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

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11
12 **KEYWORDS:** Soil-to-plant transfer; Aging effect; Caesium-137; Paddy rice; Global fallout.

13
14 **ABSTRACT**

15 Radiocaesium (^RCs) mobility in soil is initially relatively high when the nuclide first comes into
16 contact with soil, after which the mobile fraction decreases with time due to ^RCs fixation to soil

17 particles (aging effect). Consequently, the ^{137}Cs activity concentration in plants grown in soil was
18 expected to decrease with time after the Fukushima Daiichi nuclear power plant accident in
19 2011. In this study, we collated data on concentration ratios (CR) of ^{137}Cs between brown rice
20 grain and paddy soil and compared CR values reported for periods before and after the accident.
21 For this purpose, soil and rice data were collected after the accident specifically from paddy
22 fields which did not have additional potassium fertilizer added (for remediation purposes). The
23 geometric mean rice/soil CR of ^{137}Cs for all types of soil was 1.2×10^{-2} in 2011 (n=62) and by 2013
24 the value had declined to 3.5×10^{-3} (n=32) which was similar to that for 1995-2007 of 3.4×10^{-3}
25 (n=120). The comparison suggests that the mean soil-to-rice grain concentration ratio had
26 returned to that prevailing before the accident after less than three years. It was also confirmed
27 that CR values for rice sampled from paddy fields were lower than those obtained from pot
28 experiments.

29 INTRODUCTION

30 The mobility of radiocaesium (^{137}Cs) in soil is at its highest when the radionuclide initially
31 deposits onto soil; then, with time, the mobile fraction gradually decreases and reaches an
32 equilibrium due to ^{137}Cs fixation to soil particles,^{1,2} commonly called an aging effect. Because ^{137}Cs
33 in the mobile fraction in soil solution is taken up through plant roots,^{2,3} the ^{137}Cs activity
34 concentration, [^{137}Cs], in plants also decreases with time after addition to soils.⁴⁻⁸ Accordingly,
35 after the TEPCO's Fukushima Daiichi Nuclear Power Plant (FDNPP) accident occurred on
36 March 11, 2011, it was expected that the mobility of ^{137}Cs released from the accident (reported as
37 ^{134}Cs , ^{137}Cs or $^{134+137}\text{Cs}$ in this study) in agricultural fields would initially be higher than that
38 observed under equilibrium conditions. In contrast, equilibrium conditions are expected to have

39 been reached for ^{137}Cs from global fallout measured in samples taken many years after its peak
40 deposition period of the late 1950s and 1960s.

41 In this study, we focused on brown rice because rice is a staple food in Japan and other Asian
42 countries. Brown rice has the husk removed, and white rice is produced by removing ca. 9-10%
43 of the surface layer of the brown rice grain by weight (i.e. bran layer). In Japan, many
44 measurement data of $^{\text{R}}\text{Cs}$ are available for soil and brown rice samples collected both before and
45 after the FDNPP accident.⁹ The transfer of $^{\text{R}}\text{Cs}$ to crops grown in contaminated soil was expected
46 to be high in 2011 compared with that reported previously in IAEA Technical Report Series No.
47 472¹⁰ for which equilibrium conditions are assumed.

48 To avoid underestimation of brown rice [$^{\text{R}}\text{Cs}$], relatively high transfer was assumed by the
49 authorities for crops in the first year after the FDNPP accident to enable limits to be set for the
50 recommencement of rice cultivation¹¹; the $^{\text{R}}\text{Cs}$ transfer ratio from soil to brown rice was assumed
51 to be 0.1. Thus, only paddy fields in which the [$^{\text{R}}\text{Cs}$] did not exceed 5,000 Bq kg⁻¹ dry weight
52 (dw) was allowed to be used for rice cultivation,¹¹ because the provisional regulation value, 500
53 Bq kg⁻¹ in raw food materials for $^{\text{R}}\text{Cs}$ to restrict the distribution of contaminated foods, was used
54 at that time. In 2011, 26,464 brown rice samples were measured in Japan, and only 39 samples
55 (0.15%) exceeded the provisional regulation value.¹² This outcome was expected given the
56 restrictions on use of paddy fields over the conservatively set limit, but the reasons why the 39
57 samples exceeded the regulation value needed to be identified.

