Chapter 3
Annual Reduction of Transfer Factors of Radiocesium from Soil to Rice Cultivated in a KCl Fertilized and Straw Plowed-in Paddy Field from 2015 to 2021

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3.1 Introduction

Rice is the most important crop in Japan. The Tokyo Electric Power Company's Fukushima Daiichi Nuclear Power Plant (TEPCO-FDNPP) accident in March 2011 caused radioactive materials to spread out and down onto farmlands around the Plant. In the Iitate village more than 30 km far from the Plant in Fukushima Prefecture, people were evacuated from June 2011 to March 2017, except Nagadoro Ward that still needs to be 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 again, without fear of radioactive contamination. The early year results (2012–2013) (Ii et al. 2015; Ii and Tanoi 2016) 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 K_2O). 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 with less radiocesium contamination and farmers usually recycle straw for the following year rice plantation. Then we continued to cultivate rice at KCl fertilized and straw plowed-in paddy field and monitor the radiocesium concentration of brown rice and straw harvested. Similar results of importance of the exchangeable K content were reported by Fujimura and Eguchi (2016) at five paddy fields in

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Fukushima Prefecture in 2012–2014. Further Yamamura et al. (2018) 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. (2007) tested the conditions of long-term radiocesium trapping (ageing) by clay soil from the Chernobyl area, and Yamashita et al. (2016) reported yearly decrease in the TF of radiocesium from soil to grasses from 2012 to 2015 on a pasture in Iwate Prefecture. Tsukada (2014) 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 and Eguchi (2016) showed 40% and 30% decrease in TF in brown rice and inedible rice part from 2011 to 2012, by employing pot experiments using soil samples from Fukushima, suggesting an irreversible sorption of ^{137}Cs to clay minerals. Tagami et al. (2018) also showed the geometric mean of ^{137}Cs TF of brown rice to soil from paddy fields without additional K decreased from 0.012 in 2011 to 0.0035 in 2013. Wakabayashi et al. (2020) reported ^{137}Cs ageing assessed by the ratios of exchangeable ^{137}Cs/^{133}Cs 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 ^{137}Cs from soil to brown rice and straw from 2015 to 2019, based on Ii et al. (2021) and to bottom levels in 2019 to 2021. The decrease from 2015 to 2019 is suggested to be mainly due to ageing effect.

3.2 Materials and Methods

3.2.1 Test Field, Rice Cultivation, and Sampling

Rice cultivation is performed at the same field at Sasu test field (N37°44′, E140°43′) as reported (Ii et al. 2015, 2021; Ii and Tanoi 2016) (Fig. 3.1).

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 (Ii et al. 2015, 2021; Ii and Tanoi 2016). The soil radiocesium concentration was 2000–6000 Bq/kg of dry weight. Further decontamination was not performed thereafter. In 2012 and 2013, the straw harvested was not plowed in, for fear of possible radiocesium contamination. In 2014 and after, all the straw harvested was plowed in the same paddy field as fertilizer for the following cultivation. Rice seedlings of Akitakomachi (2012–2013), Hitomebore (2014–2017, 2020–2021), Koshihikari (2018–2019) were planted in the fields. Rice radiocesium absorption is not significantly different among these cultivars (Ono et al. 2013). Table 3.1 summarizes rice cultivation procedures including fertilizers used from 2014 to 2021.

Rice planting was performed in May or early June. In 2014, a basal fertilizer (18N–15P–15K; weight % as N, P_2O_5, and K_2O, 50 kg/10a) was employed and K

