

## Influence of water management and fertilizer application on $^{137}\text{Cs}$ and $^{133}\text{Cs}$ uptake in paddy rice fields

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1 **Influence of Water Management and Fertilizer Application on  $^{137}\text{Cs}$  and  $^{133}\text{Cs}$  Uptake in Paddy Rice Fields**

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## Highlights:

We examined Cs uptake by rice with varying water management in fields in 2012–2014.

Long flooding treatment enhanced  $^{137}\text{Cs}$  and  $^{133}\text{Cs}$  uptakes by rice plant.

The successive K fertilization brought cumulative effect on Cs uptake of rice plant.

Uptakes of  $^{137}\text{Cs}$  and  $^{133}\text{Cs}$  by rice became correlated over the elapsed year.

1 **Abstract**

2 Cesium-137 derived from the Tokyo Electric Power Company's Fukushima Dai-ichi  
3 Nuclear Power Plant (FDNPP) accident contaminated large areas of agricultural land in  
4 Eastern Japan. Previous studies before the accident have indicated that flooding enhances  
5 radiocesium uptake in rice fields. We investigated the influence of water management in  
6 combination with fertilizers on  $^{137}\text{Cs}$  concentrations in rice plants at two fields in southern  
7 Ibaraki Prefecture. Stable Cs ( $^{133}\text{Cs}$ ) in the plants was also determined as an analogue for  
8 predicting  $^{137}\text{Cs}$  behavior after long-term aging of soil  $^{137}\text{Cs}$ . The experimental periods  
9 comprised 3 y starting from 2012 in one field, and 2 y from 2013 in another field. These fields  
10 were divided into three water management sections: a long-flooding section without  
11 midsummer drainage, and medial-flooding, and short-flooding sections with one- or  
12 two-week midsummer drainage and earlier end of flooding than the long-flooding section. Six  
13 or four types of fertilizer subsections (most differing only in potassium application) were  
14 nested in each water management section. Generally, the long-flooding treatment led to higher  
15  $^{137}\text{Cs}$  and  $^{133}\text{Cs}$  concentrations in both straw and brown rice than medial- and short-flooding  
16 treatments, although there were some notable exceptions in the first experimental year at each  
17 site. Effects of differing potassium fertilizer treatments were cumulative; the effects on  $^{137}\text{Cs}$   
18 and  $^{133}\text{Cs}$  concentrations in rice plants were not obvious in 2012 and 2013, but in 2014, these  
19 concentrations were highest where potassium fertilizer had been absent and lowest where  
20 basal dressings of K had been tripled. The relationship between  $^{137}\text{Cs}$  and  $^{133}\text{Cs}$  in rice plants  
21 was not correlative in the first experimental year at each site, but correlation became evident  
22 in the subsequent year(s). This study demonstrates a novel finding that omitting midsummer  
23 drainage and/or delaying drainage during the grain-filling period enhances uptake of both  
24  $^{137}\text{Cs}$  and  $^{133}\text{Cs}$ .

25

26 **Keywords:** cesium, potassium fertilizer, flooding, rice, water management

27

## 28 **1. Introduction**

29 The accident at the Tokyo Electric Power Company's Fukushima Dai-ichi Nuclear Power  
30 Plant (FDNPP), triggered by the Great East Japan Earthquake and subsequent Tsunami in  
31 March 2011, widely contaminated the agricultural environment in the southern Tohoku and  
32 northern Kanto districts with radionuclides. Cesium-137 ( $^{137}\text{Cs}$ ) has a long half-life of 30 y,  
33 and its transfer to agricultural products is a long-lasting problem that demands  
34 countermeasures. The optimization of potassium (K) fertilizer application is an effective  
35 means to accomplish this (Kato et al., 2015; Saito et al., 2015) because K competes with  $^{137}\text{Cs}$   
36 in the transfer process from soil solution to plant body (Shaw and Bell, 1991; Smolders et al.,  
37 1997).

