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Changes in the soil to brown rice concentration ratio of radiocaesium before and after the Fukushima Daiichi Nuclear Power Plant Accident in 2011

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KEYWORDS: Soil-to-plant transfer; Aging effect; Caesium-137; Paddy rice; Global fallout.

ABSTRACT

Radiocesium (^{R}Cs) mobility in soil is initially relatively high when the nuclide first comes into contact with soil, after which the mobile fraction decreases with time due to ^{R}Cs fixation to soil particles (aging effect). Consequently, the ^{R}Cs activity concentration in plants grown in soil was expected to decrease with time after the Fukushima Daiichi nuclear power plant accident in 2011. In this study, we collated data on concentration ratios (CR) of ^{R}Cs between brown rice grain and paddy soil and compared CR values reported for periods before and after the accident. For this purpose, soil and rice data were collected after the accident specifically from paddy fields which did not have additional potassium fertilizer added (for remediation purposes). The geometric mean rice/soil CR of ^{R}Cs for all types of soil was 1.2×10^{-2} in 2011 (n=62) and by 2013 the value had declined to 3.5×10^{-3} (n=32) which was similar to that for 1995-2007 of 3.4×10^{-3} (n=120). The comparison suggests that the mean soil-to-rice grain concentration ratio had returned to that prevailing before the accident after less than three years. It was also confirmed that CR values for rice sampled from paddy fields were lower than those obtained from pot experiments.

INTRODUCTION

The mobility of radiocaesium (^{R}Cs) in soil is at its highest when the radionuclide initially deposits onto soil; then, with time, the mobile fraction gradually decreases and reaches an equilibrium due to ^{R}Cs fixation to soil particles,[1,2] commonly called an aging effect. Because ^{R}Cs in the mobile fraction in soil solution is taken up through plant roots,[2,3] the ^{R}Cs activity concentration, [^{R}Cs], in plants also decreases with time after addition to soils.[4-8] Accordingly, after the TEPCO's Fukushima Daiichi Nuclear Power Plant (FDNPP) accident occurred on March 11, 2011, it was expected that the mobility of ^{R}Cs released from the accident (reported as ^{134}Cs, ^{137}Cs or $^{134+137}$Cs in this study) in agricultural fields would initially be higher than that observed under equilibrium conditions. In contrast, equilibrium conditions are expected to have been reached for ^{137}Cs from global fallout measured in samples taken many years after its peak deposition period of the late 1950s and 1960s.

In this study, we focused on brown rice because rice is a staple food in Japan and other Asian countries. Brown rice has the husk removed, and white rice is produced by removing ca. 9-10% of the surface layer of the brown rice grain by weight (i.e. bran layer). In Japan, many measurement data of ^{R}Cs are available for soil and brown rice samples collected both before and after the FDNPP accident.[9] The transfer of ^{R}Cs to crops grown in contaminated soil was expected to be high in 2011 compared with that reported previously in IAEA Technical Report Series No. 472[10] for which equilibrium conditions are assumed.

To avoid underestimation of brown rice [^{R}Cs], relatively high transfer was assumed by the authorities for crops in the first year after the FDNPP accident to enable limits to be set for the recommencement of rice cultivation[11]; the ^{R}Cs transfer ratio from soil to brown rice was assumed to be 0.1. Thus, only paddy fields in which the [^{R}Cs] did not exceed 5,000 Bq kg^{-1} dry weight (dw) was allowed to be used for rice cultivation,[11] because the provisional regulation value, 500 Bq kg^{-1} in raw food materials for ^{R}Cs to restrict the distribution of contaminated foods, was used at that time. In 2011, 26,464 brown rice samples were measured in Japan, and only 39 samples (0.15%) exceeded the provisional regulation value.[12] This outcome was expected given the restrictions on use of paddy fields over the conservatively set limit, but the reasons why the 39 samples exceeded the regulation value needed to be identified.

One major potential reason for the relatively high [^{R}Cs] in rice in some paddy soils was a low exchangeable K in the soil solution according to the Ministry of Agriculture, Forestry and Fisheries (MAFF).[13] Many authors have previously shown that if K concentrations in soils (especially exchangeable K concentrations) increases, then ^{R}Cs transfer to crops decreases. [e.g.]

