# Report on "Radiation Disaster Recovery Studies"

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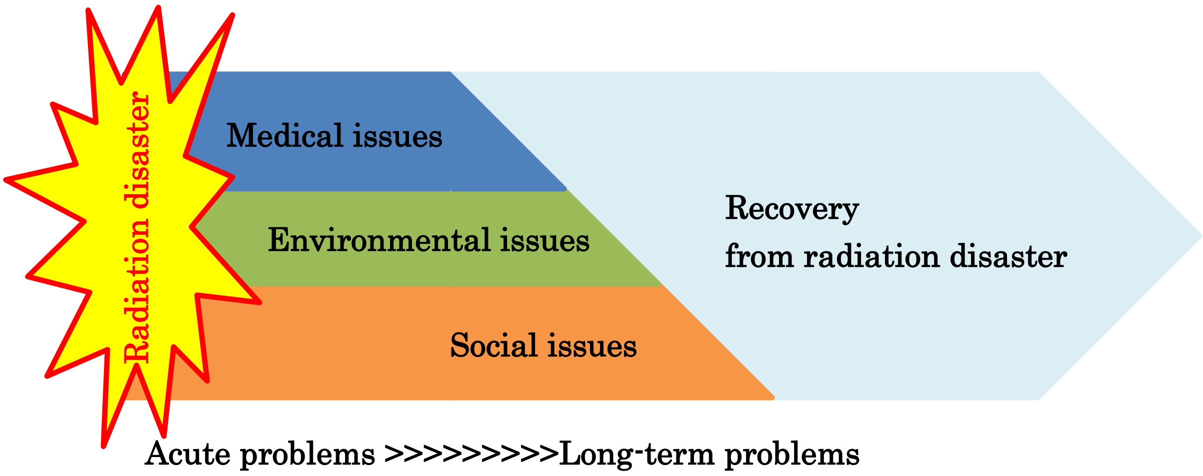
○Regarding "Radiation Disaster Recovery Studies"

Radiation disasters cause not only acute impact but also medium and long-term impact. It usually happens that both/either "exposure" by radiation and/or "contamination" by radionuclides become problems. When human bodies are exposed by radiation, the radiation causes ionization and excitation, and produces the secondary electron. This electron directory damages the DNA strand or produces the radical by reacting the water molecule which is also harmful for DNA strand. The human bodies have recovery mechanism of DNA strand's damages, but if DNA damages are caused more frequently by radiation with high dose rate, the damages cannot be recovered in time and it appears as functional obstacles (acute effect). On the other hand, cancerogenesis and genetic effect occur at a peak age of onset due to the accumulation of DNA mutation (late effect). On the other hand, if the radionuclides are released to the terrestrial environment, and if foods contaminated by radionuclides are taken, that causes the internal exposure. In particular, some isotopes have an affinity with organs and make them being continuously exposed. In Tokaimura nuclear accident occurred in 1999, three workers were severely exposed to neutron from critical uranium solution, which happened due to deviation of manufacturing manual, during the processing of nuclear fuel. Those who lived around the business office were also exposed and forced to evacuate. On the other hand, in Fukushima Daiichi Nuclear Power Station (FDNPS) accident occurred in 2011, FDNPS was collapsed by tsunami triggered by the East Japan Great Earthquake, and caused the hydrogen explosion. A lot of radionuclides were released into the terrestrial environment, and the residents were forced to evacuate and to stay in the temporary housing for a long time. Sudden change of life style and prolongation of situation led to high level of stress, and eventually caused some physical/mental diseases, such as diabetes and depression. These disadvantages were caused not by exposure or contamination directory, but by the situation of radiation disaster. In order to recover from radiation disaster, we need the different countermeasures in each phase of the disaster. In the initial stage, the protection of human life is taken on top priority. Existence or non-existence of those who are exposed by radiation or contaminated by radionuclides must be confirmed. In addition, it must be considered whether or not the residents should evacuate. In next (or same time), the types of radiation disaster must be determined (exposure and/or contamination). Since the radionuclides with short half-life decay soon, the analysis of environmental samples must be conducted immediately. At last, reassurance of people must be created through the risk communication and dialog with people based on the objective facts. The radiation disaster recovery is achieved in a stepwise approach.




