A correlation between the transfer factor of radioactive cesium from soil into rice plants and the grain size distribution of paddy soil in Fukushima

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The transfer factors (TFs) from paddy soil to rice plants of radioactive cesium (^{137}Cs and ^{134}Cs) derived from Fukushima Daiichi Nuclear Power Plant (FDNPP) accidents in March, 2011 and grain size distributions were estimated in four paddy fields, A through D, in Fukushima City. Soil and rice samples were obtained in September, 2014. The soil was measured for ^{137}Cs and ^{134}Cs radioactivity by Ge semiconductor detector and separated by the dry sieving classification. The radioactivity of radioactive cesium in the soil was comparatively higher in fields C and D (> 4000 Bq/kg), but the TF in field B was the highest (1.9E-03) among the four fields. Meanwhile, the soil in fields A and B contained a relatively large amount of larger grains, such as medium sand. Grain size distribution might be related to the TF. This study shows a new correlation between the grain size distribution and TF.

Key words: radioactive cesium, paddy soil and rice, grain size distribution, transfer factor, Fukushima

1. Introduction

We are apprehensive about a migration of radioactive cesium (^{137}Cs and ^{134}Cs) from paddy soil into rice plants since Fukushima Daiichi Nuclear Power Plant (FDNPP) accident. The East Japan Great Earthquake, which occurred on March 11th, 2011, caused the unimaginable tsunami and FDNPP accident. A lot of radioisotopes were released due to some hydrogen explosions in FDNPP and the ground and environment were contaminated.[1, 2] Radioactive cesium, which is one of radioisotopes obtained as a fission product in a nuclear reactor, has a comparatively long half-life (the half-lives of ^{137}Cs and ^{134}Cs are approximately 30 and 2 years, respectively). It stays in the environment for a long time. Meanwhile, it is known that cesium itself is absorbed to biological bodies as well as potassium because both of the elements are grouped into alkali metal and behave similarly. It is known that the decontamination of radioactive cesium from soil containing smaller grains (< 250 μm) is difficult[3], and a computational chemistry revealed that radioactive cesium can hold a place in a frayed edge site instead of potassium and be fixed strongly.[4] For a few years after the accident, the radioactivity of radioactive cesium in rice produced from Fukushima Prefecture was investigated and the excessive level of the radioactive cesium in some rice was detected.[5] The migration of radioactive cesium from soil to rice plants has been investigated. The transfer factor of radioactive cesium into brown rice was estimated.[6] Furthermore, a correlation between the concentrations of the radioactive cesium and potassium in rice plants and transfer factors were investigated.[7] But, there might be many factors to influence the transfer factor of radioactive cesium. It is also very important to investigate how a grain size distribution as a soil character affects the transfer factor of the radioactive cesium. However, a correlation

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between transfer factor of radioactive cesium and grain size distribution has been unrevealed. It is conceivable that radioactive cesium migrates into rice plants from larger grains in paddy soil, given smaller grains have the specific site, such as the frayed edge site, and fix radioactive cesium so strongly.

Contaminated rice was observed in the rice from the paddy field in Fukushima City, which is northwest, approximately 60 km far from FDNPP. The situation was investigated by Subcommittee concerning chemical treatment, Japanese Society of Radiation Safety Management in 2013.[8] In the present study, we actually have visited paddy fields in Fukushima and obtained soil and rice samples. We investigated how the grain size distribution in the soil influences the transfer factor.

2. Materials and methods

2-1. Sampling and preparations

Paddy soil and rice plant samples were obtained in four paddy fields in Oguraji, Fukushima City in September, 2014, which are located northwest approximately 60 km far from FDNPP and in Abukuma river basin (Fig. 1). The paddy fields are irrigated mainly from a pond, but a little water is supplied from Abukuma river. We sampled five soil samples from the surface (0–5 cm) at the center and four corners and single rice plant sample at the center in each field (A through D). All samples were, first, dried at room temperature for one day, and then the soil samples were dried completely at 105 °C for 24 hours by a convection oven (NDO-401W, EYELA Tokyo Rikakikai Co., LTD.). The mass of the soil samples was measured by an electronic balance (GH series, Kensei Co., LTD.) before and after complete drying in order to calculate a moisture ratio. The moisture ratio was calculated in a following equation.