[6,14-16] Most of the relevant data reported are for crops other than rice, but there was one pre-FDNPP accident study which reported this feature for brown rice.[6] Therefore, as a remediation measure, additional K fertilizer (above that normally used) was applied to soil after the FDNPP-accident that was used to grow rice plants[13,17,18] and other crops such as soybeans.[19] Because the soil exchangeable K fraction was comprehensively monitored and controlled in contaminated areas, the [^{R}Cs] in brown rice did not exceed the Japanese standard limit of [^{R}Cs] in crops of 100 Bq kg^{-1} (as of April 1, 2012) in 2015-2017.[20] The number of continuous measurements of soil-to-crop transfer of ^{R}Cs decreased with time,[21-24] with most reports providing data for only one to two years (written in Japanese and English languages). To better understand the time dependency of soil-to-crop transfer of ^{R}Cs, available data were collated from the literature through a comprehensive analysis of the peer-reviewed data. We also identified ^{137}Cs data for brown rice and associated soil samples to obtain estimates of transfer for the period before the FDNPP-accident, and carried out measurements of global fallout ^{137}Cs using archived soil and brown rice sample sets collected in 2006-2007.

The aim of the data collation and new measurements of archived samples was to enable a comparison of the transfer of ^{R}Cs from soil to brown rice for periods before and after the FDNPP accident. Although the annual intake of ^{R}Cs in Japan caused ingestion doses well below 1 mSv a^{-1} since 2011,[9,25] such information is valuable to estimate future intake of ^{R}Cs from rice as well as to prepare for potential nuclear accidents in countries where rice is a staple food.

MATERIALS AND METHODS

Calculation of Soil-to-rice Concentration Ratio (CR). To provide a simple approach for estimating the transfer of radionuclides from soil to crops in radioecological models for dose assessment, the radionuclide activity concentration on dw basis in the crop is compared with that in the ploughed soil layer.[10] In this study, the soil-to-rice concentration ratio (CR) of ^{R}Cs was calculated using the following empirical ratio:

CR= Activity concentration in rice grain (Bq kg^{-1}-dw)/ Activity concentration in soil (Bq kg^{-1}-dw)

The IAEA reported this ratio as a Transfer factor (F_v) which is calculated using the same equation above, and assumes equilibrium has been reached. Some data collated for the F_v values may still have been in a transition stage as they were collected within a few years after the Chernobyl accident. A similar situation would be expected to occur in Japan after the FDNPP accident. The transition stage is defined as the period when the mobility of ^{R}Cs in soil was not in an equilibrium condition due to aging process in physico-chemical forms after the deposition on soil.

For the collated data, if brown rice [^{R}Cs] data were reported in fresh weight, edible state, then the normal water content of 15% was applied to obtain a dw basis for the concentration data.

^{137}Cs CR Data in 2006 and 2007. The National Institute of Radiological Sciences, National Institutes for Quantum and Radiological Science and Technology (QST-NIRS) collected brown rice samples and associated soil samples throughout Japan in 2002-2007. Sample collection sites are shown in Figure 1. The global fallout [^{137}Cs] in the 2002-2005 samples (n=50) have been reported in our previous papers.[26,27] The activity concentration in brown rice and the associated soil samples ranged from 0.005-0.61 Bq kg^{-1}-dw (number of samples determined: 37) and 2.5-

31.1 Bq kg^{-1}-dw (number of samples determined: 50), respectively. Additional ^{137}Cs data were derived by measurements during this study of samples collected in 2006 (n=13) and 2007 (n=5).

The paddy field soils (ploughed soil layer: 0–15 cm) were air-dried and passed through a 2-mm mesh sieve. For brown rice grains, due to the low [^{137}Cs], each 3-5 kg sample was incinerated at 450˚C for 12 h to decrease sample volume.[28]

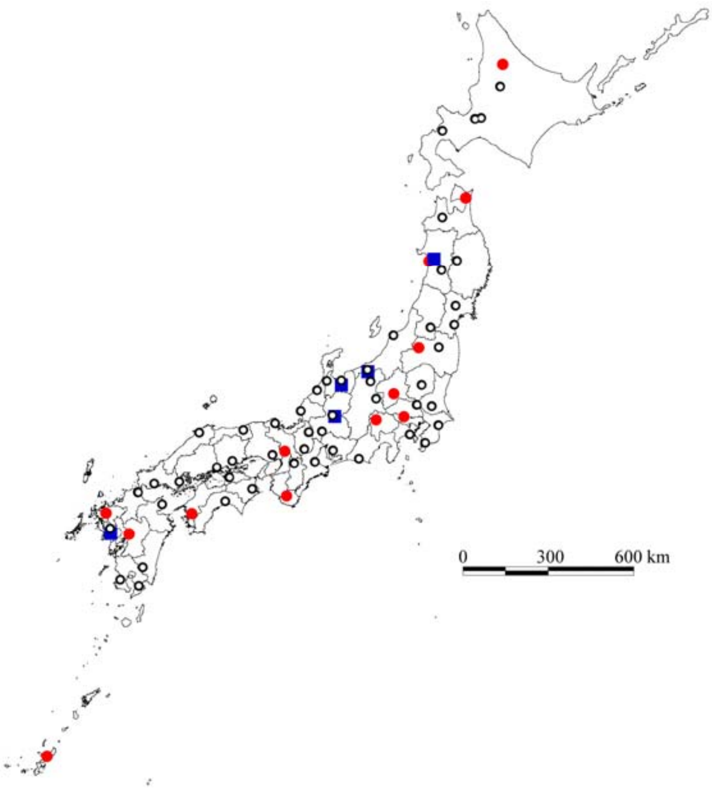

Figure 1. Paddy soil-brown rice sample set collection sites in Japan from 2002-2007. Open circle: collection sites previously reported[25,26], closed circle: collected in 2006, and closed square: collected in 2007.