**Conceptual Scheme of Radiation Disaster Recovery**

〇Title of Doctoral Thesis

Field Study on Transfer of Radioactive Cesium into Rice Plants Derived from Fukushima Daiichi Nuclear Power Station Accident

〇Summary of Doctoral Thesis

1. Introduction

Among the radionuclides released from FDNPS accident, cesium-137 ($^{137}Cs$) has a long half-life (30.17 years) considering the length of an average human life. Cesium-134 ($^{134}Cs$) has a comparatively short half-life (2.06 years), and was released in similar quantities to $^{137}Cs$. Cesium (Cs) itself is a homologous element with potassium (K), and Cs and K have similar physical and chemical properties. Thus, $^{137}Cs$ and $^{134}Cs$ may contaminate the agricultural products by replacing K. Since agriculture is the key industry of Fukushima Prefecture, and rice is a Japanese staple food, it is very important to ensure food safety. Rice contaminated by $^{137}Cs$ was observed in the private paddy farm in Fukushima City in 2013. The Japanese Society of Radiation Safety Management has investigated the cause of the contamination[1)]. In later years, we have started the studies about the factors contributing to the transfer of radioactive cesium into rice plants. We found some correlations between,

A. The grain size distribution of the soil and the transfer factor (TF) of radioactive Cs from the soil to rice plants.

B. The concentration of exchangeable cations and radioactivity concentration of radioactive Cs in the clay and silt fraction.

C. The concentration of exchangeable K in the clay and silt fraction and TF

D. The radioactivity concentration of potassium-40 ($^{40}K$) and TF in each part of rice plants, such as soil to rice roots, rice roots to leaves, and rice leaves to ears.

We believe that these are useful data as results of baseline study for developing the environmental countermeasures of radiation disaster recovery.

## 2. Experimental methods

### 2.1. Fieldwork

We conducted the fieldworks in the private farm located in northwest 60 km from FDNPS from 2014 to 2018. There are four rice fields neighboring each other (**Fig. 1**). For different aims, the fieldworks were conducted, such as the sampling of paddy soil and rice plants and/or measuring of rice plants' growth (**Table 1**).

**Table 1** Schedule of fieldworks

| **Year** | 2014 | 2016 | 2018 |
|---|---|---|---|
| **Month/Date** | 9/25 | 4/26, 5/18, 6/15, 7/5, 8/2, 8/24 and 9/24 | 3/14 |
| **Field** | A to D | C and D only | C and D only |
| **Sampling** | Soil and rice plant | Soil and rice plant | Soil only |
| **Measuring** | | Plant height and SPAD value | |

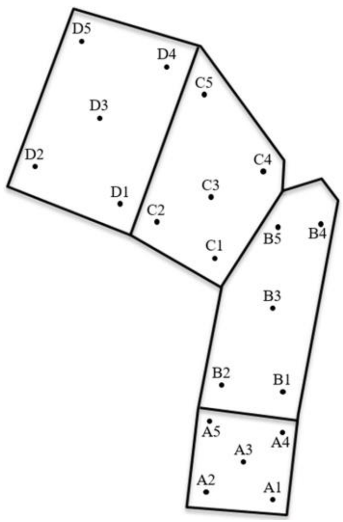



**Fig. 1** Position of rice fields

We collected the samples of the soil from the top 5 cm of soil and batch of rice plants of at a center and four corners in each rice field in each year. In 2016, the plant height was measured manually and the SPAD (Soil and Plant Analyzer Development) values were measured by a SPAD-502Plus chlorophyll meter at each sampling point.

### 2.2. Classification of grain size

To make the uniform soil samples, the gravel fraction (> 2mm) was removed by dry sieving after drying at room temperature in the laboratory for a few days. The soil samples obtained in 2014 were separated into more fractions for further analysis of grain size distribution according to "the

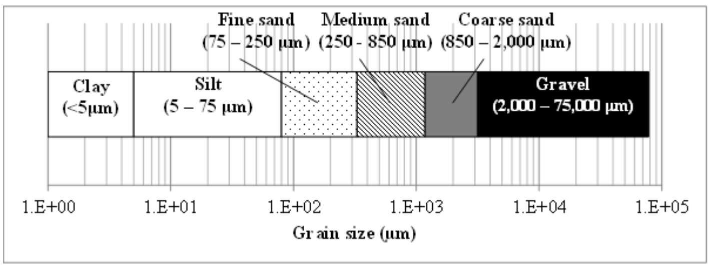



**Fig. 2** Classification of grain size

method of classification of geomaterials for engineering purpose" by The Japanese Geotechnical Society (JGS0051) (**Fig. 2**).