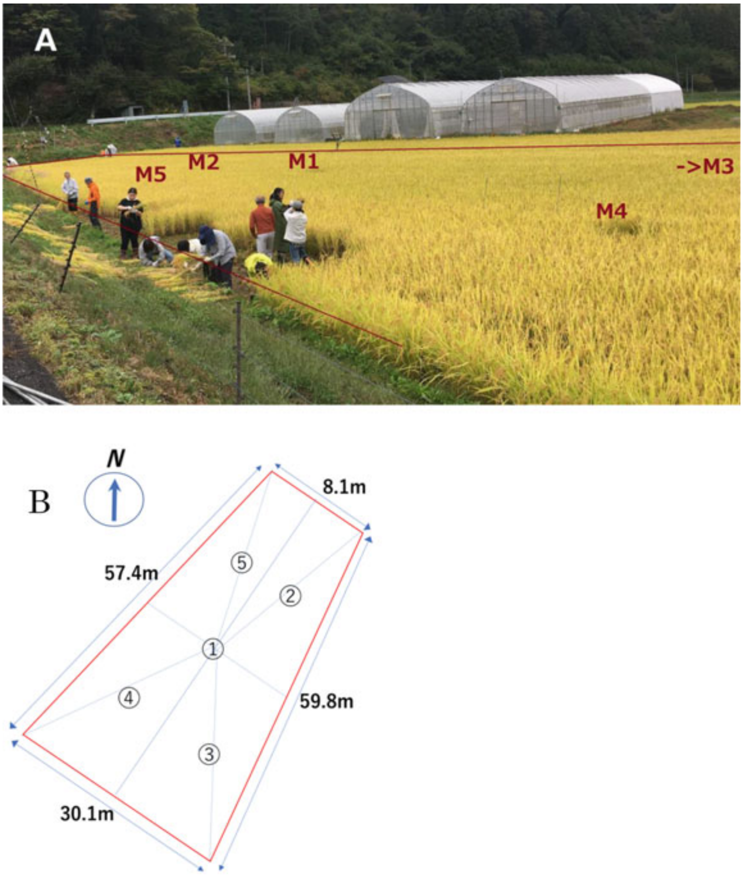

Fig. 3.1 The test field of Sasu and the 5 sampling points (**a**) the picture of rice harvest with hand, in October 6 of 2019, the next day of the sampling (sampling points M1–5 shown). (**b**) Diagram of the sampling points (1–5). [Figures from Ii et al. (2021)]

fertilizer was added as KCl (60% as K_2O:20 kg/10a) 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/10a) and KCl (20 kg/10a) were added to whole field. In 2016, the same as in 2015, except an additional fertilizer of NK (17N:17K) 2–3 kg/10a was added in July. In 2017, the same as in 2016, except the addition of Ca silicate (50 kg/10a) as a soil improver and a basal fertilizer (20N–15P–14K, 50 kg/10a) 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 4–5 kg/10a in July. In 2020 and 2021, the same as in 2019 except the amount of additional NK fertilizer of 2–3 kg/10a. In 2021, Koshihikari was also planted in a small part just for a comparison purpose.

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

Table 3.1 Rice cultivation procedures and sampling date (2014–2021)

Year	Species	Basal fertilizer	Additional fertilizer	Straw plowed in	Sampling date	Note
2014	**Hitomebore**	Fertilizer or Hitomebore (18–15–15) 50 kg/10a; KCl 20 kg/10a (KCl addition half area only)	No	Yes	Oct. 5	
2015	**Hitomebore**	Fertilizer or Hitomebore (18–15–15) 50 kg/10a; KCl 20 kg/10a	No	Yes	Oct. 3	Sep. 10, Typhoon 18 caused brook overflow with soil and sand to the test field
2016	**Hitomebore**	Fertilizer for Hitomebore (18–15–15) 50 kg/10a; KCl 20 kg/10a	NK fertilizer (17–0–17) 2–3 kg/10a	Yes	Oct. 1	
2017	**Hitomebore**	Fertilizer for Hitomebore (20–15–14) 50 kg/10a; KCl 20 kg/10a (Ca Silicate 50 kg/10a as soil improver just before addition of fertilizer)	NK fertilizer (17–0–17) 2–3 kg/10a	Yes	Oct. 9	
2018	**Koshihikari**	Fertilizer for Hitomebore (20–15–14) 50 kg/10a; KCl 20 kg/10a	NK fertilizer (17–0–17) 4–5 kg/10a	Yes	Sep. 29	
2019	**Koshihikari**	Fertilizer for Hitomebore (20–15–14) 50 kg/10a; KCl 20 kg/10a	NK fertilizer (17–0–17) 4–5 kg/10a	Yes	Oct. 5	Oct. 11–12, Typhoon 19 caused brook overflow with soil and sand to the test field
2020	**Hitomebore**	Fertilizer for Hitomebore (20–15–14) 50 kg/10a; KCl 20 kg/10a	NK fertilizer (17–0–17) 2–3 kg/10a	Yes	Oct. 2	

(continued)

Table 3.1 (continued)

Year	Species	Basal fertilizer	Additional fertilizer	Straw plowed in	Sampling date	Note
2021	**Hitomebore** **Koshihikari**	Fertilizer for Hitomebore (20–15–14) 50 kg/10a; KCl 20 kg/10a	NK fertilizer (17–0–17) 2–3 kg/10a	Yes	Oct. 2	

Data for 2014 to 2019 from Ii et al. (2021)

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 2015–2021, soil and rice sampling were 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. 3.1. 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 threshed with a foot-driven thresher to give one unhulled rice sample for each test point at Sasu (Fig. 3.2).