For ^{137}Cs determination, the volume of each sample was adjusted to 100-mL amount of the air-dried and sieved soil sample or incinerated brown rice sample in a 260-mL plastic vessel

(ASONE, Packclean) with a uniform 33-mm height from the bottom of the vessel. Their [^{137}Cs] was determined with a Ge-detection system (Seiko EG&G Ortec) by counting for 80,000 s for soil samples and 500,000-999,999 s for incinerated brown rice samples. IAEA-156 (clover), IAEA-373 (grass), and IAEA-375 (soil) were used as standard reference materials for measurements.[29]

Data Survey-1: ^{137}Cs Activity Concentrations in Brown Rice in 1965-2016. Most CR values for ^{R}Cs and brown rice after the FDNPP accident were derived for samples from Fukushima and Ibaraki prefectures. To compare the [^{137}Cs] trend in brown rice before and after the FDNPP accident within these two prefectures, and in other areas in Japan, data were selected from the Environmental Radiation Database hosted by Nuclear Regulation Authority, Japan (NRA).[30] In the dataset, rice grain data were available for both brown rice and white rice. To convert white rice data into brown rice, a ^{R}Cs concentration ratio between brown rice/white rice was applied based on measured ratios from our previous studies,[31,32] which ranged from 1.75 to 2.08 with a geometric mean of 2.0 (n=5). The data for ^{137}Cs (both estimated and measured) were classified into two groups of (1) Fukushima and Ibaraki Prefectures, and (2) Other areas in Japan. However, for category (2), we excluded data from Iwate, Miyagi, Tochigi, Gunma and Chiba prefectures where some deposition of the FDNPP releases occurred.

Data Survey-2: CR Data Survey from Literature in 2011-2014. Brown rice and associated soil data were compiled from published papers and institutional reports in Japanese and English. Only data for normal fertilizer, no-K fertilizer (hereafter, No-K), and no fertilizer application conditions were collated for rice paddies and pot experiments. A normal fertilizer application condition is defined as the amount of fertilizer added annually by the farmers before the FDNPP accident including K-fertilizer. Because soil conditions differ in paddy fields, it is difficult to specify the range in the amount of fertilizer added; however, according to the analysis results by MAFF,[33] potassium fertility in soil was almost within the range of appropriate condition for rice production with normal fertilizer application before the FDNPP-accident. There are many papers studying potassium fertilizer effect on ^{137}Cs soil to plant uptake, but few of them were for normal fertilizer conditions. In contrast, all the CR values reported before the FDNPP accident were for normal fertilizer conditions. If fertilizer conditions were not mentioned we assumed that such papers applied normal fertilizer amounts. Rice cultivars were not classified; because it is often not specified in the available Japanese literature. Thus, it was assumed that all data were for the most common Japonica variety of rice because the variety is common in Japan.

Data were accepted under the following criteria.

- If CR values of ^{R}Cs were reported in the publication.
- If [^{R}Cs] data in both brown rice and soil were reported allowing a calculation of CR values.
- data were available in figures only, in which case the [^{R}Cs] were derived from the figures and CR values were calculated.

Statistics. Data obtained in this study were usually distributed log-normally. Therefore, the logarithm of the CR value was chosen for statistical analysis. KaleidaGraph software (Synergy Software, version 4.5.2) was used for *t*-test among two items; thus, multiple *t*-test (*t*-test among all analysis items) was carried out in this study.

RESULTS AND DISCUSSION

^{137}Cs activity concentrations in brown rice since 1965. CR values in the literature after the FDNPP accident was mainly reported for brown rice from the relatively high ^{137}Cs density deposition areas, namely Fukushima and Ibaraki Prefectures. Only a limited number of CR values for brown rice were available before the FDNPP accident in these two prefectures, therefore, comparison could only be carried out with CR values from other areas in Japan. Fortunately, the soil types observed in these two prefectures are commonly occurring soil types throughout Japan,[34] which is classified into Fluvisol according to the FAO/UNESCO classification method.[35]

The levels of [^{137}Cs] in brown rice are shown in Figure 2. Using the conversion stated above, the ^{137}Cs data estimated from white rice agreed well with those of brown rice for both category areas, (1) Fukushima and Ibaraki, and (2) Other areas. The [^{R}Cs] in brown rice from Fukushima and Ibaraki Prefecture before the FDNPP accident were within the range of that of (2) Other Areas data. Therefore, CR data obtained in Fukushima and Ibaraki Prefectures should be comparable of that of Other data in Japan.