## 2.3. Radioactivity measurement

The soil and rice plant samples were packed in U8 vessels and their radioactivity concentration of $^{137}Cs$, $^{134}Cs$ and $^{40}K$ was measured by Ge semiconductor detector. The radioactivity concentration was calculated by a following equation. The detection efficiency was estimated by measuring of the standard source set. The correction constant of sum effect of $^{134}Cs$ was estimated by measuring of the soil samples obtained in the same fieldwork site.

$$A = (C - B) \times S \times \frac{1}{D} \times \frac{100}{E} \times \frac{1}{exp(-\lambda \times t)} \times \frac{1}{M}$$

***A***: the radioactivity concentration on the sampling day (Bq kg$^{-1}$)

***C***: the counts per second of gamma-ray (cps) (= net counts/live time)

***B***: the counts per second of gamma-ray of $^{40}K$ background (cps) (= 0.0053) for calculation of $^{40}K$

***S***: the correction constant of the sum effect of $^{134}Cs$ (= 1.129 ± 0.007 as the standard error) for calculation of $^{134}Cs$

***D***: the detection efficiency

***E***: the emission rate of gamma-ray (%)

***λ***: the decay constant (s$^{-1}$) (=0.693/half-life)

***t***: the elapsed time from sampling to measurement (s)

***M***: the mass of the sample in U8 vessel (kg)

## 2.4. Transfer factor (TF)

The transfer factor (TF) is the ratio of radioactivity concentration (a) divided by (b).

(a) Radioactivity concentration after the transfer (rice ears)

(b) Radioactivity concentration before the transfer (paddy soil)

For the samples in 2016, three-phase TFs were calculated. $TF_1$, $TF_2$, and $TF_3$ are the TF from soil to roots, from roots to leaves, and from leaves to ears, respectively.

## 2.5. Measurement of exchangeable cations

The concentration of four exchangeable cations, such as sodium (Na), magnesium (Mg), potassium (K) and calcium (Ca), in the clay and silt fraction, which were separated from the soil samples in 2014, were measured by ICP-AES. The exchangeable cations were extracted from about 5 g of clay and silt by 1 M ammonium acetate. The extraction was conducted three times (1st: 40 ml for overnight, 2nd and 3rd: 30 ml for 1 hour). The molar concentration of each exchange cation was calculated by a following equation.

$$MC = EC \times V \times I / AW / M$$

***MC*** : concentration of molar equivalent of exchangeable cations (mmol$_c$ kg$^{-1}$)

***EC*** : concentration of exchangeable cations (ppm ≒ mg L$^{-1}$)

***V*** : volume (L)

***I*** : valence of ion

***AW*** : atomic weight (g mol$^{-1}$)

***M*** : mass of samples (kg)

## 3. Results and discussion

### 3.1. Correlation between grain size distribution of paddy soil and TF

The fraction of clay and silt was rich in field C and D that that of A and B. The small fraction may be carried from the upstream (A and B) to the down (C and D) (**Fig. 3**). The concentrations of $^{137}$Cs and $^{134}$Cs in field C and D were higher than that of A and B by approx. 20%. On the other hand, the concentration of $^{40}$K in field B was especially lower than the others by approx. 25%. In field B, the TF of Cs-137 and Cs-134 was 1.5 ~ 2 times higher than the others (**Fig. 4**).

