Annual Reduction of Transfer Factor of Radiocesium from Soil to Rice Cultivated in a KCl Fertilized Paddy Field from 2015 to 2019

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We performed consecutive field trials of rice cultivation to monitor radiocesium contamination in harvested rice from 2012, in the Iitate Village in Fukushima Prefecture, where people were forced to be evacuated due to high level of radioactive contamination caused by the disaster at the Fukushima Dai-ichi Nuclear Power Plant of Tokyo Electric Power. The early year results (2012–2013) showed the radiocesium concentration in the brown rice was reduced depending on the decontaminated level of paddy soil and on the exchangeable K content of the soil. This report of later year results (2015–2019) showed further more than 80% reduction of \(^{137}\text{Cs}\) concentration in the brown rice and straw at KCl fertilized paddy soil, in spite of little reduction of \(^{137}\text{Cs}\) concentration of the soil. The transfer factor of \(^{137}\text{Cs}\) from soil to brown rice reduced from 0.0022 in 2015 to 0.0003 in 2019 and that to straw reduced from 0.0262 in 2015 to 0.0028 in 2019, respectively. Exchangeable positive ions of the soil were also analyzed. Multiple regression analyses of all data of transfer factor in 2015 to 2019 to year (ageing) and exchangeable K ion as variables shows that the main causal factor is year (ageing) with some supportive effect of increase of exchangeable K ion. This implicates that radiocesium in soil was gradually transformed to a form more difficult to be absorbed by rice, that is, \(^{137}\text{Cs}\) immobilization or fixation on clay minerals by ageing, not only in early years after the accident (2011–2015), but also in later years (2015–2019). This implication was supported by comparative analysis of exchangeable \(^{137}\text{Cs}\) of dry soil of 2017, 2018 and 2019.

Key Words: radioactive fallout, soil, rice, caesium-137, transfer factor, Fukushima Dai-ichi Nuclear Power Plant Accident

1. Introduction

Rice is the most important crop in Japan. The Fukushima Dai-ichi Nuclear Power Plant (FDNPP) Accident in March 2011 caused radioactive materials to spread out and down onto farm lands from the Plant of Tokyo Electric Power Company. In the Iitate village in Fukushima Prefecture, people were evacuated from June 2011 to March 2017, except Nagadoro Ward still evacuated. In the village we performed consecutive field trials of rice cultivation and monitored radiocesium contamination in rice from 2012 to judge how we can cultivate rice in the village, without fear of radioactive contamination. The early
year results (2012–2013) showed KCl fertilization can reduce the transfer factor (TF) of radiocesium in brown rice from soil to 0.003–0.004 in 2012 when the exchangeable K content in soil was higher than 20 mg/100 g (as K2O). From 2014, all bags of brown rice harvested at Sasu test field have passed the Fukushima Prefecture inspection (below the detection level of 25 Bq/kg with screening measurement). However, consumers prefer food of less radiocesium contamination. Then we continued to cultivate rice and monitor the radiocesium concentration of brown rice and straw. Similar results of importance of the exchangeable K content were reported by Fujimura et al. at five paddy fields in Fukushima Prefecture in 2012–2014. Further Yamamura et al. proposed a statistical model for estimating the radiocesium TF from soil to brown rice using the soil exchangeable K content, based on the field data obtained from 2012 to 2015. From their model, they also estimated the year factor and a yearly decline rate of 17%.

On the other hand, Roig et al. tested the conditions of long-term radiocesium trapping (ageing) by clay soil from the Chernobyl area, and Yamashita et al. reported yearly decrease in the TF of radiocesium from soil to grasses from 2012 to 2015 on a pasture in Iwate Prefecture. Tsukada reported that the ratio of radiocesium in the exchangeable fraction decreased from 12.8% in April of 2012 to 6.9% in October of 2013 in a paddy field in Fukushima. Further Fujimura et al. showed 40% and 30% decrease in TF in brown rice and inedible rice part from 2011 to 2012, employing pot experiments using soil samples from Fukushima, suggesting an irreversible sorption of 137Cs to clay minerals. Tagami et al. also showed the geometric mean of 137Cs TF of brown rice to soil from paddy fields without additional K decreased from 0.012 in 2011 to 0.0035 in 2013. Most recently Wakabayashi et al. reported 137Cs ageing assessed by the ratios of exchangeable 137Cs/133Cs in a field study of a rice paddy in allophanic Andosol in Tsukuba from 2011 to 2015. All the data reported are from those in 4 years after the accident. Here, we report yearly decrease in the TF of 137Cs from soil to brown rice and straw from 2015 to 2019, and suggest that the decrease is mainly due to ageing effect.