Figure 2 shows that the [^{137}Cs] in brown rice collected in Fukushima and Ibaraki Prefectures in 2016 (0.8-1.24 Bq kg^{-1}-raw, n=5) were one order of magnitude higher than that before the FDNPP accident, i.e. geometric mean [^{137}Cs] for all data was 0.076 Bq kg^{-1}-raw in 2006-2010 (n=151). In Fukushima and the surrounding contaminated prefectures, the sampling areas where continuous ^{137}Cs measurement in brown rice had been carried out before the FDNPP accident were not used afterwards so the data collated by NRA are not directly comparable. Therefore, the order of magnitude difference is somewhat imprecise. Overall, the data show a decreasing trend in [^{R}Cs] in brown rice in Area (1).

We could not find openly available continuous soil-to-brown rice data sets from the same sampling field(s) to provide site-specific ^{R}Cs time trends in rice grains over the five-year period. Nevertheless, for herbaceous plants collected from Chiba campus of the QST-NIRS located about 220 km south from the FDNPP, we observed an almost constant [^{R}Cs] from 1000 d after March 11, 2011.[36] Such data suggests that in Area (1) most of the decline occurred in 2011-2013 with less decline in 2013-2014. Since 2015, the total [^{R}Cs] in all brown rice produced in Fukushima Prefecture has not exceeded the standard limit of 100 Bq kg^{-1}.[20] Thus, the aging effect, together with remediation measures (e.g., addition of K fertilizers to keep exchangeable K concentration to a suitable condition in soil, addition of soil amendments to fix ^{R}Cs in soils, or removal of surface layer of agricultural soils to reduce ^{R}Cs), has ensured that the [^{R}Cs] in rice had decreased considerably since deposition.

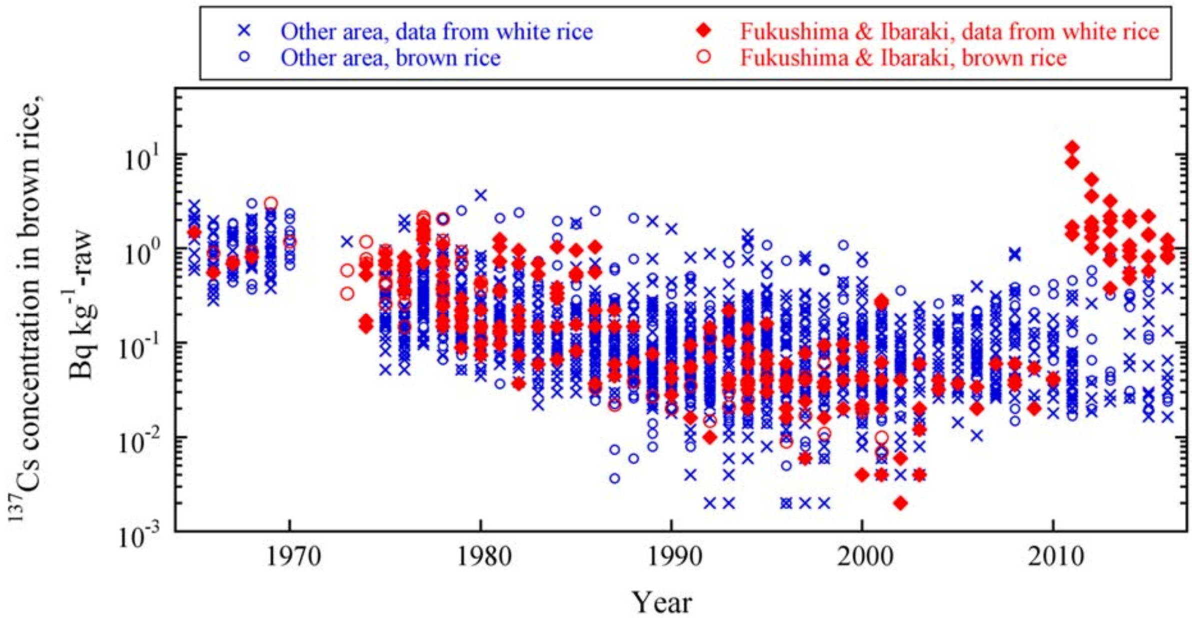

Figure 2. Measured and estimated [^{137}Cs] in brown rice in Japan in 1965-2016. ^{137}Cs in brown rice was estimated using the data in white rice, using a brown rice/white rice ^{R}Cs concentration ratio of 2.0.