The soil sample and the unhulled rice sample were sent to “Circle Madei” (Ii and Tanoi 2016), a volunteer employee and student group at Tokyo University and three aliquots of co. 20 mL of soil in vials were prepared from each soil in a bag for the radioisotope measurement (Fig. 3.3). The unhulled rice sample was kept for a week or more in the room, and then brown rice sample was prepared with a hulling machine. The straw sample was dried naturally in a greenhouse for a week or more. Then the straw sample for radioisotope measurement was prepared by cutting the straw part by 1 cm, sent to “Circle Madei,” and further dried in the room for a week or more.

3.2.2 *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 1–24 h in 100 mL or 250 mL containers. The ^{137}Cs value above each detection level of 2σ was adopted. 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 (Tl) scintillation counter (2480WIZARD2Autoɣcounter:Perkin Elmer) in 20 mL vials (Nobori et al. 2013) 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) (Ii et al. 2015). The dry

Fig. 3.2 Photos of the sampling of rice and soil (left), and the rice bundles were threshed with a foot driven thresher (right)

Fig. 3.3 Photo of recently renewed “Circle Madei” room, the members preparing soil samples for measurement of radiocesium

soil was crushed and sieved with 2 mm sieve, and 4 or 10 g of the sieved dry soil was mixed with 10 vol. of 1 M ammonium acetate solution at room temperature for more than 1 h and the supernatant was obtained by centrifugation and the filtrate with 0.2 μm filter 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 80 mL of

0.2 μm filtrate was used for the measurement by a Ge semiconductor detector. The data was divided by ^{137}Cs value of 8 g of the 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. To compare the soils in 2019, 2020 and 2021, 2019 soil stored for 15 months, 2020 soil for 3 months and 2021 soil was stored for 5 months. The 2019 and 2020 soils were measured simultaneously.

3.3 Results and Discussion

3.3.1 *Yearly Change of ^{137}Cs in Brown Rice, Straw and Paddy Soil*

Figures 3.4 and 3.5 show the yearly change of ^{137}Cs in brown rice (a) and straw (b) from 2015 to 2021 at each sampling point. Though some data of brown rice below the detection level were not presented in Fig. 3.4a and one missing data in Fig. 3.5b, annual decrease of ^{137}Cs in brown rice and in straw are obvious, though variance is observed. The variance may be due to possible contamination with dirt and sampling variance.

Figure 3.6 shows yearly results of the ^{137}Cs concentration of paddy soil at each sampling point (M①, M②, M③, M④ and M⑤) from 2015 to 2021 as shown in

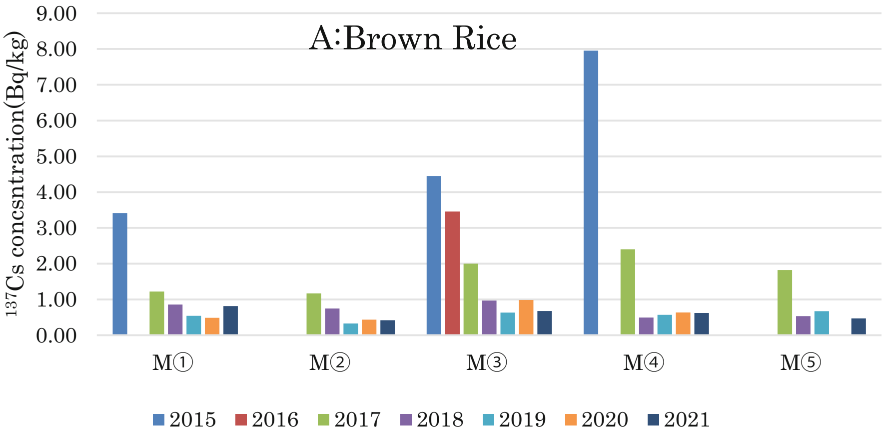

Fig. 3.4 The yearly change of ^{137}Cs in brown rice (**a**) at the sampling points (2015–2021). The values are adjusted to the date of measurement of the soil within a week after the rice sampling. Data are not presented for M2 in 2016, M5 in 2015, 2016, and 2020, because they were below the detection level (2σ). [Figure modified from Ii et al. (2021) including the new data of 2020 and 2021]

