the highest ^{137}Cs concentration.
2. ^{137}Cs distribution across the landscape through the effect of ground water.
3. ^{137}Cs uptake by wild plants affected by soil factors, regardless of plant species. Soil factors include competitive ions (exchangeable K) or cooperative components such as NH$_4^+$, SOM.

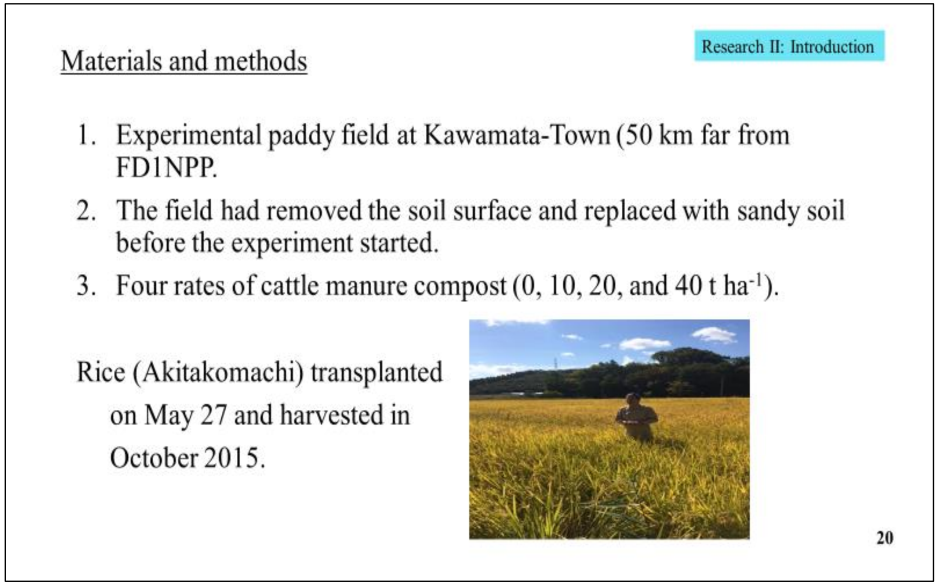


Research II: Introduction

Research II: ^{137}Cs distribution an rice plant parts affected by cattle manure compost application

Objective:

To evaluate the effects of cattle manure compost application at various rates on rice yields and ^{137}Cs distribution in rice parts.



Research II: Introduction

Materials and methods

1. Experimental paddy field at Kawamata-Town (50 km far from FD1NPP.
2. The field had removed the soil surface and replaced with sandy soil before the experiment started.
3. Four rates of cattle manure compost (0, 10, 20, and 40 t ha^{-1}).

Rice (Akitakomachi) transplanted on May 27 and harvested in October 2015.

Research II: Introduction

Materials and methods (continued)

4. Soil and rice samples were collected at harvest.
5. ^{137}Cs concentration in soil and rice parts (stems, leaves, and panicles) measured by a Ge semiconductor.
6. Measuring time for soil was 30 minutes and for rice was 24 hrs.

Chemical components of cattle manure compost.

Components	Dried weight bases
Total-N (%)	1.2
Total-C (%)	39.7
P_2O_5 (%)	0.86
CaO (%)	1.3
MgO (%)	0.6
K_2O (%)	3.13
Ash (%)	24.6

Research II: ^{137}Cs in rice

1. Soil chemical components increased with increase rates of compost.

Treatment t ha^{-1}	Total C mg kg^{-1}	Total N mg kg^{-1}	K cmol$_c$ kg^{-1}	Mg cmol$_c$ kg^{-1}	Ca cmol$_c$ kg^{-1}
0	10.7 b	0.96 b	0.17 b	0.82 b	6.41 b
10	15.6 ab	1.29 ab	0.41 ab	0.95 ab	7.47 ab
20	22.2 a	1.68 a	0.51 a	1.02 a	8.09 a
40	18.6 a	1.41 ab	0.42 ab	0.88 ab	6.73 ab

Application rates of 20 t ha^{-1} was significantly increased soil components.

Research II: ^{137}Cs in rice

2. Rice components increased with increase rates of compost.

Treatments t ha^{-1}	Total dry mass g m^{-2}	No. of panicles m^{-2}	No. of spiklets panicle^{-1}	Ripened grains %	1000 gains weight g	Grain yield g m^{-2}
0	1116	371	61	86.7	23.1	453
10	1139	366	64	85.0	22.9	458
20	1359	417	64	85.7	23.4	593
40	1499	438	67	86.9	23.7	601