Comparison of CR values for ^{R}Cs in brown rice before and after the FDNPP accident. In the NRA database, combined brown rice-soil datasets were difficult to reliably identify. Therefore, other reported data were used including our own data. The National Institute for Agro-Environmental Sciences (NIAES) had been measuring radiocesium in brown rice and associated soil samples continuously in 1959-2000 from all over Japan (approximately 15 stations each year).[37] The resulting brown rice and associated soil data were used to calculate CR values from 1995-2000. We also used our previously reported values and the data we obtained in this study for 2002-2007 (QST-NIRS): the geometric mean value of CR was 2.9×10^{-3}. In some incinerated brown rice samples, ^{137}Cs was not detectable due to the low levels originating from global fallout; nevertheless it was detectable in 37 data of 50 brown rice samples from 2002-2005, 7 data of 13 brown rice samples from 2006 (0.0086-0.094 Bq kg^{-1}-dw) and all 5 samples from 2007 (0.027-0.41 Bq kg^{-1}-dw). For comparison, [^{137}Cs] in soil samples in 2006-2007 were 3.2 - 32.1 Bq kg^{-1}-dw. If we applied the detection limit value of [^{137}Cs] 0.005 Bq kg^{-1}-dw to undetected data in 2002-2007, the geometric mean value of CR was 1.9×10^{-3}. From this result, the estimated CR values using detected [^{137}Cs] data are likely a bit higher than those that would have been derived if all samples were above detection limits.

The literature survey data for CR values of ^{R}Cs for brown rice after the FDNPP accident collected in open paddies with normal fertilizer conditions are shown in Figure 3 together with the data for before the FDNPP accident from NIAES[34] and QST-NIRS. Data in each sampling year were close to log-normal distributions. The yellow band shows the 95% range of CR values in 1995-2007 (4.6×10^{-4} - 2.5×10^{-2}), the geometric mean value was 3.4×10^{-3} in the same year range.

The CR value was clearly higher in 2011 in Fukushima and Ibaraki prefectures. Because rice planting in open paddy fields starts around early May, and by that time in 2011 the amounts of newly deposited ^{R}Cs was smaller than those in March and April, the direct deposition effect to rice grains (typical flowering time is in July-August) was considered to be negligible.

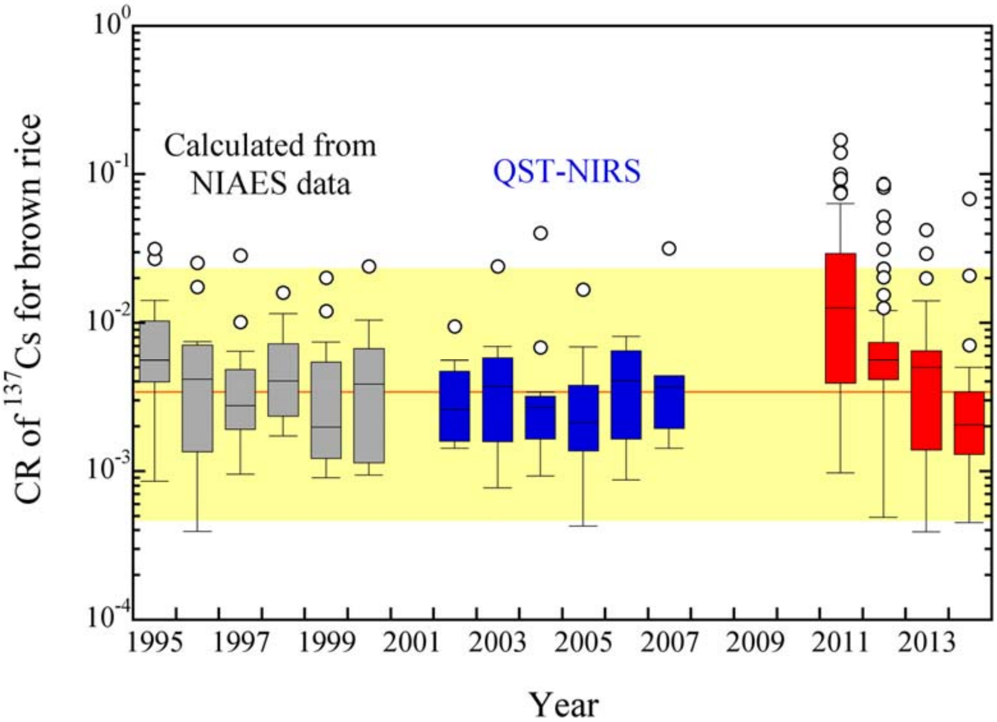

Figure 3. Box-plots of the concentration ratio (CR) of radiocaesium in brown rice in Japan in 1995-2000,[37] 2002-2007 and in Fukushima and Ibaraki Prefectures in 2011-2014. Open circles show outliers. The yellow band shows 95% range of data in 1995-2007 and the red line shows geometric means of the relevant year range.