38 Rice (*Oryza sativa*) cropping in flooded soil is the representative agricultural system in  
39 Japan. Some previous studies before the FDNPP accident indicate that soil flooding enhances  
40 radiocesium uptake by the rice plant. Tensho et al. (1961) demonstrated using pot-culture  
41 experiments with artificial addition of radiocesium that the uptake of radiocesium by rice  
42 plants is much greater from flooded soil than from unflooded soil. They suggested that  
43 ammonium ( $\text{NH}_4^+$ ) as the main form of inorganic nitrogen in flooded soil might enhance  
44 availability of radiocesium to plants because  $\text{NH}_4^+$  could exchange with radiocesium  
45 selectively adsorbed in soil. Pot-culture experiments by D'Souza and Mistry (1980) generated  
46 results similar to those of Tensho et al. (1961). Verfaillie et al. (1967) demonstrated in an  
47 actual paddy field in Northern Italy in 1964 that preventing flooding decreased  $^{137}\text{Cs}$   
48 concentration in rice grain. Although these studies compared the effects of flooding and  
49 upland cultivation practices on radiocesium uptake by rice plants, it remains unknown  
50 whether variations in the flooding period within a practical range for paddy rice cropping

51 affect  $^{137}\text{Cs}$  uptake by rice plants. Smolders and Tsukada (2011) suggested that water  
52 management systems that suppress  $\text{NH}_4^+$  concentration might be a potential countermeasure  
53 against  $^{137}\text{Cs}$  transfer to rice crops, and advocated factorial experiments focused on water  
54 management and nitrogen (N) fertilization.

55 We conducted field experiments in paddy crops to investigate the influence of water  
56 management (flooding period) on  $^{137}\text{Cs}$  concentration in rice plants. Additionally, the effects  
57 of different K application treatments were investigated. Stable cesium-133 ( $^{133}\text{Cs}$ ) is regarded  
58 as a useful analogue for long-term assessment of  $^{137}\text{Cs}$  in agricultural environments, because  
59 the fate of radionuclides in the environment follows the behavior of their stable isotopes  
60 (Tsukada et al., 2002; Uchida and Tagami, 2007). In this study,  $^{133}\text{Cs}$  in rice plants was also  
61 determined so the influence of water management and fertilizer treatment on  $^{137}\text{Cs}$  uptake  
62 long after the fallout event might be predicted.

63

## 64 **2. Materials and Methods**

65

### 66 **2.1. Experimental Fields**

67 The experimental area comprised two paddy fields in southern Ibaraki Prefecture. Site YWR  
68 is located in the alluvial plain of the Kokaigawa River in Tsukubamirai City. Site KND is  
69 located on the Hitachi tableland in Tsukuba City. According to the soil classification system in  
70 Japan (Obara et al., 2011) or the World Reference Base for Soil Resources (International  
71 Union of Soil Sciences Working Group WRB, 2014) respectively, YWR soil is Gray Lowland  
72 soil or Gleyic Fluvisol, and KND soil is Upland Reformed soil or Thaptandic Regosol  
73 (Transportic). These fields had no history of deep tillage after the FDNPP accident, therefore  
74  $^{137}\text{Cs}$  was mainly distributed within topsoil.

75 In previous rice cropping before this experiment in each field, water management had been

76 spatially uniform for more than a decade. A common practice in these areas, called  
77 “midsummer drainage,” is aimed at enhancing root growth and reducing the number of  
78 unproductive tillers of rice plants. It also promotes subsurface drainage through formation of  
79 drying cracks in the soil and increases the soil’s bearing capacity for machinery operation  
80 (Inoue and Tokunaga, 1995). In previous rice cropping at YWR, field flooding was started and  
81 topsoil was puddled in early May, a few days before transplanting, as was customary for the  
82 water management system. Basal-dressing fertilizer had been applied before flooding. In late  
83 June, ponding water was temporarily drained for 2–3 weeks as midsummer drainage. Field  
84 flooding ended in early September, a week before harvest. As was customary at KND, field  
85 flooding was started and topsoil was puddled in early May. A few days later, basal-dressing  
86 fertilizer was applied to the flooded soil. Rice seedlings were transplanted a few days later  
87 thereafter. The midsummer drainage was conducted for a week from early July. During  
88 drainage period in KND, drainpipe buried in the soil at a depth of 90 cm was opened to  
89 promote underdrainage. Field flooding ended at the end of August, two weeks before harvest.  
90 (Other details on field management before this experiment, such as fertilizer application, are  
91 summarized in Supplementary Tables.)