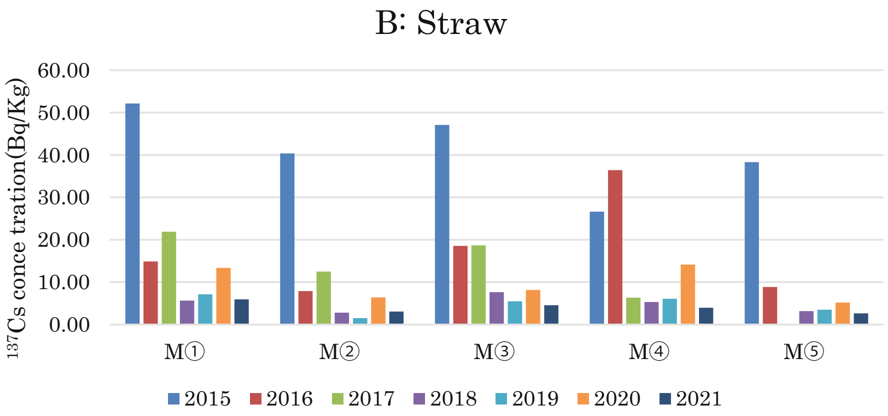

Fig. 3.5 The yearly change of ^{137}Cs in straw (**b**) at the sampling points (2015–2021). The values are adjusted to the date of measurement of the soil within a week after the rice sampling. Data are not presented for M⑤ in 2017, because the spectrum peak shape did not show Gaussian distribution. [Figure modified from Ii et al. (2021) including the new data of 2020 and 2021]

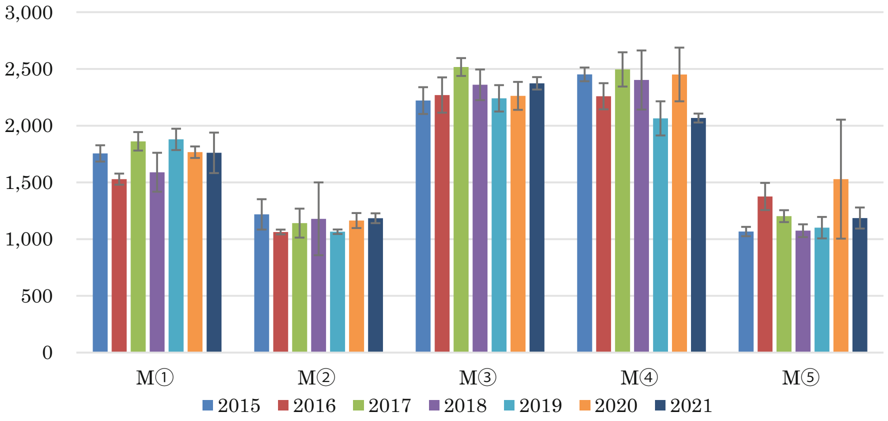

Fig. 3.6 ^{137}Cs concentration (Bq/kg of dry soil) of paddy soil at each sampling point (M①, M②, M③, M④ and M⑤) from 2015 to 2021. [Figure modified from Ii et al. (2021) including the new data of 2020 and 2021]

Fig. 3.1. Those of M③ and M④ are about 2200 Bq/kg, and that of M① is about 1700 Bq/kg, keeping the gradient of ^{137}Cs concentration at the decontamination time in 2012$^{1),2)}$. The yearly change of the ^{137}Cs concentration from 2015 to 2021 is not obvious at each point. This means relatively slow soil ^{137}Cs movement from 2015 to 2021. Theoretically ^{137}Cs (half-life: 30.08 year) should reduce 13% during 6 years. However, the 13% reduction was not obvious because of experimental variance and due to probable introduction of some quantity of ^{137}Cs from outer contaminated fields with water and dust.

3.3.2 Yearly Change of Transfer Factors of ^{137}Cs to Brown Rice and Straw from Soil

Figures 3.7 and 3.8 show the yearly change of TF of ^{137}Cs from soil to brown rice (a) and to straw (b), respectively, which can be calculated from Figs. 3.4, 3.5 and 3.6. 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. Little decrease was observed from 2019 to 2021.