2. Materials and methods

2.1 Test field, Rice cultivation, and Sampling

Rice cultivation is performed at the same field at Sasu test field (N37°44′, E140°43′) as reported in April 2012 by shallow irrigation and the muddy water swept out from North-East to South-West as reported. The soil is classified as an Andosol (D2z1) by Japanese soil inventory system (http://soil-inventory.dc.affrc.go.jp). The field was partially decontaminated in April 2012 by shallow irrigation and the muddy water swept out from North-East to South-West as reported.1,2) The soil radiocesium concentration was 2,000 to 6,000 Bq/kg of dry weight. Further decontamination was not performed thereafter. Rice seedlings of Akitakomachi (2012–2013), Hitomebore (2014–2017), Koshihikari (2018–2019) were planted in the fields. Rice radiocesium absorption is not significantly different among these cultivars. Table 1 summarize rice cultivation procedures including fertilizers used from 2014 to 2019. Rice planting was performed in May or early June. In 2014, a basal fertilizer (18N-15P-15K; weight% as N, P2O5, and K2O, 50 kg/10 a) was employed and K fertilizer was added as KCl (60% as K2O: 20 kg /10 a) to south-east half area (K test field) and mixed with the plowed soil before planting. In 2015, the basal fertilizer (18N-15P-15K, 50 kg/10 a) and KCl (20 kg /10 a) were added to whole field. In 2016, the same as in 2015, except an additional fertilizer of NK (17N:17K) 2–3 kg/10 a was added in July. In 2017, the same as in 2016, except the addition of Ca silicate (50 kg/10 a) as a soil improver and a basal fertilizer (18N-15P-15K, 50 kg/10 a) was used. In 2018 and 2019, the same as in 2017, except for no soil improver added and the amount of additional
NK fertilizer (17N-17K) of 4 to 5 kg/10a in July. After sampling by hand, harvest was performed in October and the remnant of straw was plowed in the paddy field. In September 10 in 2015, typhoon 18 caused the brook overflow at the south-west side, and the soil and sand covered part of the south-west side but not to the sampling points in the field and soil and rice sampling was performed at 23 days after the flood. In 2015 to 2019, soil and rice sampling was performed between late in September and early in October at the 5 points (M₁, M₂, M₃, M₄, M₅) of the test field, as shown in Fig. 1. The soil of 0–15 cm at each point was taken into a plastic bag with a long scoop, and 10–15 sheaves of rice plant were cut and collected. The bundles of rice plant were dried in doors for more than 1 week, and unhulled rice were prepared by a thresher and then brown rice sample was prepared with a hulling machine. The straw sample was prepared by cutting the straw part by 1 cm, and further dried for more than a week.

Fig. 1 The test field of Sasu and the 5 sampling points. (A) The picture of rice harvest with hand, in October 6 of 2019, the next day of the sampling (sampling points M1–5 shown). (B) Diagram of the sampling points (1–5) (Color online).

| Year | Species     | Basal Fertilizer                                                                 | Additional Fertilizer in July | Straw plowed in | Sampling Date | Note                                                                 |
|------|-------------|---------------------------------------------------------------------------------|------------------------------|-----------------|--------------|----------------------------------------------------------------------|
| 2014 | Hitomebore  | Fertilizer (18N-15P-15K) 50kg/10a; KCl 20kg/10a (KCl addition south-east half area only) | No                           | Yes             | Oct.5        |                                                                      |
| 2015 | Hitomebore  | Fertilizer (18N-15P-15K) 50kg/10a; KCl 20kg/10a                                | No                           | Yes             | Oct.3        | Sep.10, Typhoon18 caused brook overflow with soil and sand to the test field (south west side) |
| 2016 | Hitomebore  | Fertilizer (18N-15P-15K) 50kg/10a; KCl 20kg/10a                                | N K fertilizer (17N-17K) 2~3kg/10a | Yes             | Oct.1        |                                                                      |
| 2017 | Hitomebore  | Fertilizer (17N-15P-14K) 50kg/10a; Ca Silicate 50kg/10a as soil improver just before addition of fertilizer | N K fertilizer (17N-17K) 2~3kg/10a | Yes             | Oct.9        |                                                                      |
| 2018 | Koshihikari | Fertilizer (17N-15P-15K) 50kg/10a; KCl 20kg/10a                              | N K fertilizer (17N-17K) 4~5kg/10a | Yes             | Sep.29       |                                                                      |
| 2019 | Koshihikari | Fertilizer (17N-15P-15K) 50kg/10a; KCl 20kg/10a                              | N K fertilizer (17N-17K) 4~5kg/10a | Yes             | Oct.5        | Oct.11-12, Typhoon19 caused brook overflow with soil and sand to the test field (south west side) |
2.2 Measurement of radiocesium and exchangeable cations