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

62 ^{6,14-16} Most of the relevant data reported are for crops other than rice, but there was one pre-
63 FDNPP accident study which reported this feature for brown rice.⁶ Therefore, as a remediation
64 measure, additional K fertilizer (above that normally used) was applied to soil after the FDNPP-
65 accident that was used to grow rice plants^{13,17,18} and other crops such as soybeans.¹⁹ Because the
66 soil exchangeable K fraction was comprehensively monitored and controlled in contaminated
67 areas, the [^RCs] in brown rice did not exceed the Japanese standard limit of [^RCs] in crops of 100
68 Bq kg⁻¹ (as of April 1, 2012) in 2015-2017.²⁰ The number of continuous measurements of soil-to-
69 crop transfer of ^RCs decreased with time,²¹⁻²⁴ with most reports providing data for only one to
70 two years (written in Japanese and English languages). To better understand the time dependency
71 of soil-to-crop transfer of ^RCs, available data were collated from the literature through a
72 comprehensive analysis of the peer-reviewed data. We also identified ¹³⁷Cs data for brown rice
73 and associated soil samples to obtain estimates of transfer for the period before the FDNPP-
74 accident, and carried out measurements of global fallout ¹³⁷Cs using archived soil and brown rice
75 sample sets collected in 2006-2007.

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

81

82 MATERIALS AND METHODS

83 **Calculation of Soil-to-rice Concentration Ratio (CR).** To provide a simple approach for
84 estimating the transfer of radionuclides from soil to crops in radioecological models for dose

85 assessment, the radionuclide activity concentration on dw basis in the crop is compared with that
86 in the ploughed soil layer.¹⁰ In this study, the soil-to-rice concentration ratio (CR) of ^RCs was
87 calculated using the following empirical ratio:

88

89 $CR = \text{Activity concentration in rice grain (Bq kg}^{-1}\text{-dw)} / \text{Activity concentration in soil (Bq kg}^{-1}\text{-}$
90 dw)

91

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

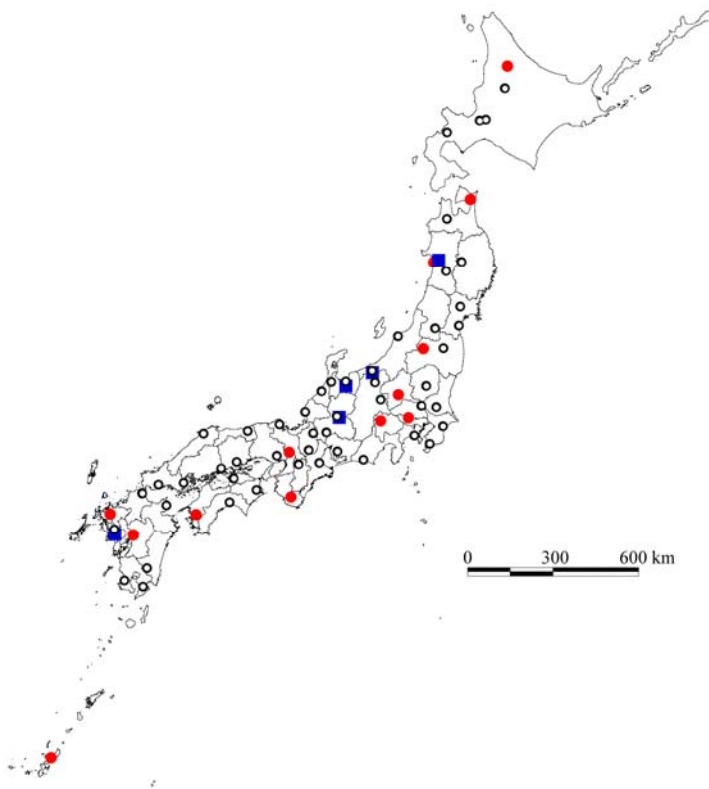
99 For the collated data, if brown rice [^RCs] data were reported in fresh weight, edible state, then
100 the normal water content of 15% was applied to obtain a dw basis for the concentration data.