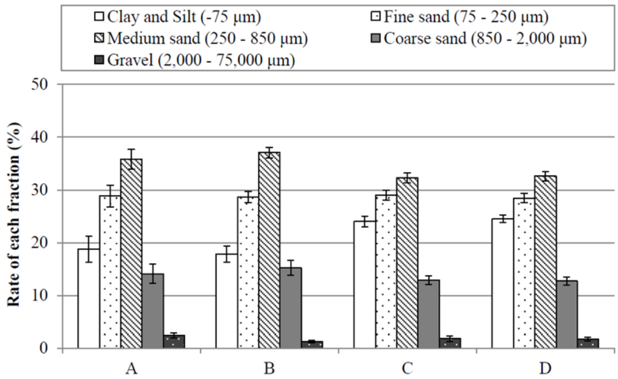



**Fig. 3** Grain size distribution of paddy soil in 2014

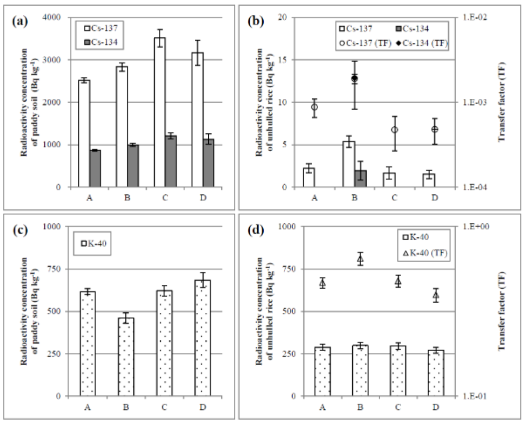



**Fig. 4** Radioactivity concentration of paddy soil and rice ears and TF in 2014

When the percentage of clay and silt increased, or medium sand relatively decreased, the TF became lower (**Fig. 5**). It is conceivable that the small fraction, such as clay and silt, includes much minerals having a layer structure known as "fried edge site" where cesium ion is fixed strongly. In the field where the clay and silt fraction is enriched, the radioactive cesium is fixed in the paddy soil, and it is difficult to transfer to rice ears.

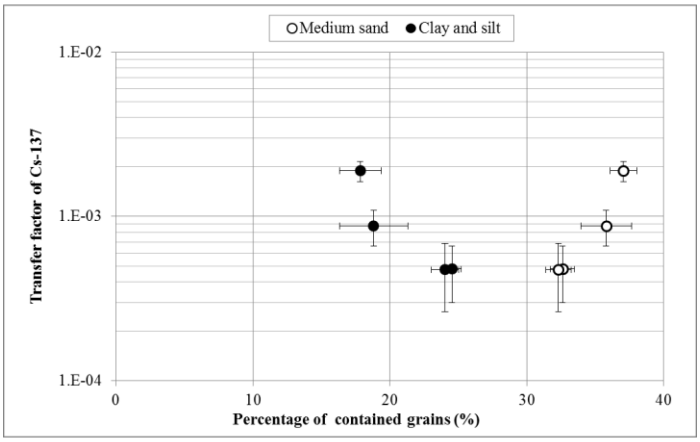



**Fig. 5** Correlation b/w grain size distribution and TF in 2014.

### 3.2. Correlation between the concentration of exchangeable cations and radioactivity concentration

In the clay and silt fraction in 2014, there is a competitive correlation between the total concentrations of exchangeable cations (Na, Mg, K, Ca) and radioactivity concentration of radioactive Cs ($^{137}$Cs, $^{134}$Cs) (**Fig. 6**). However, the inverse number of the concentration of exchangeable K in the clay and silt fraction was proportional relation with TF (**Fig. 7**). The exchangeable K behaves competitively with radioactive Cs on the transfer into rice plants, while the exchangeable cations do on the placing in the small fractions of paddy soil.