Figure 3.9 shows semilog plots of all the data of TF of ^{137}Cs to year axis (c). Regression lines and formulas for brown rice and straw are included. Semilog plots show better fitting than usual plots (a and b). Regression analyses of the plots show that R^2 values are 0.72 for brown rice ($N = 31$) and year, and 0.60 for straw ($N = 34$) and year, which still means strong correlations between the TF and year (ageing), and the correlations are significant (P value: 1.58E−09 for brown rice and 9.15E−08 for straw). From the regression formulas, annual decreasing rates of TF are calculated to be 28% (95% confidence range: 22–33%) for brown rice and 28% (95% confidence range: 21–35%) for straw, respectively. These values are lower than the values of 39% for brown rice and 43% for straw calculated from the data of 2015 to 2019, but close to 30–40% decrease was reported by Fujimura and Eguchi (2016) employing pot experiments from 2011 to 2012, though cultivation years are much earlier. Our early results (Ii et al. 2015; Ii and Tanoi 2016) 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 2012–2014 were not included in this report, since the fields were divided into fields with KCl and without KCl and the sampling points were different from those of 2015 to 2021.

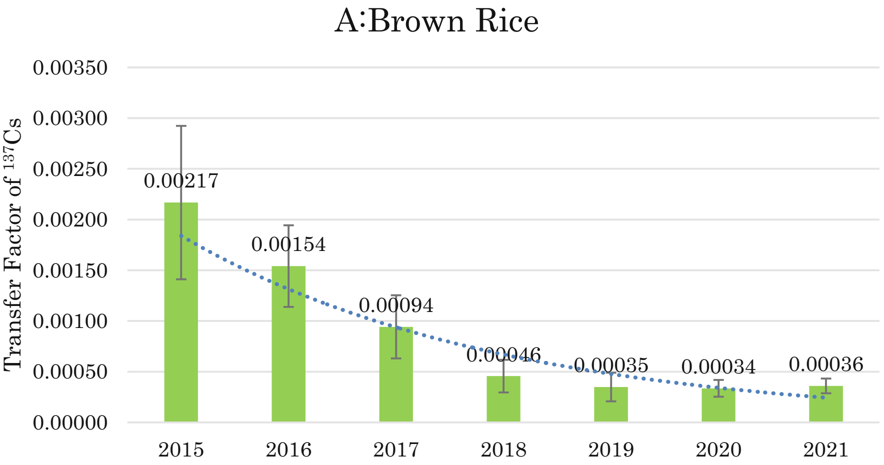

Fig. 3.7 The yearly decrease in transfer factor of ^{137}Cs from soil to brown rice (**a**) [Figure modified from Ii et al. (2021) including the new data of 2020 and 2021]

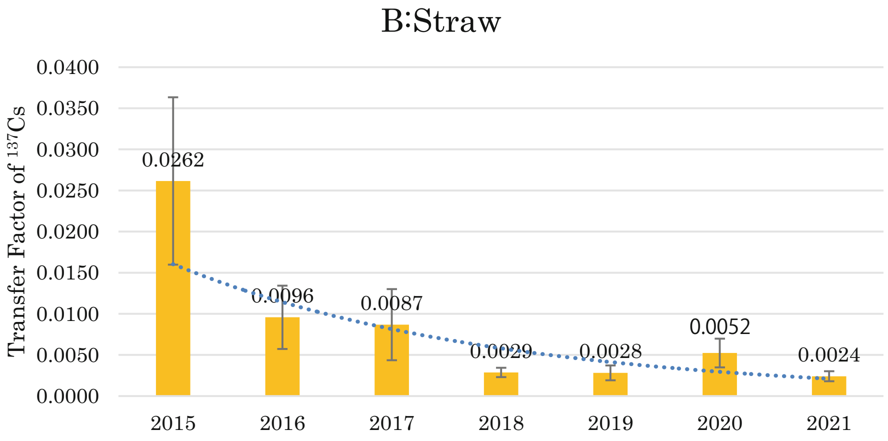

Fig. 3.8 The yearly decrease in transfer factor of ^{137}Cs from soil to straw (**b**) Figure modified from Ii et al. (2021) including new data of 2020 and 2021. Straw values (red number) in 2020 are high, probably due to sampling of more leaf part. [Figure modified from Ii et al. (2021) including the new data of 2020 and 2021]