101

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

108 31.1 Bq kg⁻¹-dw (number of samples determined: 50), respectively. Additional ¹³⁷Cs data were
109 derived by measurements during this study of samples collected in 2006 (n=13) and 2007 (n=5).

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



113
114 **Figure 1.** Paddy soil-brown rice sample set collection sites in Japan from 2002-2007. Open
115 circle: collection sites previously reported^{25,26}, closed circle: collected in 2006, and closed
116 square: collected in 2007.

117
118
119 For ¹³⁷Cs determination, the volume of each sample was adjusted to 100-mL amount of the air-
120 dried and sieved soil sample or incinerated brown rice sample in a 260-mL plastic vessel

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

126

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

139

140 **Data Survey-2: CR Data Survey from Literature in 2011-2014.** Brown rice and associated
141 soil data were compiled from published papers and institutional reports in Japanese and English.
142 Only data for normal fertilizer, no-K fertilizer (hereafter, No-K), and no fertilizer application
143 conditions were collated for rice paddies and pot experiments. A normal fertilizer application

144 condition is defined as the amount of fertilizer added annually by the farmers before the FDNPP
145 accident including K-fertilizer. Because soil conditions differ in paddy fields, it is difficult to
146 specify the range in the amount of fertilizer added; however, according to the analysis results by
147 MAFF,³³ potassium fertility in soil was almost within the range of appropriate condition for rice
148 production with normal fertilizer application before the FDNPP-accident. There are many papers
149 studying potassium fertilizer effect on ¹³⁷Cs soil to plant uptake, but few of them were for normal
150 fertilizer conditions. In contrast, all the CR values reported before the FDNPP accident were for
151 normal fertilizer conditions. If fertilizer conditions were not mentioned we assumed that such
152 papers applied normal fertilizer amounts. Rice cultivars were not classified; because it is often
153 not specified in the available Japanese literature. Thus, it was assumed that all data were for the
154 most common Japonica variety of rice because the variety is common in Japan.

155 Data were accepted under the following criteria.

- 156 - If CR values of ^RCs were reported in the publication.
- 157 - If [^RCs] data in both brown rice and soil were reported allowing a calculation of CR values.
- 158 - data were available in figures only, in which case the [^RCs] were derived from the figures and
159 CR values were calculated.