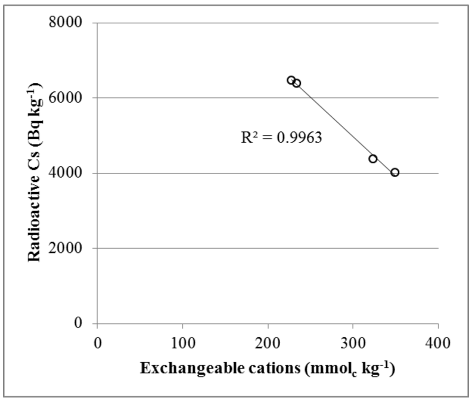



**Fig. 6** Correlation b/w exchangeable cations and radioactive Cs in the clay and silt fraction in 2014.

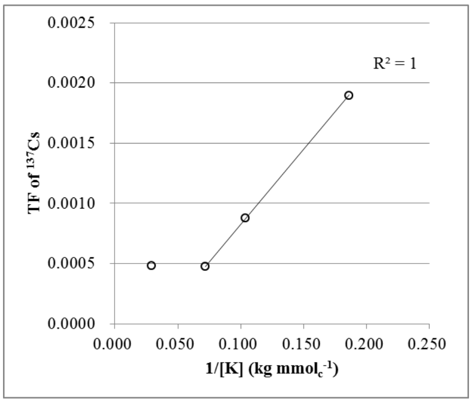



**Fig. 7** Correlation b/w exchangeable K in the clay and silt fraction and TF in 2014.

3.3. Correlation between three-phase TFs and radioactivity concentration of $^{40}K$

Transfer factor 1 of $^{137}Cs$ and $^{134}Cs$ from the soil to roots became high when $^{40}K$ was poor in the soil, while $TF_2$ of $^{137}Cs$ from roots to leaves and $TF_3$ of $^{137}Cs$ from leaves to ears became high when the radioactivity concentration of $^{40}K$ in the roots and leaves were also high, respectively. It suggested that existence of K is competitive on the absorption of Cs by roots, but corporative on the transportation of Cs inside of rice plants.

**(a)** $TF_1$ and $^{40}K$ in the soil **(b)** $TF_2$ and $^{40}K$ in the roots **(c)** $TF_3$ and $^{40}K$ in the leaves

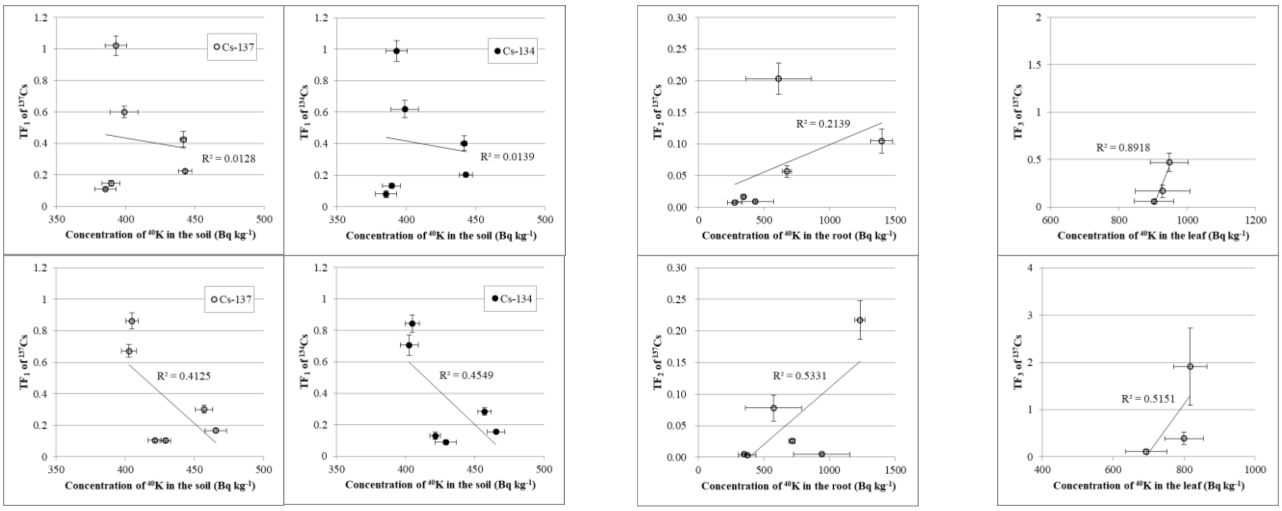



**Fig. 8** Correlation b/w three-phase TFs and radioactivity concentration of $^{40}K$ in each place (upper: Field C, lower: Field D)

3.4. Correlation between three-phase TFs and rice plants' growth

The $TF_1$ increased with the rice plants' growth when the plant height increased or SPAD value decreased, but $TF_2$ and $TF_3$ decreased (**Fig. 9 and 10**). In the early stage of the rice plants' growth, the roots is not likely to absorb the radioactive Cs while the once absorbed radioactive Cs is transferred to upper parts of rice plants, such as leaves.