The moisture ratio (%) =

$$\frac{\text{The mass of water contained in the soil sample* (g)}}{\text{The mass of the soil sample before complete drying (g)}} \times 100$$

*The mass of water contained in the soil sample
= (The mass of the soil sample *before* complete drying) − (The mass of the soil sample *after* complete drying)

In the meantime, unhulled rice was removed from rice plant samples and contained in U-8 vessels. Its mass was measured by the electronic balance.

2-2. Dry sieving

2-2-1. *Classification of soil*

Soil can be classified by its grain size. We categorized the
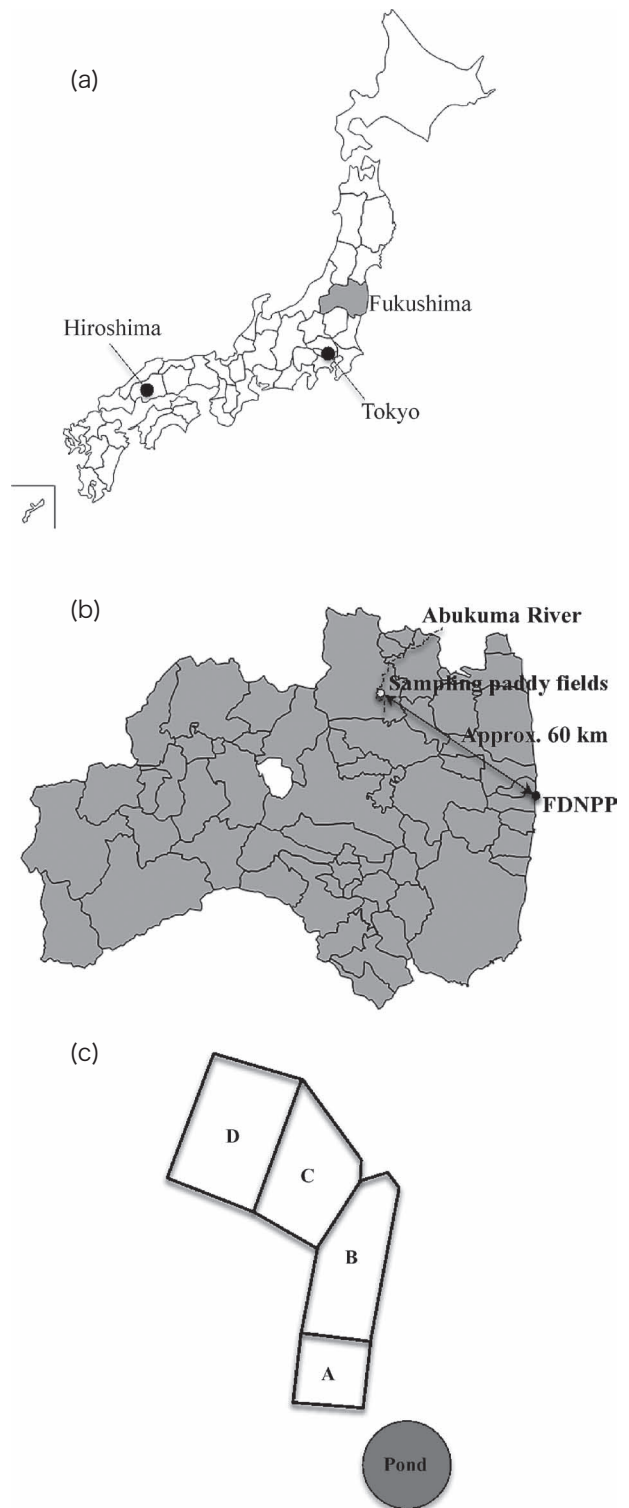

Fig. 1. (a) The location of Fukushima Prefecture in Japan. (b) Positional relationships among sampling paddy fields, Abukuma River, and FDNPP. (c) A view of the paddy fields and a pond as a source of water.