The CR values of ^{R}Cs for brown rice grown under normal fertilizer-open field conditions in each year from 2011 to 2014 and the CR data of global fallout ^{137}Cs in 1995-2007 are listed in Table 1. Individual CR data from literature (2011-2014) is given in the Supporting Information Table 1 (Table S1). When logarithm data of the CRs were compared, no difference was found among CR values for Fluvisol, Andosol and Cambisol in 1995-2007. After the FDNPP accident, using all the data in 2011 and 2012, CR values were significantly higher than those in 1995-2007 by multiple *t*-test ($p<0.001$), especially in 2011. The post-accident CR values in 2011 have a large variation with a geometric standard deviation (GSD) of 3.4 compared with the global fallout value of 2.7 when all soil types were included. Comparing soil types in 2011, the CR values of Fluvisol were significantly lower than those of Andosol and Gleysol; thus, soil type would affect the variation of CR values in 2011. Unfortunately, in 2012 the only soil type recorded was Fluvisol so soil type comparison was not possible. In 2013 and 2014, for all soil types, there was no significant difference in CR to those estimated for before the FDNPP accident, probably due to the aging effect in soil. Thus, it is likely that even if additional K fertilizer had not been applied, the CR values would have returned to that prevailing before the FDNPP accident within three years after ^{R}Cs deposition for the FDNPP. Although soil types were different, the post-Chernobyl data from sandy or organic soils also showed a clear decrease in CR with time.[8] These results suggest that in mineral soils the majority of 'aging' occurs quickly; the information would be useful for post-accident management.

Fluvisol is the only soil type for which CR can be compared before and after the FDNPP accident. Fluvisol occupies ca. 80% of paddy fields in Japan (including Fukushima and Ibaraki prefectures).[34] The GM of the CR of radiocaesium in 1995-2007, 2011, 2012, 2013 and 2014 were 3.1×10^{-3}, 4.4×10^{-3}, 4.8×10^{-3}, 2.2×10^{-3}, and 2.1×10^{-3}, respectively. According to *t*-test analysis the GM value was significantly higher than before the FDNPP accident only in 2012; however, the difference was small.

The caesium fixation capacity in soil can vary depending on the different contents of different types of clay minerals, organic matter, competitive ions (e.g., K^+ and NH_4^+), and pH in soil.[2,10,38-41] Nakao et al.[40] found that micaceous minerals in soil collected in Fukushima largely contributed to the ^{R}Cs retention ability of the soil clays. Conversely, soil organic matter can increase ^{R}Cs mobility in soil by forming soluble organic-matter-bound-Cs[38] and by inhibition of Cs fixation on clay mineral.[41] Furthermore, the passage of time and the water management regime also affects Cs fixation. Takeda et al.[2] reported that a drying-wetting cycle affected Cs sorption sites of smectite in soil; 1M NH_4OAc extractability of added Cs in gray lowland soil, which is a Fluvisol type soil, decreased with time possibly due to increasing the number of Cs selective sites in smectite during the drying-wetting cycle. Under typical water management of rice paddy fields, soils are flooded during the rice growing season and then dried after harvest. Therefore, drying-wetting cycle could influence the decrease in Cs uptake by plants with time. To quantify ^{R}Cs bioavailability, the Radiocaesium Interception Potential (RIP) was proposed by Cremers et al.,[42] and the method was incorporated into the transfer parameter handbook by the IAEA.[10] Inherent variability in the CR value is likely to be at least a factor of 10 due to the different soil characteristics in paddy fields, variable rice growing conditions, impact of catchment water dynamics and soil management every year in rice paddy fields.

Table 1. Summary of brown rice/soil concentration ratios of radiocaesium

Year	Soil group	N	GM	GSD	min	max
1995-2007	All	120	3.4×10^{-3}	2.7	3.9×10^{-4}	4.1×10^{-2}
	Andosol	15	3.1×10^{-3}	1.8	9.8×10^{-4}	6.9×10^{-3}
	Fluvisol	60	3.1×10^{-3}	2.9	7.7×10^{-4}	4.1×10^{-2}
	Gleysol	48	4.5×10^{-3}	2.8	3.9×10^{-4}	2.9×10^{-2}
	Others	7	1.7×10^{-3}	3.1	4.3×10^{-4}	8.1×10^{-3}
2011	All	62	1.2×10^{-2}	3.4	9.7×10^{-4}	1.7×10^{-1}
	Andosol	10	1.7×10^{-2}	1.4	1.2×10^{-2}	2.8×10^{-2}
	Fluvisol	25	4.4×10^{-3}	2.1	9.7×10^{-4}	3.7×10^{-2}
	Gleysol	9	2.5×10^{-2}	2.3	6.3×10^{-3}	7.7×10^{-2}
	Others	18	1.9×10^{-2}	4.6	1.3×10^{-3}	1.7×10^{-1}
2012	All	70	5.6×10^{-3}	2.6	4.9×10^{-4}	8.6×10^{-2}
	Fluvisol	54	4.8×10^{-3}	2.2	4.9×10^{-4}	4.4×10^{-2}
	Others	16	9.5×10^{-3}	3.8	5.8×10^{-4}	8.6×10^{-2}
2013	All	32	3.5×10^{-3}	3.3	3.9×10^{-4}	4.3×10^{-2}
	Fluvisol	18	2.2×10^{-3}	3.3	3.9×10^{-4}	2.9×10^{-2}
	Others	14	6.4×10^{-3}	2.5	9.6×10^{-4}	4.3×10^{-2}
2014	All	27	2.3×10^{-3}	3.1	4.5×10^{-4}	6.9×10^{-2}
	Fluvisol	17	2.1×10^{-3}	2.1	5.3×10^{-4}	7.1×10^{-3}
	Others	10	2.8×10^{-3}	4.9	4.5×10^{-4}	6.9×10^{-2}