92 The experimental periods comprised 3 y from 2012 at the YWR field, and 2 y from 2013 at  
93 the KND field. The cultivar “Koshihikari” was transplanted in May and raised until harvest in  
94 September. The schedule of paddy field management during the experiment is shown in Table  
95 1. During the season, air temperature ranged from 4.9 to 36.8°C (mean temperature of  
96 23.4°C) and precipitation was recorded as 580, 380 and 760 mm in 2012, 2013 and 2014,  
97 respectively at the nearest meteorological observatory (Japan Meteorological Agency, 2015).

98

## 99 **2.2. Experimental design**

100 The experimental designs of the two fields are shown in Fig. 1. Each field was divided into

101 three sections for different water management treatments using two plastic corrugated sheets  
102 (Nami-ita in Japanese). Each section was identified by the three type of water management as  
103 follows: A long-flooding (LF) section, which was flooded from before transplanting to the end  
104 of August or early September; a medial-flooding (MF) section, which was drained in  
105 midsummer for 7 or 8 d and 7–12 d earlier at the end of flooding than the LF section; and a  
106 short-flooding (SF) section, which was drained in midsummer for 14–16 d and 15–20 d  
107 earlier at the end of flooding than the LF section. In 2014, however, midsummer drainage for  
108 the MF and SF sections was increased to 20 d because of prolonged rainy weather. At KND,  
109 only one water outlet existed for surface drainage; therefore, pumps were used to discharge  
110 standing water at that site.

111 Each water management section was divided into six (at YWR) or four (at KND)  
112 subsections for different fertilizer treatments. Control subsections (CR) at each site received  
113  $3.0 \text{ g m}^{-2}$  of N as ammonium sulfate ( $(\text{NH}_4)_2\text{SO}_4$ ),  $2.6 \text{ g m}^{-2}$  of P as calcium superphosphate  
114 (mainly  $\text{Ca}(\text{H}_2\text{PO}_4)_2 \cdot \text{H}_2\text{O}$ ), and  $5.0 \text{ g m}^{-2}$  of K as potassium chloride (KCl) as basal dressing,  
115 and  $3.0 \text{ g m}^{-2}$  of N as  $(\text{NH}_4)_2\text{SO}_4$  and  $2.5 \text{ g m}^{-2}$  of K as KCl as top-dressing in late July. Three  
116 types of subsections at each site were defined according to the KCl application strategy; no K  
117 fertilizer application (0K: no K), triple basal dressing without top-dressing (3BK: totally  $15 \text{ g}$   
118  $\text{m}^{-2}$  K), and triple top-dressing without basal dressing (3TK: totally  $7.5 \text{ g K m}^{-2}$  K). Of the  
119 remaining subsections at YWR, one type received  $7.5 \text{ g m}^{-2}$  of K as potassium silicate  
120 ( $\text{K}_2\text{SiO}_3$ ), which is a release fertilizer, as basal dressing instead of KCl (KS). The other type  
121 differs from the control subsections in having had no top-dressing of KCl (0TK) in 2012 and  
122 increased N fertilizer application in 2013 and 2014 (IN:  $4 \text{ g m}^{-2}$  of N as  $(\text{NH}_4)_2\text{SO}_4$  and the  
123 same amount of N as controlled-release coated urea, LPSS100) as basal dressing.

124 The field position of each treatment-defined subsection was not changed during the  
125 experimental years in order to observe the cumulative effect of field management, especially



126 fertilizer application, over the years.

127

### 128 **2.3. Sample collection, and radiometric and chemical analysis**

129 In mid-September, the aerial parts of matured rice were harvested at about 2 cm above  
130 ground from a 5.0 or 6.5 m<sup>2</sup> area of each subsection. A sample of the underlying topsoil,  
131 which was 17 cm thick on average, was also collected from a harvest area of each subsection  
132 with a shovel or a root auger (4 cm diameter; DIK-102A, Daiki Rika Kogyo Co., Ltd., Japan).  
133 Brown rice samples were obtained after air-drying, husking, and sieving through 1.8 mm  
134 sieves. Straw samples were dried at 70 °C, brushed to remove surface tissue stained with soil  
135 remnants, and milled. Soil samples were air-dried and sieved through 2.0 mm sieves.