Table 1 The mass of sea sands and Montmorillonite mixed as the test samples.

Test	Sea sand (g)			Montmorillonite	Total mass	Sieving area	Sieving area density
No.	1400–850 μm	850–425 μm	600–300 μm	(g)	(g)	(cm²)	(g/cm²)
X1	19.642	19.668	19.596	19.701	78.607		1.00
X2	19.604	19.592	19.619	19.999	78.814	78.5	1.00
Y1	2.088	1.962	2.002	1.925	7.977		0.10
Y2	1.955	1.966	1.969	1.989	7.879		0.10

soil samples in accordance with the method of classification of geomaterials for engineering purposes by The Japanese Geotechnical Society (JGS0051). The soil samples with a grain size X (X < 5 μm, 5 μm < X < 75 μm, 75 μm < X < 2 mm, and 2 mm < X) were grouped into clay, silt, sand, and gravel, respectively. The sand samples were additionally grouped into fine, medium, and coarse sand by a grain size Y (75 μm < Y < 250 μm, 250 μm < Y < 850 μm, and 850 μm < Y < 2 mm).

2-2-2. *Pretest for dry sieving*

To optimize experimental conditions, a dry sieving test by using sea sands and a clay mineral was done. The sea sands whose grain size is 300–600 μm, 425–850 μm, and 850–1400 μm (191–15955, 196–08175, and 190–11405, Wako Pure Chemical Industries, Ltd.), and Montmorillonite (281522, Sigma-Aldrich Co. LLC.) were mixed as test samples of soil containing sand and clay and passed through a 10 cm diameter, 75 μm mesh size testing sieve (JIS Z 8801, NONAKA RIKAKI Co., LTD.). The mass of the mixed sea sands and Montmorillonite is shown in Table 1. The mass of the test samples per sieving area was adjusted to 1 g/cm² and 0.1 g/cm². The dry sieving was performed in a shaker (FMC-100, EYELA Tokyo Rikakikai Co., LTD.) for 10, 20, 30, 60, and 120 min, and a sieving ratio was calculated in a following equation after sieving in each time.

The sieving ratio (%) =

$$\frac{\text{The mass of Montmorillonite passed through the sieve in the time (g)}}{\text{The mass of Montmorillonite mixed in the test sample (g)}} \times 100$$

2-2-3. *Dry sieving classification*

The soil samples were separated by using the 20 cm diameter sieves (JIS Z 8801, SANPO Co., LTD.) as previously mentioned in chapter 2-2-1. This dry sieving was performed with the appropriate quantities of the soil samples (< 0.1 g/cm²) for
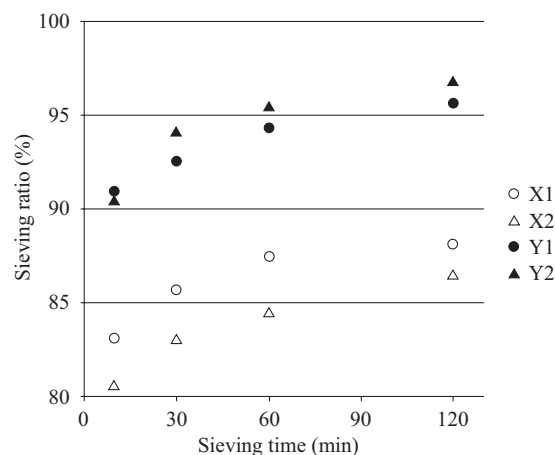

Fig. 2. Results of the dry sieving test. X and Y stand for the results of the sieving of 1 g/cm² and 0.1 g/cm²-test samples, respectively. Two tests were performed in each condition. White and black circles and triangles stand for test number X1, X2, Y1, and Y2, respectively, shown in Table 1.

enough time (> 3 hours) (Fig. 2). After sieving, each grain (clay and silt, fine sand, medium sand, coarse sand, and gravel) were contained in U-8 vessels, and the mass of each grain was measured by the electronic balance. The result was plotted on a grain size accumulation curve.