Comparison of study CR values with other literature data. The CR values derived from the dataset for (i) open field-normal fertilizer, (ii) pot experiments with normal fertilizer, and (iii) open field experiments with No-K fertilizer and have been compared in Figure 4. The number of

CR values for pot and No-K were smaller than those for open field-normal fertilizer, and were only available for 2011 and 2012. The geometric mean for CR values of pot-normal fertilizer, and open field-No-K in 2011 were 1.2×10^{-2} and 2.2×10^{-2}, respectively, and those in 2012 were 1.8×10^{-2} and 1.4×10^{-2}, respectively. Tests of significant difference using a multiple *t*-test were carried out on the CR data after conversion into logarithm data. There was no difference amongst the three treatments in the 2011 data, so the data in the pot experiment were within the range of CR values determined in field conditions. In contrast, for 2012 the CR value for pot experiment data were significantly higher than those for open field-normal fertilizer ($p<0.01$). The GM value of No-K data was slightly higher than those of open field-normal fertilizer, but there were no significant differences in CR values between the two treatments.

Saito and Sakuma[43] reported that for the same soil type, the [^{R}Cs] in rice grains was higher in pot studies than for rice collected from open field experiments. No soil concentration data was given in this paper so that we could not include their data in our analysis. It is possible that the pot conditions encourage higher extract of ^{R}Cs from the soil into the soil solution from which it would be taken up by the plant roots. Possible reasons for the effect include (i) the relatively high density of plant roots in small pots enabling close root contact with soil, and (ii) the release of organic acids from roots into soil to extract exchangeable cations.[44] However, further studies are necessary to clarify whether these or other mechanisms are responsible for the observed difference.

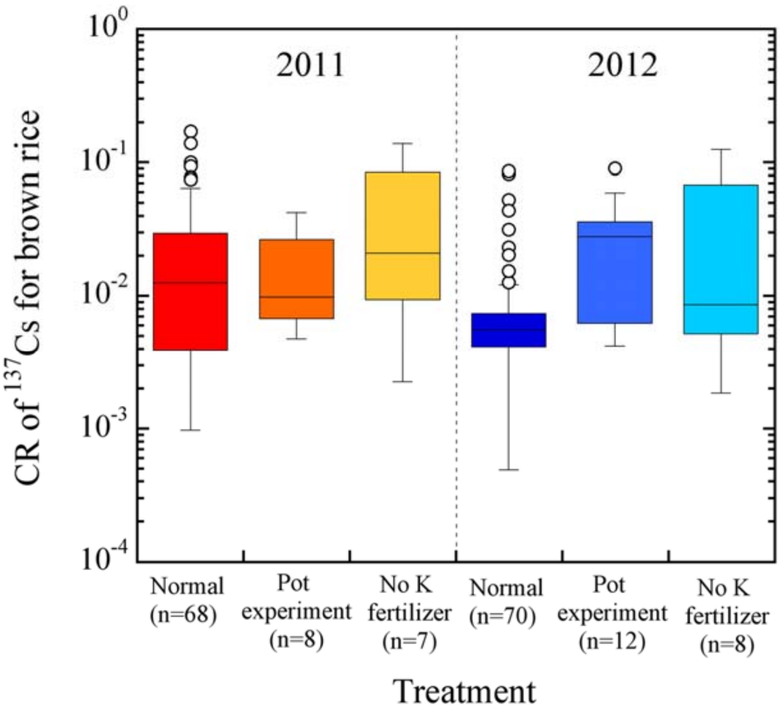

Figure 4. Box plots of concentration ratios from soil to brown rice in Japan for three different treatments in 2011 and 2012. Open circles show outliers. Normal: Open field data with normal fertilizer, Pot experiment: Pot experiment data with normal fertilizer; and No K: Open field data fertilized without K.