160

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

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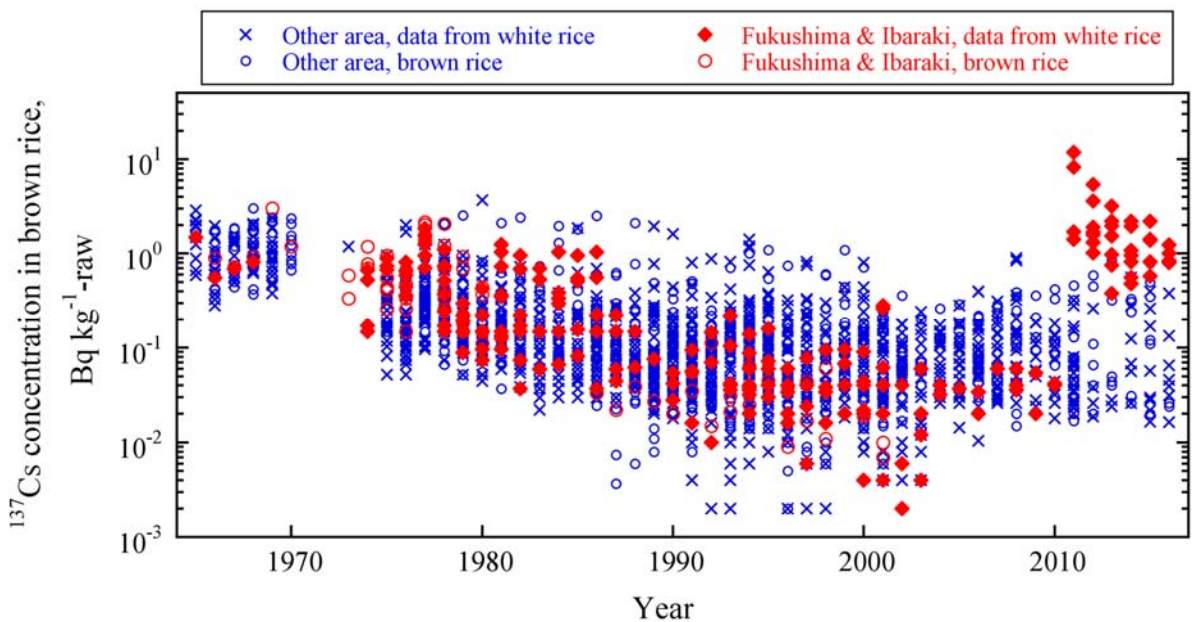
166 RESULTS AND DISCUSSION

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

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

181 Figure 2 shows that the [¹³⁷Cs] in brown rice collected in Fukushima and Ibaraki Prefectures in
182 2016 (0.8-1.24 Bq kg⁻¹-raw, n=5) were one order of magnitude higher than that before the
183 FDNPP accident, i.e. geometric mean [¹³⁷Cs] for all data was 0.076 Bq kg⁻¹-raw in 2006-2010
184 (n=151). In Fukushima and the surrounding contaminated prefectures, the sampling areas where
185 continuous ¹³⁷Cs measurement in brown rice had been carried out before the FDNPP accident
186 were not used afterwards so the data collated by NRA are not directly comparable. Therefore, the
187 order of magnitude difference is somewhat imprecise. Overall, the data show a decreasing trend
188 in [^RCs] in brown rice in Area (1).

189 We could not find openly available continuous soil-to-brown rice data sets from the same
 190 sampling field(s) to provide site-specific ^{137}Cs time trends in rice grains over the five-year period.
 191 Nevertheless, for herbaceous plants collected from Chiba campus of the QST-NIRS located
 192 about 220 km south from the FDNPP, we observed an almost constant [^{137}Cs] from 1000 d after
 193 March 11, 2011.³⁶ Such data suggests that in Area (1) most of the decline occurred in 2011-2013
 194 with less decline in 2013-2014. Since 2015, the total [^{137}Cs] in all brown rice produced in
 195 Fukushima Prefecture has not exceeded the standard limit of 100 Bq kg^{-1} .²⁰ Thus, the aging
 196 effect, together with remediation measures (e.g., addition of K fertilizers to keep exchangeable K
 197 concentration to a suitable condition in soil, addition of soil amendments to fix ^{137}Cs in soils, or
 198 removal of surface layer of agricultural soils to reduce ^{137}Cs), has ensured that the [^{137}Cs] in rice
 199 had decreased considerably since deposition.
 200