The $^{137}$Cs in brown rice and straw were measured by a Ge semiconductor detector (GEM and GMX type; Seiko EG&G) for 1 h to 24 h in 100 mL or 250 mL containers. The $^{137}$Cs value above each detection level of $2\sigma$ was adopted instead of $3\sigma$. The water content of brown rice and straw samples were around 10%. The values were adjusted as 10% water content. Further to calculate TF, the values are adjusted to the date of measurement of the soil, using a half-life of 30.08 year. The soil samples were measured by a NaI (TI) scintillation counter (2480WIZARD2Auto-counter: Perkin Elmer) in 20 mL vials within a week after the sampling date. The value of the soil was corrected per dry weight by measuring the soil weight after drying the soil at 60°C for more than 6 days. The exchangeable cations in the soil extracts were analyzed using ICP-OES (Optima 7300DV). The dry soil was crushed and sieved with 2 mm sieve, and 4 g of the sieved dry soil was mixed with 40 mL of 1M ammonium acetate solution at room temperature for more than 1 h and the supernatant obtained by centrifugation was used for the analysis. For the analysis of exchangeable $^{137}$Cs, 10 g of the sieved dry soil and 100 mL of ammonium acetate was used, and the whole 0.2 µm filtrate (co.100 mL) was used for the measurement by a Ge semiconductor detector. The data was divided by $^{137}$Cs value of 10 g of the sieved dry soil measured by the NaI scintillation counter and adjusted to the Ge measurement date, using the half-life. For the measurement, the dry soil was prepared at 60°C for 7 days and stored at room temperature (15–25°C) for 15 months in case of 2017 soil and for 3 months in case of 2018 and 2019 soils. The 2017 soil and 2018 soil were measured simultaneously.

3. Results and discussion

3.1 Yearly change of $^{137}$Cs in brown rice, straw and paddy soil

Fig. 2 shows the yearly change of $^{137}$Cs in brown rice (A) and straw (B) from 2015 to 2019 at each sampling point. Though some data of brown rice below the detection level were not presented in Fig. 2 (A) and one missing data in Fig. 2 (B), annual

![Graph showing the yearly change of $^{137}$Cs concentration in brown rice and straw from 2015 to 2019.](image-url)
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Decrease of $^{137}$Cs in brown rice and in straw are obvious, though variance is observed especially in straw (B). The variance may be due to possible contamination with dirt.

Fig. 3 shows yearly results of the $^{137}$Cs concentration of paddy soil at each sampling point ($M_1$, $M_2$, $M_3$, $M_4$ and $M_5$) from 2015 to 2019 as shown in Fig. 1. Among the points, $^{137}$Cs concentrations of $M_2$ and $M_5$ are about 1,100 Bq/kg, those of $M_3$ and $M_4$ are about 2,200 Bq/kg, and that of $M_1$ is about 1,700 Bq/kg, keeping the gradient of $^{137}$Cs concentration at the decontamination time in 2012. The yearly change of the $^{137}$Cs concentration from 2015 to 2019 is not obvious within experimental errors at each point. This means relatively slow soil $^{137}$Cs movement from 2015 to 2019. Theoretically $^{137}$Cs (half-life: 30.08 year) should reduce 9% during 4 years. However, the 9% reduction was not obvious because of experimental variance and due to probable introduction of small quantity of $^{137}$Cs from outer contaminated fields with water and wind.