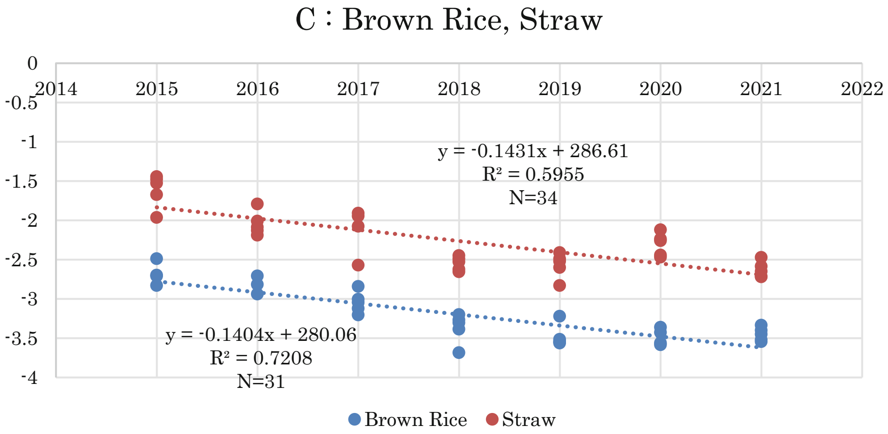

Fig. 3.9 Semilog plots of all data obtained of transfer factors of ^{137}Cs from soil to brown rice (blue dots: below) and to straw (red dots: upper) to year axis (**c**). Regression lines and formulas with *N* (number of data analyzed). [Figure modified from Ii et al. (2021) including new data of 2020 and 2021]

Figure 3.10 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 floodwater by typhoon 18 on September 10 before the soil sampling. The exchangeable 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 K_2O, and

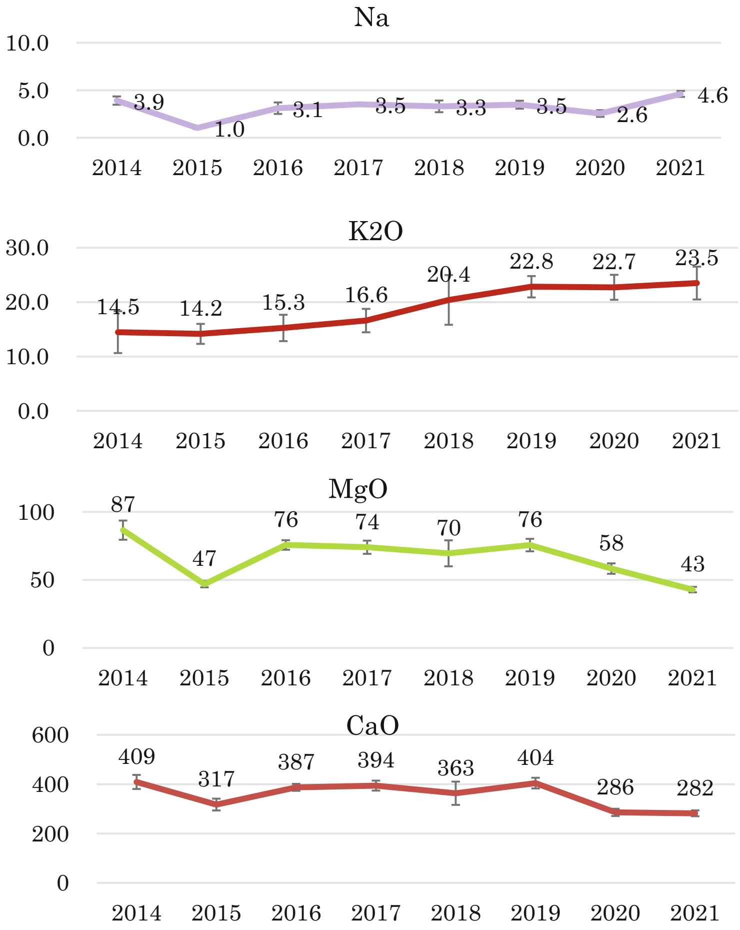

Fig. 3.10 The exchangeable cation concentrations of the soil (2014–2021). Each point is the average with a range of SD. $N = 5$ except 3 in 2015. The values are expressed as mg of Na, K_2O, MgO and CaO per 100 g of dry soil (vertical axis). [Figure modified from Ii et al. (2021) including new data of 2020 and 2021]

little change from 2019 to 2021. 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 K_2O may cause multicollinearity and multiple regression analyses may be hampered, then Variance Inflation Factor (VIF) between year and K_2O of the data sets from 2015 to 2021 was calculated to be 3.76 for brown rice ($N = 29$) and 2.54 for straw ($N = 32$), respectively, and the multicollinearity is not serious for the analyses. Multiple regression analyses of all sets of data of logarithm of TF in 2015–2021 to year and K_2O as variables were performed and