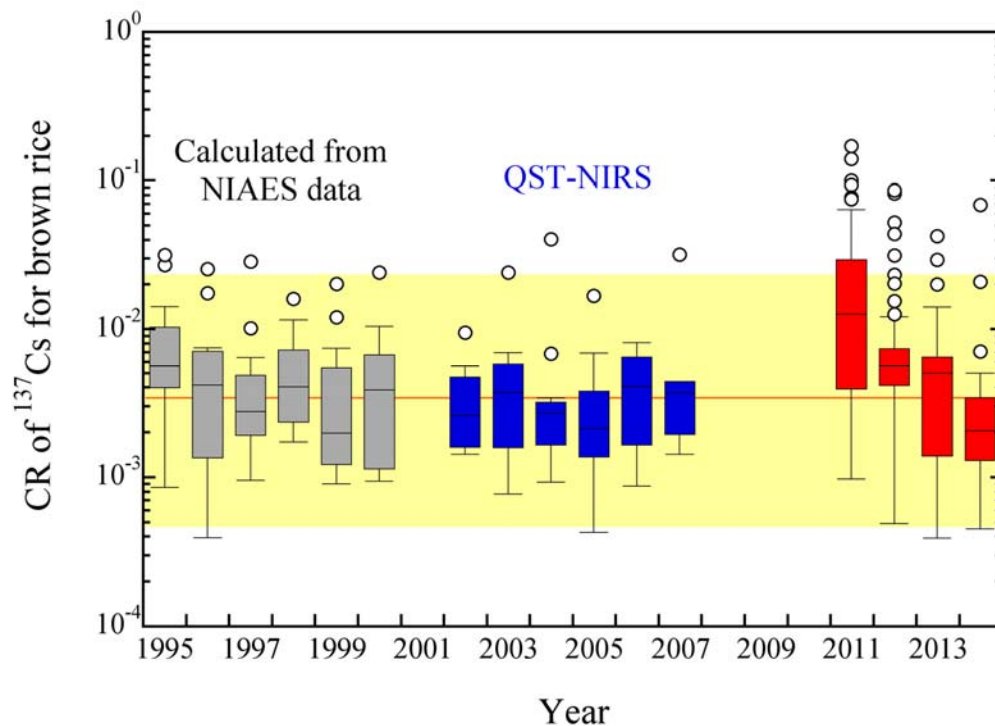
201
 202 **Figure 2.** Measured and estimated [^{137}Cs] in brown rice in Japan in 1965-2016. ^{137}Cs in brown
 203 rice was estimated using the data in white rice, using a brown rice/white rice ^{137}Cs concentration
 204 ratio of 2.0.

205

206 **Comparison of CR values for ^{137}Cs in brown rice before and after the FDNPP accident.** In
207 the NRA database, combined brown rice-soil datasets were difficult to reliably identify.
208 Therefore, other reported data were used including our own data. The National Institute for
209 Agro-Environmental Sciences (NIAES) had been measuring radiocesium in brown rice and
210 associated soil samples continuously in 1959-2000 from all over Japan (approximately 15
211 stations each year).³⁷ The resulting brown rice and associated soil data were used to calculate CR
212 values from 1995-2000. We also used our previously reported values and the data we obtained in
213 this study for 2002-2007 (QST-NIRS): the geometric mean value of CR was 2.9×10^{-3} . In some
214 incinerated brown rice samples, ^{137}Cs was not detectable due to the low levels originating from
215 global fallout; nevertheless it was detectable in 37 data of 50 brown rice samples from 2002-
216 2005, 7 data of 13 brown rice samples from 2006 (0.0086-0.094 Bq kg⁻¹-dw) and all 5 samples
217 from 2007 (0.027-0.41 Bq kg⁻¹-dw). For comparison, [^{137}Cs] in soil samples in 2006-2007 were
218 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
219 undetected data in 2002-2007, the geometric mean value of CR was 1.9×10^{-3} . From this result,
220 the estimated CR values using detected [^{137}Cs] data are likely a bit higher than those that would
221 have been derived if all samples were above detection limits.

222 The literature survey data for CR values of ^{137}Cs for brown rice after the FDNPP accident
223 collected in open paddies with normal fertilizer conditions are shown in Figure 3 together with
224 the data for before the FDNPP accident from NIAES³⁴ and QST-NIRS. Data in each sampling
225 year were close to log-normal distributions. The yellow band shows the 95% range of CR values
226 in 1995-2007 (4.6×10^{-4} - 2.5×10^{-2}), the geometric mean value was 3.4×10^{-3} in the same year
227 range.