2-3. Measurement of radioactivity

Radioactivity of ^{137}Cs, ^{134}Cs, and ^{40}K in the soil and unhulled rice samples was measured by Ge semiconductor detector (GEM 30-70, ORTEC). The radioactivity of the soil samples was measured in advance before dry sieving and that of sand after dry sieving was done as well. The radioactivity in the soil in each field was estimated as an average of that of the five soil samples. The radioactivity in the rice was done as a result of that of the single unhulled rice sample. To calculate the radioactivity of ^{137}Cs and ^{134}Cs, the 662 and 794 keV γ-rays were targeted to be detected, respectively. The measurement was

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performed for enough time until the standard error of NET count of 794 keV γ-rays becomes smaller than 5 %. The count efficiency, which comes from the differences of heights of the sample sources and energies of the detected γ-ray, is estimated by measuring a radioactivity standard gamma volume source set (MX033U8PP, Japan Radioisotope Association). The count efficiency of the 794 keV γ-ray was calculated by logarithmic and logarithmic interpolation between the count efficiency of the 662 keV (derived from ^{137}Cs) and 835 keV (derived from ^{54}Mn) γ-ray. In addition, in order to estimate a coefficient, which calibrates the underestimation of ^{134}Cs radioactivity because of Sum effect (a Sum peak formed by 605 and 794 keV derived from ^{134}Cs), some soil samples were placed 20 cm and 1 cm from the detector to measure their radioactivity. This coefficient to calibrate Sum effect was calculated in a following equation.

The coefficient to calibrate Sum effect =

$$\frac{\text{The ratio of } ^{134/137}\text{Cs radioactivity at 20 cm from the detector}}{\text{The ratio of } ^{134/137}\text{Cs radioactivity at 1 cm from the detector}}$$

The decay correction gave the radioactivity of ^{137}Cs and ^{134}Cs on the date of sampling as the result.

2-4. Calculation of transfer factors

Transfer factors in which the radioactive cesium migrated from the paddy soil to rice plants were calculated in a following equation.

The transfer factor =

$$\frac{\text{The radioactivity in the unhulled rice (Bq/kg)}}{\text{The radioactivity in the soil before sieving (Bq/kg)}}$$

2-5. Handle of errors

For the case that the two and more data were obtained as a result, a standard error was calculated from a standard deviation as an external error. The data of radioactivity of ^{137}Cs, ^{134}Cs, and ^{40}K in the soil had standard errors, and internal errors were also calculated by conforming to the rule of error propagation. Larger one of them was adopted (Table 2(a) and (b)).

3. Results and discussion

3-1. Dry sieving test

Montmorillonite could be separated from the test samples by over 95% for up to 2 hours when the dry sieving tests were performed with the test samples of 0.1 g/cm^2 (Fig. 2). On the other hand, only less than 90% of Montmorillonite could be separated from the test samples of 1 g/cm^2.
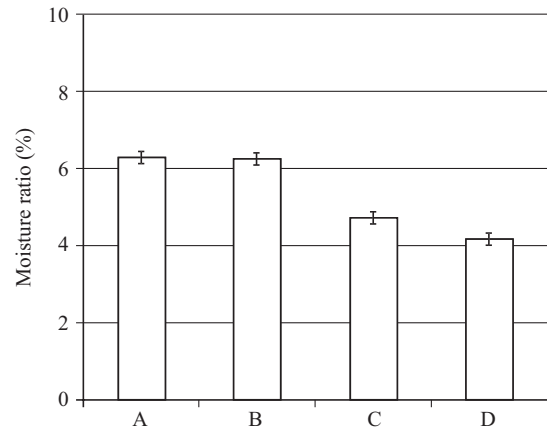

Fig. 3. Moisture ratio of soil in paddy fields A through D. The error bars show the standard deviation.