3.2 Yearly change of transfer factors of $^{137}$Cs to brown rice and straw from soil

Fig. 4 shows the yearly change of TF of $^{137}$Cs from soil to brown rice (A) and to straw (B), which can be calculated from Figs. 2 and 3. These show more than 80% decreases in the TF, those are, brown rice: 0.0022 ± 0.0008 in 2015 to 0.0003 ± 0.0001 in 2019 and straw: 0.0262 ± 0.0102 in 2015 to 0.0028 ± 0.0009 in 2019. Fig. 4C shows semi-log plots of all the data of TF of $^{137}$Cs to year axis and regression lines and formulas for brown rice and straw are included. Semi-log plots show better fitting than usual plots (A and B). Regression analyses of the plots show that $R^2$ values are 0.81 for brown rice ($N=22$) and year, and 0.76 for straw ($N=24$) and year, which means strong correlations between the TF and year (ageing), and the correlations are significant ($P$-value: 7.56E-09 for brown rice and 2.62E-08 for straw). From the regression formulas, annual decreasing rates of TF are calculated to be 39% (95% confidence range: 32–45%) for brown rice and 43% (95% confidence range: 34–50%) for straw, respectively. These values are close to 30–40% decrease reported by Fujimura et al. employing pot experiments from 2011 to 2012, though cultivation years are earlier. Our early results in 2012 and 2013 at the same Sasu field also showed about 30% decrease in the TF of brown rice, compared in the KCl-fertilized fields. The data of 2012 to 2014 were not included in this note, since the fields were
divided to fields with KCl and without KCl and the sampling points were different from those of 2015 to 2019.

Fig. 5 shows results of measurement of exchangeable positive ions (Na, K, Mg and Ca) of the sampled soil. In 2015, all the ion concentrations were low, compared to 2014 and 2016–2019, this may be due to outflow of surface fine soils and sedimentation of fines flowed in with the flood-water by typhoon 18 on September 10 before the soil sampling. The ex-

![Diagram of exchangeable cation concentrations of the soil (2014–2019). Each point is the average with a range of SD. N=5 except 3 in 2015. The values are expressed as mg of Na, K2O, MgO and CaO per 100 g of dry soil (vertical axis) (Color online).](image)

![Diagram of the yearly change of transfer factor of 137Cs from soil to brown rice (A) and straw (B). Column is the average of the data obtained of the 5 sampling points and bar is the SD. C: Semi-log plots of all data obtained of transfer factors of 137Cs from soil to brown rice (blue dots: below) and to straw (red dots: upper) to year axis. Regression lines and formulas with N (number of data analyzed) are shown in Fig. 4C (Color online).](image)
changeable K ion increased from 14.2±1.8 mg/100 g of soil (2015) to 22.8±2.0 mg/100 g of soil (2019) as K₂O. Since the increase of exchangeable K ion reduces radiocesium absorption by plant from soil, the increase could explain the decrease of the TF from 2015 to 2019, and the above single regression analyses of TF with year as a single variable may overestimate the decreasing rates by year. Further possibly strong relationship between year and K₂O may cause multicollinearity and multiple regression analyses may be hampered, then Variance Inflation Factor (VIF) between year and K₂O of the data sets was calculated to be 2.23 for brown rice and 2.42 for straw, respectively, and the multicollinearity is not serious for the analyses. Multiple regression analyses of all sets of data of logarithm of TF in 2015 to 2019 to year and K₂O as variables were performed and show that R² values are 0.81 for brown rice (N=20) and 0.77 for straw (N=22) with year and K₂O, and significant P-values for year of 0.0006 for brown rice and of 0.0002 for straw, respectively, assuming significant P level of 0.05. From the following multi-regression formulas:

Log (TF of brown rice) = 309.61-0.1548 Year-0.02103 K₂O (mg/100 g of soil),

Log (TF of straw) = 456.65-0.2274 Year-0.01135 K₂O (mg/100 g of soil),

annual decreasing rates of TF are calculated to be 30% for brown rice (95%confidence range:16–41%) and 41% for straw (95%confidence range:24–53%). These mean that the decreases in TF from 2015 to 2019 are mainly due to year (ageing) effect, and that the K₂O annual increase (2.15 mg as average) may add the decrease by 10% for brown rice and 5% for straw as a supplemental manner, the values of decrease are comparable with 8% decrease for brown rice and 9% for straw, calculated by the K₂O increase with the formulas of the relation between TF and exchangeable K₂O concentration obtained at Sasu and Komiya in 2016, reported in preliminary form (http://fukushima-saisei.jp/app-def/s-102/madei/wp-content/uploads/2017/10/20171022_poster21.pdf). These suggest that 137Cs in soil was gradually transformed to a form more difficult to be absorbed by rice, and we assessed possible annual change of exchangeable 137Cs fraction in the soil.