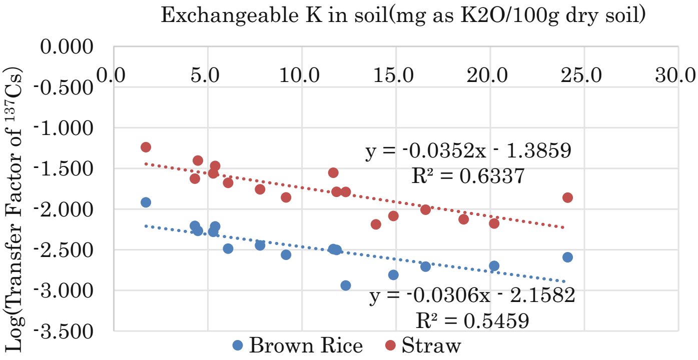

Fig. 3.11 Relation between TF and exchangeable K_2O concentration obtained at Sasu and Komiya paddy fields in Iitate Village in 2016. Straw (red points: above). Brown rice (blue points: below)

show that R^2 values are 0.73 for brown rice ($N = 29$) and 0.61 for straw ($N = 32$) with year and K_2O, and significant P values for year of 0.0002 for brown rice and of 0.0014 for straw, and significant P values for K_2O of 0.016 for brown rice but nonsignificant P value of 0.079 for straw, respectively, assuming significant P level of 0.05. From the following multiregression formulas:

Log (TF of brown rice) = 161.704 – 0.08147 Year – 0.02694 K_2O (mg/100 g of soil),

Log (TF of straw) = 183.652 – 0.09186 Year – 0.02856 K_2O (mg/100 g of soil),

annual decreasing rates of TF are calculated to be 17% for brown rice (95% confidence range, 7–26%) and 19% for straw (95% confidence range: 4–32%). These mean that the decreases in TF from 2015 to 2021 are mainly due to year (ageing) effect, but that the K_2O annual increase (1.55 mg as average) may add the decrease by 9% for brown rice and 10% 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 K_2O increase with the formulas of the relation between TF and exchangeable K_2O concentration obtained at Sasu and Komiya in 2016, as shown in Fig. 3.11.

These suggest that ^{137}Cs in soil was gradually transformed to a form more difficult to be absorbed by rice, and we assessed possible annual change of exchangeable ^{137}Cs fraction in the soil.

3.3.3 Analysis of Exchangeable ^{137}Cs Fraction in the Soil Sampled in 2019, 2020 and 2021

In a previous report (Ii et al. 2021), we compared the fraction of exchangeable ^{137}Cs to whole ^{137}Cs in the soil sampled in 2017, 2018, and 2019 at each sampling point. At all the points, the exchangeable ^{137}Cs fraction decreased from 2017 to 2019 and

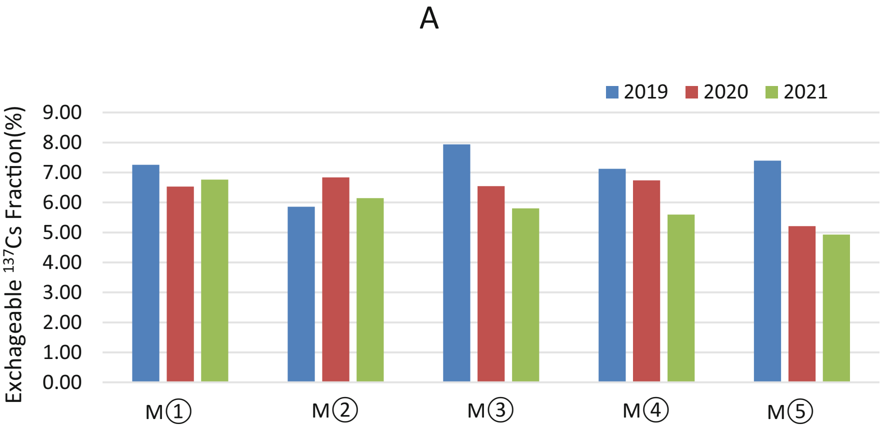

Fig. 3.12 (**a**) Comparison of the fraction (%) of exchangeable ^{137}Cs in the soil at the sampling points (2019–2021)

multiple regression analysis of the set of the data with year and K_2O showed that annual decrease rate of exchangeable fraction is 13% (95% confidence range: 0.4–24%) and annual decrease fraction by the annual increase of K_2O (3.1 mg/100 g of soil) could be 4%. A similar analysis was performed using the soil samples of 2019, 2020, and 2021.