3-2. Profile of soil and irrigation water

The soil in the fields A and B included slightly more water than that of C and D (Fig. 3). This may be because the fields A and B are located close to a pond, which supplies the irrigation water, and the fields C and D are done in the ends of the running water. This locational fact might be also able to explain the difference among the grain size distributions (Fig. 1c). According to the grain size accumulation curves, a composition of soil is similar between A and B, and between C and D (Fig. 4(a)). The soil in the fields A and B had slightly less clay and silt than that of C and D. The running water might carry the clay and silt from the fields A and B to C and D. However, medium sand abounded in the fields A and B than C and D (Fig. 4(b)). Whereas the soil was composed chiefly of sand this time, in our experiment, some clay and silt might be still attached on surface of sand. Additionally, according to the previous study, radioactivity of ^{137}Cs and ^{134}Cs in the both of irrigation water of the pond and Abukuma river makes little differences and is sufficiently low.[8]

3-3. Radioactivity in the soil and rice and transfer factor

The radioactivity of ^{137}Cs and ^{134}Cs in the soil before sieving in the fields C and D was higher than that of A and B, and the radioactivity in the sand after sieving denoted the same tendency (Fig. 5(a) and (b)). However, the radioactivity of ^{137}Cs and ^{134}Cs in the unhulled rice in the field B was specifically the highest and the transfer factor in the field B was also as well (Fig. 5(c)). Although the transfer factor of ^{137}Cs in the field A was placed second behind that of B, the radioactivity of ^{134}Cs in the rice in the field A, C, and D could not be estimated due to the size of

Table 2(a)   The radioactivity of ^{137}Cs, [134]Cs, and [40]K in the soil before sieving and comparison of external and internal errors.

Fields	Radioactivity of cesium-137 (Bq/kg)	Error (Bq/kg)		Radioactivity of cesium-134 (Bq/kg)	Error (Bq/kg)		Radioactivity of potassium-40 (Bq/kg)	Error (Bq/kg)	
		External	Internal		External	Internal		External	Internal
A	2523	61	54	872	21	23	616	7	18
B	2835	100	62	992	36	28	460	31	26
C	3513	204	77	1207	73	35	621	19	31
D	3169	294	71	1131	119	35	684	37	44

The radioactivity (Bq) per dry weight (kg) of the samples was calculated.

Table 2(b)   The radioactivity of ^{137}Cs, [134]Cs, and [40]K in the sand after sieving and comparison of external and internal errors.

Fields	Radioactivity of cesium-137 (Bq/kg)	Error (Bq/kg)		Radioactivity of cesium-134 (Bq/kg)	Error (Bq/kg)		Radioactivity of potassium-40 (Bq/kg)	Error (Bq/kg)	
		External	Internal		External	Internal		External	Internal
A	2209	65	49	745	20	22	623	34	36
B	2604	125	58	904	47	27	541	29	35
C	2866	169	64	1004	71	31	648	22	45
D	2447	243	57	900	96	30	756	65	56

The radioactivity (Bq) per dry weight (kg) of the samples was calculated.

error. At any rate, the radioactivity of ^{137}Cs and [134]Cs between the soil and unhulled rice was not simply correlated with each other. It was inspired that the higher radioactivity of radioactive cesium in soil does not always cause more migration of radioactive cesium into rice plants. Meanwhile, the lack of [40]K was found in the soil in the field B (Fig. 5(d)). The same tendency was confirmed in sand (Fig. 5(e)), though the radioactivity of ^{137}Cs, [134]Cs, and [40]K was different: the radioactivity of ^{137}Cs and [134]Cs in the total soil was 0.8–0.9 times higher in the sand, but that of [40]K in the total soil was 1.0–1.2 times higher in the sand. The transfer factor of [40]K was the highest in the field B (Fig. 5(f)) because rice plants absorb the determinate amount of potassium independent of the concentration in the paddy soil.