136 Concentrations of <sup>137</sup>Cs in straw, brown rice, and soil samples were determined with  
137 germanium (Ge) gamma-ray detectors (GEM55P, GEM20-70, SEIKO EG&G, Co., Ltd.,  
138 Japan; GC2020, GC2520, GC4020-7500SL-2002SCL, Canberra, USA) using 2.0 L of plant  
139 samples or 100 mL of soil samples. Before measurement, each sample was mixed to  
140 homogenize the material and was then uniformly packed into a plastic container. The counting  
141 efficiency of the Ge gamma-ray detectors was calibrated using gamma-ray reference source  
142 (MX033MR and MX033U8PP, Japan Radioisotope Association, Tokyo, Japan). The decay  
143 corrections were made to the harvest day in each year.

144 To measure <sup>133</sup>Cs and K concentration in brown rice and straw, 100 mg of milled sample  
145 was digested in duplicate with 70% nitric acid on the hot plate at 105°C. The digestion  
146 solution was analyzed using inductively coupled plasma mass spectrometry (Agilent 7700x,  
147 Agilent Technologies, Japan) and atomic absorption spectrometry (iCE 3300, Thermo Fisher  
148 Scientific K.K., Japan) to measure <sup>133</sup>Cs and K concentrations, respectively. The standards for  
149 calibration were prepared using multi-element calibration standard 3 (containing 10.0 mg L<sup>-1</sup>  
150 of <sup>133</sup>Cs; PerkinElmer, Inc., USA) and KCl powder (>99.5%; Wako Pure Chemical Industries,

151 Ltd., Japan). The average standard error between duplicates was 3% and 1% of the analytical  
152 value for  $^{133}\text{Cs}$  and K analysis, respectively.

153 To estimate exchangeable K, soil samples were shaken with  $1 \text{ mol L}^{-1}$  ammonium acetate  
154 solution at pH 7.0 for 1 h at a solution/soil ratio of  $10 \text{ mL g}^{-1}$ . The K concentration of the  
155 supernatant solution after centrifugation and filtration was determined by atomic absorption  
156 spectrometry. The extraction was duplicated, and the average standard error between  
157 duplicates was determined to be 1% of the analytical value.

158 To investigate the influence of water management and increasing N fertilizer application on  
159 soil  $\text{NH}_4^+$  concentration, the exchangeable  $\text{NH}_4^+$  content of soils in the CR and IN subsections  
160 was determined before and during cultivation in 2014 at the YWR field. Soil cores (3 cm  
161 diameter and 10 cm depth) were collected five times in triplicate: the days before  
162 transplanting (15 May), before midsummer drainage (23 June), after midsummer drainage in  
163 the MF and SF sections (16 July), after heading stage (6 August), and after drainage in the LF  
164 section (5 September). Each wet core sample was stirred to homogenize, and a part of it was  
165 used for 1-h extraction with  $2 \text{ mol L}^{-1}$  KCl solution (solution/soil ratio of  $10 \text{ mL g}^{-1}$ ) within  
166 the sampling day. After centrifugation and filtration, the  $\text{NH}_4^+$  concentration of the  
167 supernatant solution was determined using an Autoanalyzer (QuAatro 2-HR, BL-TEC, Japan).  
168 The remaining wet samples were used to estimate exchangeable K contents during cultivation.  
169 Similar to KCl extractions, 1-h extractions with  $1 \text{ mol L}^{-1}$  ammonium acetate solution were  
170 conducted. The K concentration of the supernatant solutions was determined using atomic  
171 absorption spectrometry.

172 The mass of dried plant material or soil is used in expression of all measured quantities  
173 (plant yields, amounts of chemical species).

174

## 175 **2.4. Statistical analysis**

176 The Grubbs' test was applied to identify outliers in datasets for each field and year using R  
177 software version 3.1.1 (The R Project for Statistical Computing, Vienna, Austria. URL  
178 <http://www.r-project.org>). A value of brown rice  $^{137}\text{Cs}$  for subsection 3TK of the SF section at  
179 KND in 2013 was regarded as an outlier at  $p < 0.001$ , and was thereby excluded from  
180 analyses.