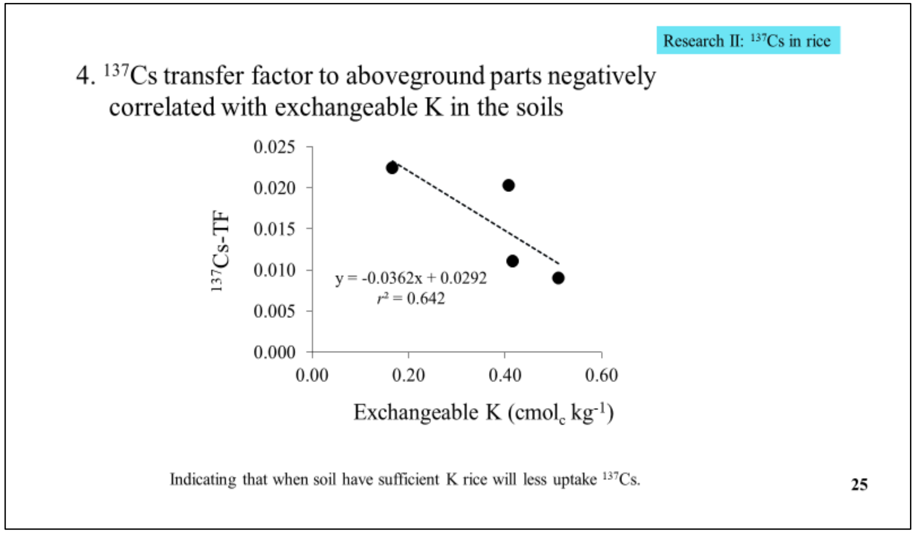
These were not significantly different among treatments, but increased by compost application.

Research II: ^{137}Cs in rice

3. Concentration of ^{137}Cs and transfer factor (TF) of rice plant parts.

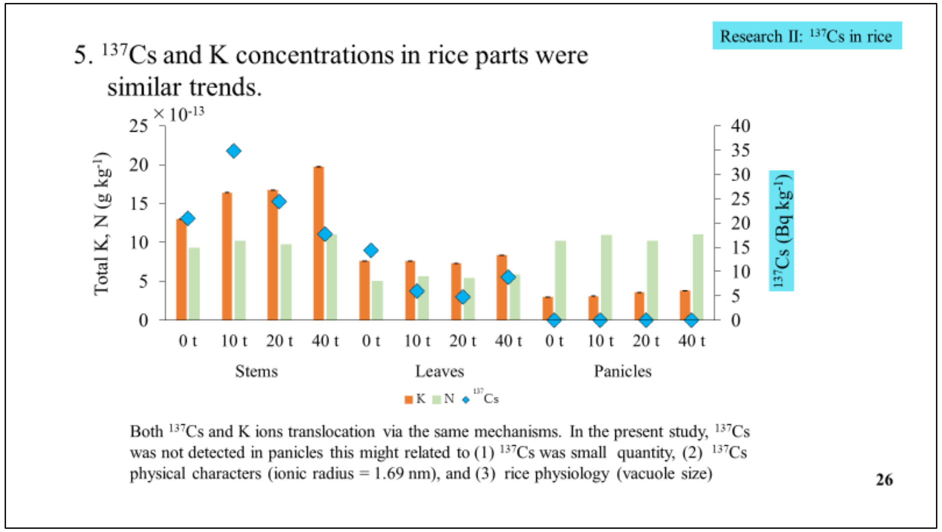
Treatments t ha^{-1}	Stems Bq kg^{-1}	Leaves Bq kg^{-1}	Panicles Bq kg^{-1}	TF of stems	TF of leaves	TF of aboveground parts
0	21	14	ND	0.050	0.034	0.022
10	35	6	ND	0.051	0.009	0.020
20	24	5	ND	0.024	0.005	0.009
40	18	9	ND	0.026	0.013	0.011

^{137}Cs in rice plants decreased by compost application.
TF was lowest in rice at 20 t ha^{-1} , which was 2.4 times lower than that at control plot.



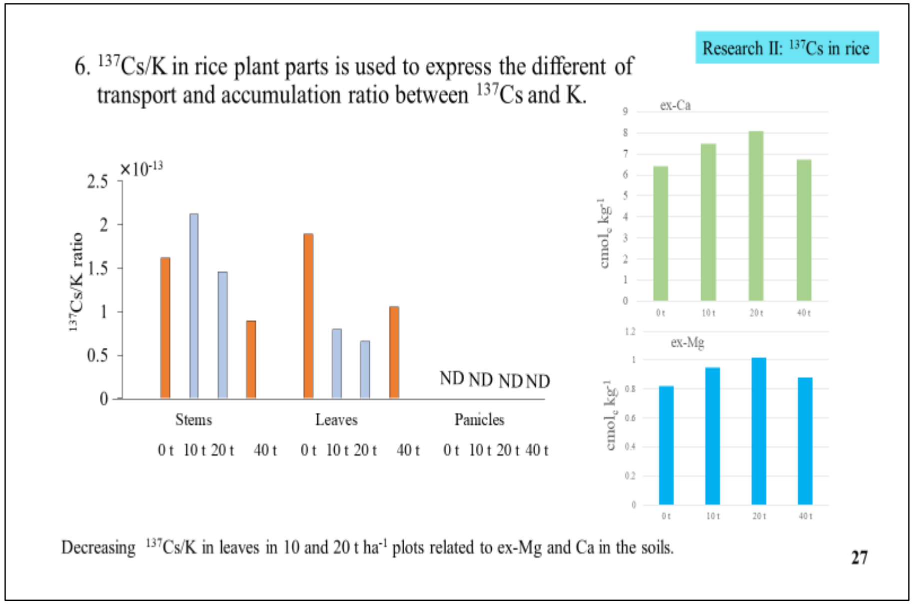
4. ^{137}Cs transfer factor to aboveground parts negatively correlated with exchangeable K in the soils

Indicating that when soil have sufficient K rice will less uptake ^{137}Cs.