**(a)** $TF_1$ and plant height **(b)** $TF_2$ and plant height **(c)** $TF_3$ and plant height

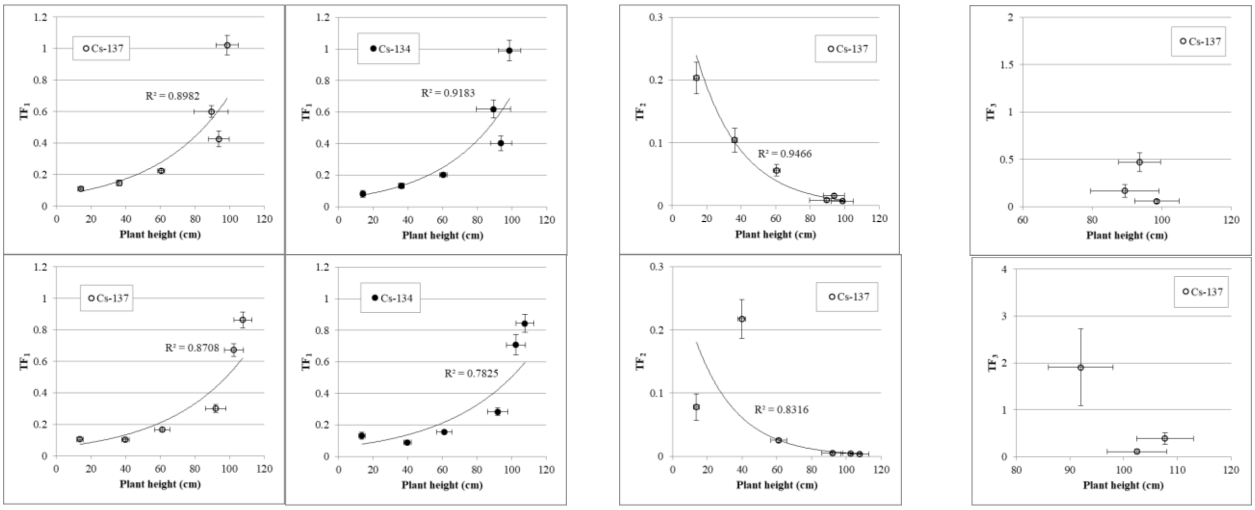



**Fig. 9** Correlation b/w three-phase TFs and plant height (upper: Field C, lower: Field D)

**(a)** $TF_1$ and SPAD value **(b)** $TF_2$ and SPAD value **(c)** $TF_3$ and SPAD value

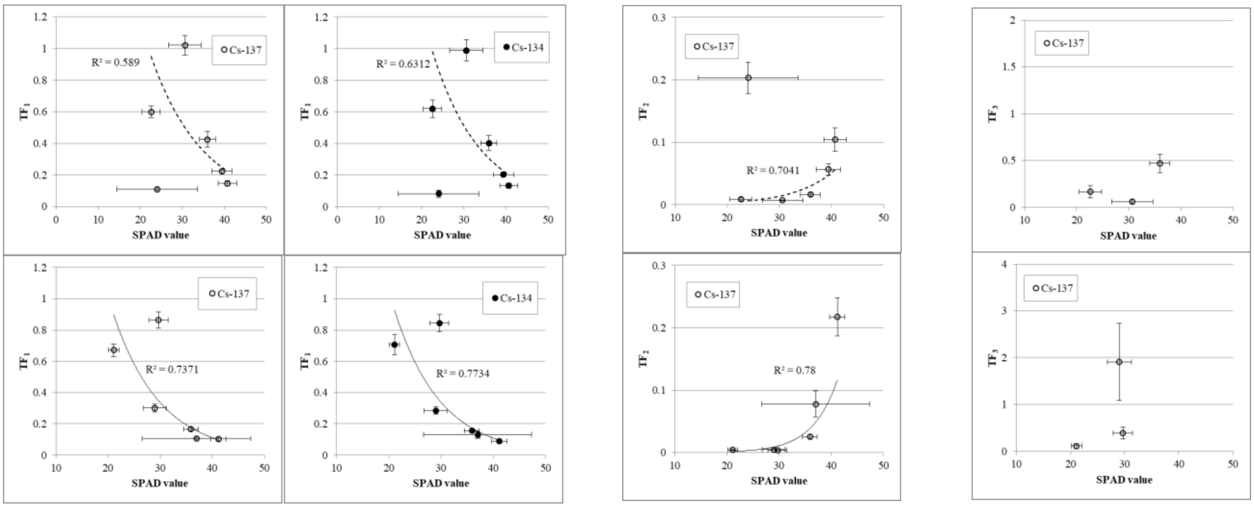



**Fig. 10** Correlation b/w three-phase TFs and SPAD value (upper: Field C, lower: Field D)

3.5. Yearly decay of radioactivity of radioactive Cs

The concentration of $^{137}$Cs and $^{134}$Cs decreased according to their half-life (**Fig. 11**). It can be estimated that 3,500 Bq kg$^{-1}$ of $^{137}$Cs and $^{134}$Cs (7,000 Bq kg$^{-1}$ in total) remained in both field C and D in March 2011.

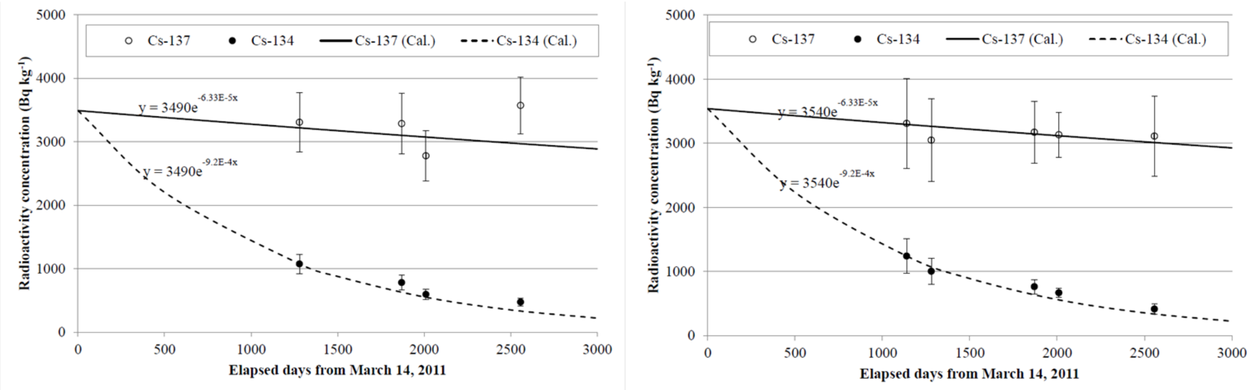



**Fig. 11** Comparison of radioactivity concentration of $^{137}$Cs and $^{134}$Cs in the paddy soil with the estimated values (left: Field C, right: Field D)

## 4. Conclusion

Transfer factor of radioactive Cs from paddy soil to rice plant is subject to some parameters as below.

(1) the grain size distribution of soil (percentage of clay and silt fraction)

(2) the concentration of exchangeable cations (in particular, K) in clay and silt fraction

(3) the radioactivity concentration of $^{40}$K

Distribution of KCl for the enrichment of K ion in paddy has been recommended to prevent the absorption of Cs by rice plant. But K inside of rice plant helps Cs to transfer into upper parts, such as leaves and ears, which is edible part for human. Since the radioactivity concentration of Cs decreases by no reason other than own decay according to its half-life, the remove of small fraction of soil and/or rice roots after harvesting is effective decontamination to reduce the concentration of Cs in soil.

## Reference

1) Matsuda, N. and Nakashima, S.: Radioactive caesium in water and soil and its absorption by rice plant (in Japanese). Japanese J. Rad. Safety. Manag., 13, 89-91, 2014

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(2) Tsujimoto, M., Miyashita, S., Nguyen, T. H., and Nakashima, S.: Monthly change in radioactivity concentration of $^{137}$Cs, $^{134}$Cs, and $^{40}$K of paddy soil and rice plants in Fukushima Prefecture, Radiation Safety Management (accepted in February, 2020).