3-4. Correlation between the transfer factor of ^{137}Cs and grain size distribution

The amount of medium sand in the soil influenced the migration of the radioactive cesium into the rice plants. The transfer factor of ^{137}Cs was high in the field where the soil was composed mainly of medium sand and, in contrast, less silt and clay (Fig. 6). The transfer factor was relatively higher in the fields A and B where the soil contained more large grains, such as medium sand (and less small grains, such as clay and silt). As the specific site to adsorb the cesium, say frayed edge site, abounds in small grains in soil, it is presumable that the migration of the radioactive cesium into rice plants from soil containing the small grains is unfavorable. Just for the record, the moisture ratio of the soil in the fields A and B was also, coincidentally, relatively higher as well as the transfer factor, though water is likely to be a carrier of radioactive cesium.

4.   Conclusion

It has been thought that a concentration of potassium in paddy soil can mainly contribute to a transfer factor of radioactive cesium into rice plants. And, in our knowledge, a correlation between a grain size distribution of paddy soil and a transfer
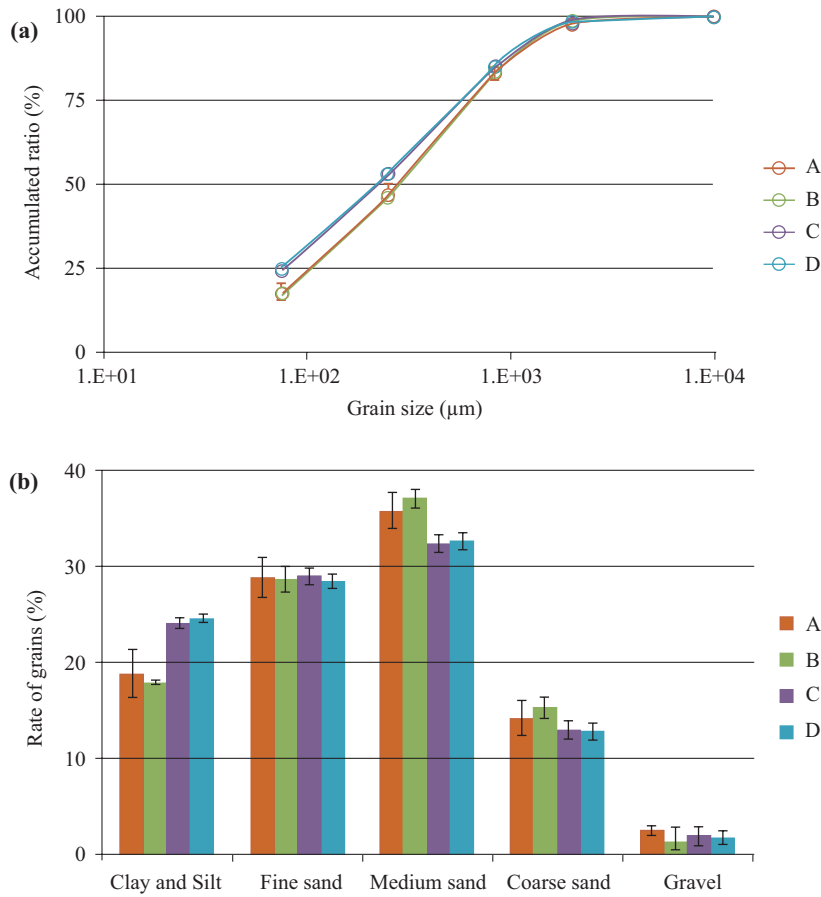
Fig. 4.   (a) Grain size accumulation curves. (b) Rate of each grain contained in the soil. The error bars show the standard error.
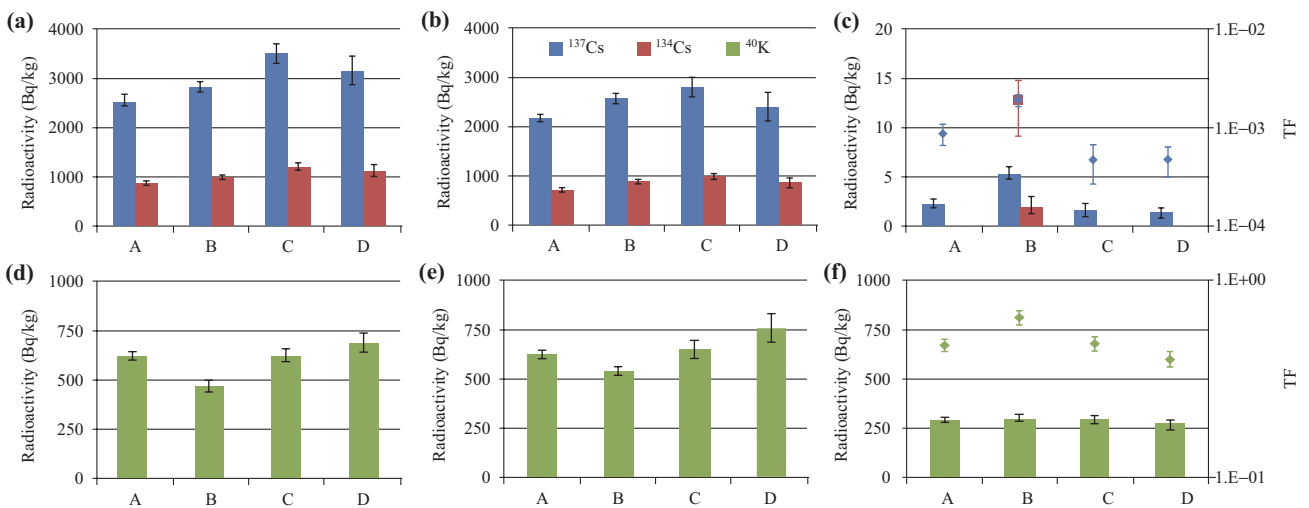