3.3 Analysis of exchangeable 137Cs fraction in the soil sampled in 2017, 2018 and 2019

Fig. 6A shows the yearly change of the fraction of exchangeable 137Cs to whole 137Cs in the soil sampled in 2017, 2018 and 2019 at each sampling point. At all the points, the exchangeable 137Cs fraction decreased from 2017 to 2019. A significant difference (P=0.011, t-test) between the fractions of 2017 and 2018, and a significant difference (P=0.016, t-test) between the fractions of 2018 and 2019. Fig. 6B shows semi-log plots of all data (N=15) of exchangeable 137Cs fraction (%) to year axis. R² value of 0.55 with significant P-value of 0.002. Employing the regression formula:

Log (Exchangeable 137Cs fraction: %) = 160.22-0.079x(year), annual decrease rate of exchangeable 137Cs fraction is calculated to be 17% (95% confidence range: 8–24%). The exchangeable K₂O increased from 2017 to 2019 as shown in Fig. 5, and the increase may cause the decrease of exchangeable 137Cs fraction. Further possibly strong relationship between year and K₂O in the data may cause multicollinearity and multiple regression analysis may be hampered, then VIF between year and K₂O of the data sets was calculated to be 1.81 and the multicollinearity is judged not serious for the analysis. Multiple regression analysis of all sets of data of logarithm of exchangeable 137Cs fraction in 2017 to 2019 to year and exchangeable K₂O (mg/100 g of soil) as variables were performed and show that R² value is 0.59 (N=15) and significant P-value for
year of 0.04, but non-significant $P$-value for K$_2$O of 0.296. The regression formula is:

$$\log (\text{Exchangeable}^{137}\text{Cs fraction: } \%) = 121.15 - 0.0596 \times \text{year} - 0.0062 \times \text{K}_2\text{O}$$

This consecutive field work from 2015 to 2019 indicates that $^{137}$Cs in the soil was gradually transformed to a form more difficult to be absorbed by rice, that is considered due to gradual radiocesium fixation with ageing, as shown by Roig et al., Abusalom et al., Yamaguchi et al., and Takeda et al. who investigated fixation mechanism and conditions in details mainly under artificial conditions. However, their data are from those within 3 years from radiocesium fall-out or addition to soils. Our work showed that phytoavailability of $^{137}$Cs assessed by TF of rice and exchangeable $^{137}$Cs fraction of the soil continue to decrease even after 4 to 8 years from the fall out in an actual paddy field in Iitate Village in Fukushima Prefecture. This slow decrease may be partly due to the type of soil of the allophanic Andosol of the test field, lower ageing speed, compared to mineral soil, and also due to probable presence of $^{137}$Cs in organic materials not exchangeable (co. 6% in case of Date-city soil) in soil and $^{137}$Cs in harvest residue returned which may
gradually be transformed exchangeable and further fixed to mineral content in the soil. The mechanism remains to be clarified.

4. Conclusion

Just after the disaster of FDNPP, we decontaminated the paddy field by removing the contaminated surface soil by mixing the surface (0–5 cm) and flow-out. During the period between 2012 and 2014, we reconfirmed the importance of KCl addition to reduce radiocesium concentration of rice, and could clear the safety standard for food in Japan (<100 Bq/kg). Natural reduction of radioactivity with decay of $^{134}$Cs (half-life of 2 y) helped the reduction by 75% for 4 years by 2015. However, we cannot further expect such yearly natural reduction of radioactivity with decay of $^{137}$Cs (half-life of 30 y). Nevertheless, consecutive yearly reduction of radiocesium activity in harvested rice was observed (2015 to 2019) at a KCl-fertilized and straw plowed-in paddy field and this is suggested mainly due to gradual radiocesium fixation by soil with ageing. This ageing effect should help further reduction of radiocesium contamination in rice and other crops by year. Further this field work also shows the robustness of the rice cultivation that we could harvest rice far below the safety standard, even though the paddy field suffered a flood by a typhoon in September 2015.

We wish to extend rice cultivation trials to other paddy fields and sooner resurrection of agriculture in Fukushima.

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