In this study, in the FDNPP-accident affected areas, we found a roughly four-fold increase in the geometric mean ^{R}Cs CR in 2011 compared with that observed before the FDNPP accident. This increased CR had the potential to increase the amount of radiocaesium ingested in both 2011 and 2012, because once a year rice production is common in Japan so that rice consumption continues to the next year harvest season. For example, in 2011, 6.3-18.6% of total ^{R}Cs intake was from rice consumption.[45] Intake of radiocaesium via rice was markedly reduced by the intensive restrictions and monitoring of foodstuff that occurred after the FDNPP accident.

Because the CR of rice decreased rapidly, the amount of ^{R}Cs ingestion from rice would also have decreased and reached a similar level to that occurring before the FDNPP-accident as estimated by Smith et al.[9]

Although the aging effect was clearly found in mineral soils, the equilibrium CR would be affected by the caesium fixation capacity in soil due to the different constituents and rice variety/cultivars. In this study, we did not find CR differences among soil types, however, CR values of Cs for rice has been reported to vary considerably in other reports.[10] Also, in this study, we did not consider rice variety/cultivar differences previously reported by Kojima et al.[46] It would also be useful to include variety/cultivar specification as well as soil type into the future dataset, although data analysis may be restrained by the small number of data for each variety/cultivar and soil types.

ASSOCIATED CONTENT

Supporting Information.

The following file is available free of charge.

Brown rice/soil concentration ratio of radiocaesium (PDF)

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Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

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Notes

The authors declare no competing financial interest.

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ABBREVIATIONS

CR, Concentration ratio; FDNPP, Fukushima Daiichi Nuclear Power Plant; MODARIA, Development, Testing and Harmonization of MOdels and DAta for Radiological Impact Assessment.

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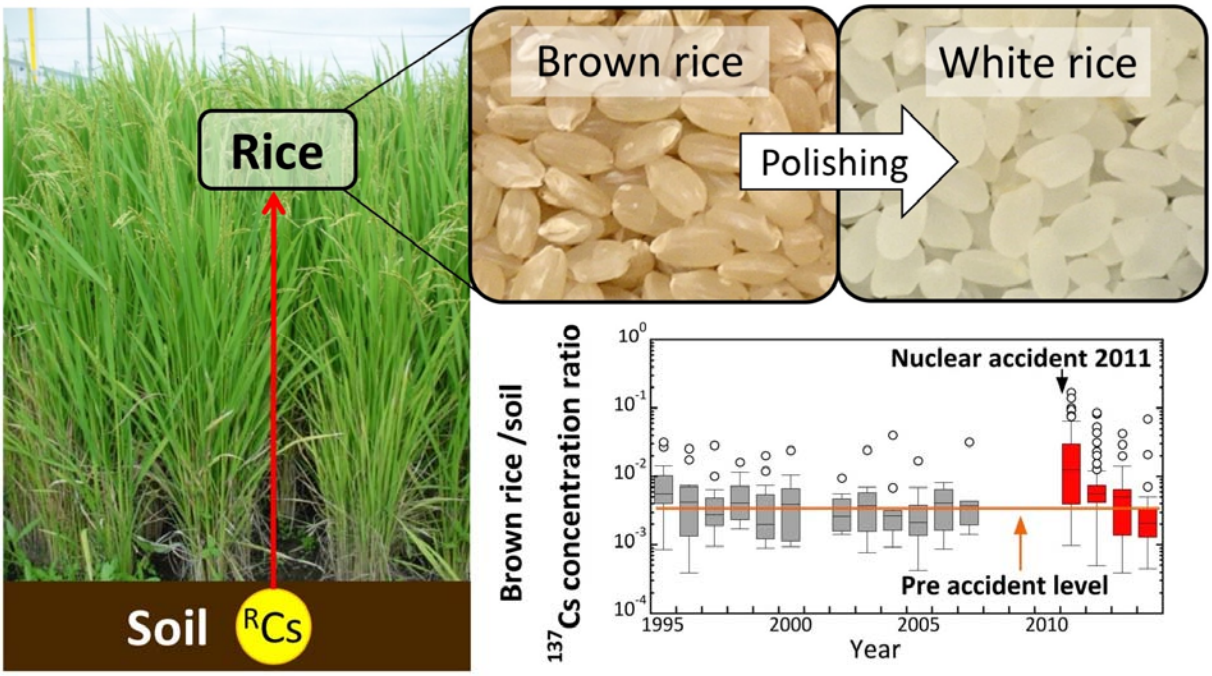
TOC



Rice
Brown rice
White rice
Polishing
Soil
RCs
Brown rice /soil
137Cs concentration ratio
Nuclear accident 2011
Pre accident level
Year
1995
2000
2005
2010
10^0
10^-1
10^-2
10^-3
10^-4