Fig. 5.   The results of radioactivity and transfer factor (TF) of ^{137}Cs and ^{134}Cs (a) in the soil before sieving, (b) in the sand after sieving, and (c) in the rice. (d), (e), and (f) show those of ^{40}K, respectively. The error bars show the standard error.
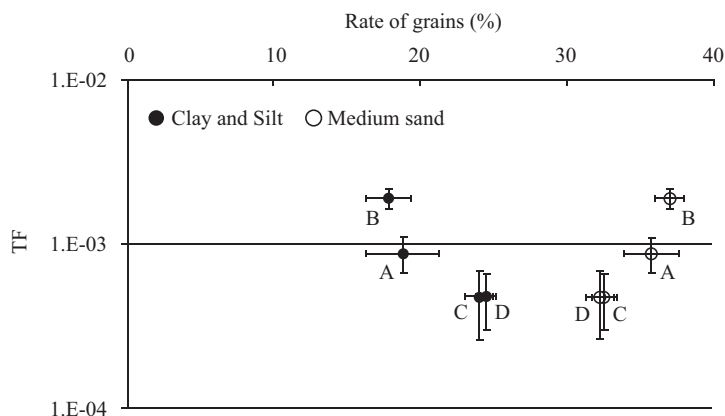
Fig. 6. A correlation between the transfer factor of ^{137}Cs and rate of grains. The error bars show the standard error.