228 The CR value was clearly higher in 2011 in Fukushima and Ibaraki prefectures. Because rice
 229 planting in open paddy fields starts around early May, and by that time in 2011 the amounts of
 230 newly deposited ^{137}Cs was smaller than those in March and April, the direct deposition effect to
 231 rice grains (typical flowering time is in July-August) was considered to be negligible.
 232



233
 234 **Figure 3.** Box-plots of the concentration ratio (CR) of radiocaesium in brown rice in Japan in
 235 1995-2000,³⁷ 2002-2007 and in Fukushima and Ibaraki Prefectures in 2011-2014. Open circles
 236 show outliers. The yellow band shows 95% range of data in 1995-2007 and the red line shows
 237 geometric means of the relevant year range.

238
 239

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

259 Fluvisol is the only soil type for which CR can be compared before and after the FDNPP
260 accident. Fluvisol occupies ca. 80% of paddy fields in Japan (including Fukushima and Ibaraki
261 prefectures).³⁴ The GM of the CR of radiocaesium in 1995-2007, 2011, 2012, 2013 and 2014
262 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

263 analysis the GM value was significantly higher than before the FDNPP accident only in 2012;
264 however, the difference was small.

265 The caesium fixation capacity in soil can vary depending on the different contents of different
266 types of clay minerals, organic matter, competitive ions (e.g., K^+ and NH_4^+), and pH in soil.^{2,10,38-}
267 ⁴¹ Nakao et al.⁴⁰ found that micaceous minerals in soil collected in Fukushima largely
268 contributed to the ^RCs retention ability of the soil clays. Conversely, soil organic matter can
269 increase ^RCs mobility in soil by forming soluble organic-matter-bound-Cs³⁸ and by inhibition of
270 Cs fixation on clay mineral.⁴¹ Furthermore, the passage of time and the water management
271 regime also affects Cs fixation. Takeda et al.² reported that a drying-wetting cycle affected Cs
272 sorption sites of smectite in soil; 1M NH_4OAc extractability of added Cs in gray lowland soil,
273 which is a Fluvisol type soil, decreased with time possibly due to increasing the number of Cs
274 selective sites in smectite during the drying-wetting cycle. Under typical water management of
275 rice paddy fields, soils are flooded during the rice growing season and then dried after harvest.
276 Therefore, drying-wetting cycle could influence the decrease in Cs uptake by plants with time.
277 To quantify ^RCs bioavailability, the Radiocaesium Interception Potential (RIP) was proposed by
278 Cremers et al.,⁴² and the method was incorporated into the transfer parameter handbook by the
279 IAEA.¹⁰ Inherent variability in the CR value is likely to be at least a factor of 10 due to the
280 different soil characteristics in paddy fields, variable rice growing conditions, impact of
281 catchment water dynamics and soil management every year in rice paddy fields.