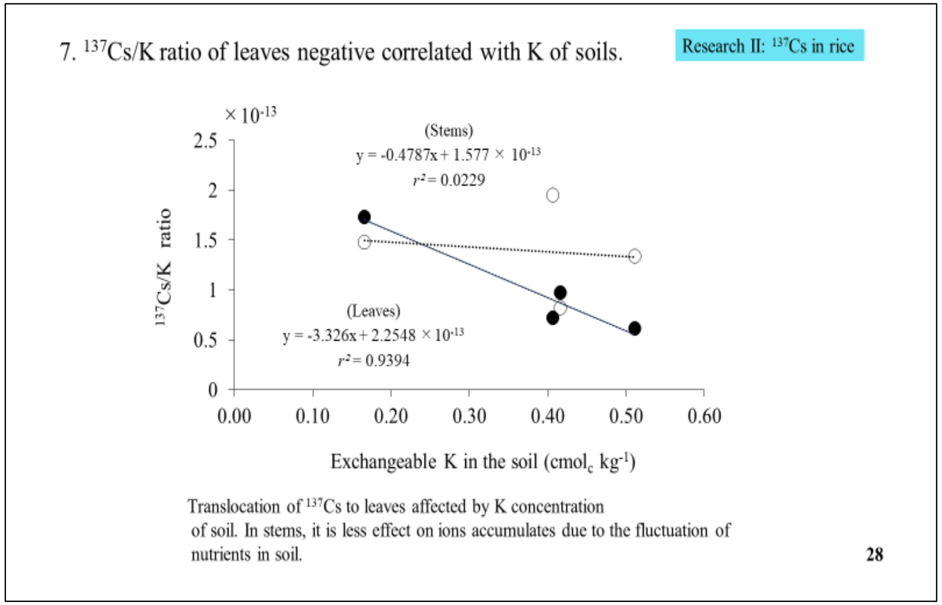
5. ^{137}Cs and K concentrations in rice parts were similar trends.

Both ^{137}Cs and K ions translocation via the same mechanisms. In the present study, ^{137}Cs was not detected in panicles this might related to (1) ^{137}Cs was small quantity, (2) ^{137}Cs physical characters (ionic radius = 1.69 nm), and (3) rice physiology (vacuole size)



6. ^{137}Cs/K in rice plant parts is used to express the different of transport and accumulation ratio between ^{137}Cs and K.

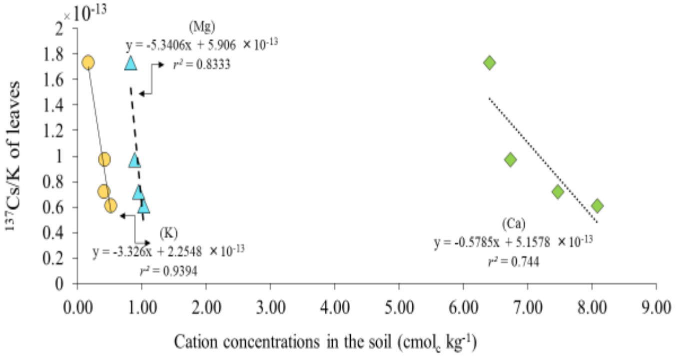
Decreasing ^{137}Cs/K in leaves in 10 and 20 t ha^{-1} plots related to ex-Mg and Ca in the soils.



7. ^{137}Cs/K ratio of leaves negative correlated with K of soils.

Translocation of ^{137}Cs to leaves affected by K concentration of soil. In stems, it is less effect on ions accumulates due to the fluctuation of nutrients in soil.

8. ^{137}Cs/K ratio of leaves negative correlated with cations in the soils.



Indicating that these cations affecting ^{137}Cs transfers (accumulates) to rice leaves.

Conclusion

1. Rice yields increased by cattle manure compost application.
2. Increasing of exchangeable K due to cattle manure compost application caused reduce ^{137}Cs uptake by rice.
3. ^{137}Cs and K distribution in rice plant parts were similar trends, stems > leaves > panicle (in the present study, ^{137}Cs was not detected in the panicles of all rice).
4. Exchangeable K, Mg, and Ca in soil affected ^{137}Cs distribution in rice plants.
5. Application rate of 20 t ha^{-1} was the most effective to both increasing the rice yields and reducing ^{137}Cs uptake by rice.

From our research

1. The information of ^{137}Cs contamination in soils of different uses might be useful for decontamination works.
2. In the ^{137}Cs contaminated agricultural lands, the contamination of food crops can avoided by soil management.
3. Application of compost increased soil productivity and reduced ^{137}Cs uptake by rice plants.
4. Effects of exchangeable Mg and Ca on ^{137}Cs translocation between rice parts should more studied.