factor has been unrevealed. However, this paper could have given a consideration for this correlation. In this study, we actually used the samples of the soil and rice plant obtained in Fukushima. The dry sieving classification has been performed adequately based on the result of the pretest, and the radioactivity of the radioactive cesium has been measured and calculated properly with the revisions as far as we can see. We found the differences of the moisture ratio and grain size distribution with a few % among the soil samples in fields A through D, even though the locations of these fields neighbor to each other. And moreover, there were the difference in the radioactivity of cesium among fields and the lack of potassium in field B. The radioactivity of cesium was not corresponded with the difference in the transfer factors. They were affected by the concentration of potassium and difference in the grain size distributions. The migration of radioactive cesium from paddy soil to rice plants easily occurs in a poor-potassium field.[7] In our experimental fields, the radioactivity of [40]K in the field B was comparatively low, and the transfer factor of the radioactive cesium was high. It exactly supports the previous study. However, the transfer factor of radioactive cesium in the field A was also relatively higher than that of C and D, even though these fields likely had an enough concentration of potassium (the radioactivity of [40]K was more than 600 Bq/kg in these fields). The rate of sand, especially medium sand in soil, was also one of key factors to determine the transfer factor of radioactive cesium from soil to rice plants. These findings provided fresh insight into the migration of radioactive cesium into rice plants. Additionally, this study indicated a possibility to prevent radioactive cesium from moving to rice plants by controlling the grain size distribution in paddy soil. For example,

decreasing the proportion of medium sand in paddy fields, by design, might be able to block the migration of radioactive cesium from soil into rice in spite of the loss of potassium and the contamination level of radioactive cesium, though it should be considered whether that grain size distribution is appropriate for growing rice plants. However, decline of the radioactivity over time will be shown, and the continued observations are necessary. Therefore, future research should include follow-up works how the transfer factor, grain size distribution and their correlation will change.

5.   Acknowledgement

6.   References

1) Iwata, H., Shiotsu, H., Kaneko, M., and Utsunomiya, S.: Nuclear accidents in Fukushima, Japan, and exploration of effective decontamination for the ^{137}Cs-contaminated soils, Advances in nuclear fuel Chap. 6 (2012).

2) Endo, S., Kimura, S., Takatsuji, T., Nanasawa, K., Imanaka, T., Shizuma, K.: Measurement of soil concentration by radionuclides due to Fukushima Dai-ichi Nuclear Power Plant accident and associated cumulative external dose estimation, J. Environm. Radioact., 111, 18–27 (2012).

3) Nikolaevsky, V. B., Lifanov, F. A., Batusov, S. S.: Improving decontamination of ^{137}Cs contaminated soil, Atomic Energy. 114, 51–55 (2013).

4) Okumura, M., Nakamura, H., Machida, M.: Mechanism of strong affinity of clay minerals to radioactive cesium: first-principles cal-

culation study for adsorption of cesium at frayed edge sites in Muscovite, J. Phys. Jpn., 82, 033802 (2013).

5) Nihei, N., Tanoi, K., Nakanishi, T. M.: Inspections of radiocesium concentration levels in rice from Fukushima Prefecture after the Fukushima Dai-ichi Nuclear Power Plant accident, Scientific Reports. 5, 8653 (2015).

6) Uchida, S., Tagami, K.: Soil-to-plant transfer factors of fallout ^{137}Cs and native ^{133}Cs in various crops collected in Japan, J.

Radioanal. Nucl. Chem., 273, 205–210 (2006).

7) Endo, S., Kajimoto, T., Shizuma, K.: Paddy-field contamination with ^{134}Cs and ^{137}Cs due to Fukushima Dai-ichi Nuclear Power Plant accident and soil-to-rice transfer coefficients, J. Environm. Radioact., 116, 59–64 (2013).

8) Matsuda, N., Nakashima S.: Radioactive cesium in water and soil, and its absorption to rice plant, Jpn. J. Radiat. Safety Manag. 13, 84–91 (2014), (in Japanese).