Figure 3.12a shows of exchangeable ^{137}Cs fraction (%) in the soil at the sampling points from 2019 to 2021. Above 90% ^{137}Cs of the paddy soil is not exchangeable, that is, not extracted with 1 M CH_3COONH_4 solution. Figure 3.13 shows the regression analysis of all the data to year axis, a little decrease of co. 0.6% per year observed in exchangeable ^{137}Cs fraction. R^2 value of 0.40 with P value of 0.011. However, almost no change in the TF for brown rice and straw from 2019 to 2021.

The exchangeable K_2O changed little from 2019 to 2021 (23 mg/100 g of dry soil) as shown in Fig. 3.10, and multiple regression analysis was not performed this time. The decrease appeared to reach close to the bottom level.

The annual decrease rate by year of 13% from 2015 to 2019 is much less than those of the TF of about 40%, suggesting other factor(s) not identified concerned. Further, this is less than the annual decrease rate of 34% calculated from data in Tsukada's report (2014) of a paddy field in Date-city in Fukushima during 1.5 years from 2012 to 2013, but more than that of 4–10% calculated from the half-time of 6.6–17.7 year of ^{137}Cs/^{133}Cs in the exchangeable fraction, using the data from 2011 to 2015, as better index of ^{137}Cs fixation by ageing (Wakabayashi et al. 2020).

This consecutive field work from 2015 to 2021 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. (2007), Absalom et al. (1995), Yamaguchi et al. (2016, 2019) and Takeda et al. (2013, 2020) who investigated fixation mechanism and conditions in detail

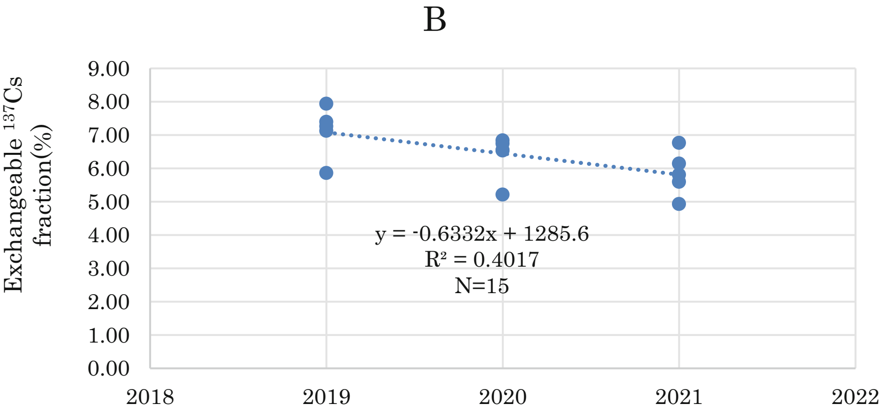

Fig. 3.13 (**b**) Plot and analysis of all the data of exchangeable ^{137}Cs fraction (%) in the soil to year axis in (**a**)

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 4–8 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 (Absalom et al. 1995; Yamaguchi et al. 2019), and also due to probable presence of ^{137}Cs in organic materials not exchangeable in soil (co. 6% in case of Date-city soil, Tsukada 2014) and ^{137}Cs in harvest residue returned and incorporated outer organic matters, which may gradually be transformed exchangeable and further fixed to mineral content in the soil. The mechanism remains to be clarified.

3.4 Conclusive Remark

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 years) helped the reduction by 75% for 4 years by 2015. However, we cannot further expect such natural reduction of radioactivity with decay of ^{137}Cs (half-life of 30 years). Nevertheless, consecutive yearly reduction of radiocesium activity in harvested rice was observed (2015–2019) at a KCl-fertilized and straw plowed-in paddy field and this is suggested mainly due to gradual radiocesium fixation by soil with ageing and further reduction of radiocesium contamination in rice was expected. However, the following study of 2019–2021 suggests that the TF of brown rice and straw may almost reach the bottom values of 0.00035 for brown rice and 0.002 for straw (Figs. 3.7 and 3.8). Further this field work also shows the robustness of the rice cultivation that we could annually harvest rice far below the safety standard, even though the paddy field suffered floods by typhoons in September 2015 and in October 2019.

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

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