282

283 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}

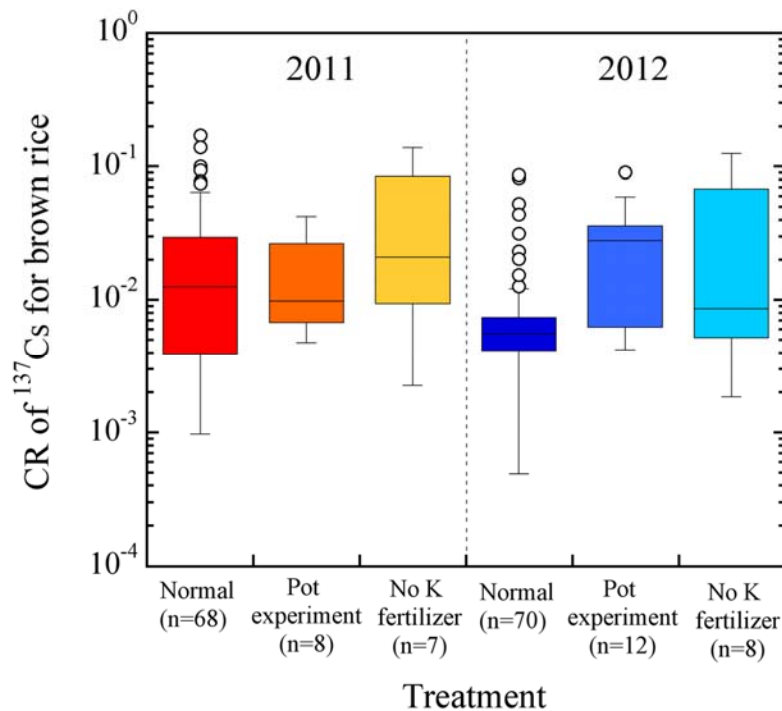
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286 **Comparison of study CR values with other literature data.** The CR values derived from the
 287 dataset for (i) open field-normal fertilizer, (ii) pot experiments with normal fertilizer, and (iii)
 288 open field experiments with No-K fertilizer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

289 CR values for pot and No-K were smaller than those for open field-normal fertilizer, and were
290 only available for 2011 and 2012. The geometric mean for CR values of pot-normal fertilizer,
291 and open field-No-K in 2011 were 1.2×10^{-2} and 2.2×10^{-2} , respectively, and those in 2012 were
292 1.8×10^{-2} and 1.4×10^{-2} , respectively. Tests of significant difference using a multiple *t*-test were
293 carried out on the CR data after conversion into logarithm data. There was no difference amongst
294 the three treatments in the 2011 data, so the data in the pot experiment were within the range of
295 CR values determined in field conditions. In contrast, for 2012 the CR value for pot experiment
296 data were significantly higher than those for open field-normal fertilizer ($p < 0.01$). The GM value
297 of No-K data was slightly higher than those of open field-normal fertilizer, but there were no
298 significant differences in CR values between the two treatments.

299 Saito and Sakuma⁴³ reported that for the same soil type, the [^RCs] in rice grains was higher in
300 pot studies than for rice collected from open field experiments. No soil concentration data was
301 given in this paper so that we could not include their data in our analysis. It is possible that the
302 pot conditions encourage higher extract of ^RCs from the soil into the soil solution from which it
303 would be taken up by the plant roots. Possible reasons for the effect include (i) the relatively high
304 density of plant roots in small pots enabling close root contact with soil, and (ii) the release of
305 organic acids from roots into soil to extract exchangeable cations.⁴⁴ However, further studies are
306 necessary to clarify whether these or other mechanisms are responsible for the observed
307 difference.



308

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

313

314 In this study, in the FDNPP-accident affected areas, we found a roughly four-fold increase in
 315 the geometric mean ^RCs CR in 2011 compared with that observed before the FDNPP accident.
 316 This increased CR had the potential to increase the amount of radiocaesium ingested in both
 317 2011 and 2012, because once a year rice production is common in Japan so that rice
 318 consumption continues to the next year harvest season. For example, in 2011, 6.3-18.6% of total
 319 ^RCs intake was from rice consumption.⁴⁵ Intake of radiocaesium via rice was markedly reduced
 320 by the intensive restrictions and monitoring of foodstuff that occurred after the FDNPP accident.

321 Because the CR of rice decreased rapidly, the amount of ¹³⁷Cs ingestion from rice would also have
322 decreased and reached a similar level to that occurring before the FDNPP-accident as estimated
323 by Smith et al.⁹

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

332

333 ASSOCIATED CONTENT

334 **Supporting Information.**

335 The following file is available free of charge.

336 Brown rice/soil concentration ratio of radiocaesium (PDF)

337

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343 **Author Contributions**

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350

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352 The authors declare no competing financial interest.

353

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357

358 **ABBREVIATIONS**

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

362

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