181 The data of four subsections (CR, 0K, 3BK, and 3TK) for 2013–2014, which have  
182 counterparts in all sections of both fields, were subjected to mixed model analysis of variance  
183 (ANOVA) with the SAS Add-In for Microsoft office version 6.1 M1 (SAS Institute Inc.,  
184 USA). This dataset is structured as a split-plot design with site as the block, water  
185 management as the primary factor, fertilizer as the secondary factor, and experimental year as  
186 the tertiary factor. In this paper, the statistical term “significant” refers to  $p < 0.050$  and  
187 “significant tendency” to  $0.050 < p < 0.100$ . When the effects of water management or  
188 fertilizer were significant without any interaction in ANOVA, Tukey's post hoc multiple  
189 comparison test was performed to determine significant difference among sections or  
190 subsections. When significant interactions between experimental year and water management  
191 or fertilizer were revealed by ANOVA, a post-hoc Tukey's test was performed separately for  
192 both 2013 and 2014 datasets at an adjusted significance level ( $p < 0.050 / 2 = 0.025$ ).

193 Correlations between  $^{137}\text{Cs}$  and  $^{133}\text{Cs}$  concentrations in straw and brown rice were calculated  
194 by linear regression using Microsoft Excel 2013.

195

### 196 **3. Results**

197 The soil  $^{137}\text{Cs}$  concentrations were closely similar in the two fields (Table 2). For each field  
198 in each year, the average soil  $^{137}\text{Cs}$  concentration for each type of water management or  
199 fertilizer treatment ranged within a narrow interval (for example, 153–177 Bq kg<sup>-1</sup> at YWR  
200 and 152–185 Bq kg<sup>-1</sup> at KND in 2014). The ANOVA results for the 2013–2014 dataset

201 revealed no significant differences in  $^{137}\text{Cs}$  among soils that experienced different water  
202 management or fertilizer treatment, but there were significant differences for the different  
203 experimental years. The values were lower in 2013 than in other years, which could be  
204 attributed to a sampling error caused by using a different tool in 2013 (root auger) than in the  
205 other years (shovels). When root augers were vertically inserted into the soil during sampling,  
206 the penetration resistance appeared to be lower than when a shovel was used, indicating that  
207 the sampling with the auger may have been deeper. This could have allowed accidental  
208 inclusion of the less-contaminated soil from the plow sole in the topsoil samples.

209 The exchangeable K content after harvest was higher at YWR than at KND (Table 2). The  
210 values for 0K and 3BK were the lowest and the highest, respectively, among the values for  
211 different fertilizer treatments, corresponding to their ranking in respect to K application  
212 amounts. The average exchangeable K content for 3BK increased annually in each field, in  
213 contrast to stable or decreasing values for 0K, so the differences widened over years. Water  
214 management sections exhibited small exchangeable K differences at KND. At YWR, the  
215 difference was negligible in 2012, but in the subsequent years, the exchangeable K content  
216 was highest in the soil that had experienced longer drainage treatment (SF). The ANOVA  
217 results for the 2013–2014 dataset revealed significant fixed effects of fertilizer and  
218 experimental year and their interaction (data not shown).

219 The annual mean values of yields of straw and brown rice varied slightly across the different  
220 kinds of water management or fertilizer treatments (Table 3). Although these yields for LF at  
221 YWR were relatively low in 2014, ANOVA for the 2013–2014 dataset revealed no significant  
222 fixed effects for yields of both materials (data not shown). Straw yields were lower in the first  
223 experimental year than in the subsequent year(s) in each field.

224 The K concentration in straw and brown rice varied more between experimental years than  
225 between experimental treatments (Table 4). Water management practices and fertilizer

226 treatments did not cause the value to vary by more than 10% from the grand mean in each  
227 year at each field. The ANOVA results for the 2013–2014 dataset revealed a significant  
228 difference only between experimental years (data not shown).

229 The  $^{137}\text{Cs}$  and  $^{133}\text{Cs}$  concentrations found in straw and brown rice are presented in Fig. 2.  
230 The ANOVA results for the 2013–2014 dataset are summarized in Table 5.

231 Concentration of  $^{137}\text{Cs}$  in straw was higher for LF than for MF and SF in each case, except  
232 for 0K at KND in 2013 (Fig. 2a). The annual mean values for each water management are  
233 shown in Fig. 2 together with the annual grand mean (white bars in Fig. 2). Compared to MF,  
234 the annual mean values for LF were higher by 28%–52%, while those of SF were almost the  
235 same, except in each field's first experimental year when annual mean values for SF were  
236 lower by 12% (YWR) and 32% (KND). Among the different fertilizer treatments, the mean  
237 values in 2014 were highest for 0K at both fields and lowest for in 3BK at KND. The values  
238 in each section and subsection mostly decreased from year to year. The ANOVA results for the  
239 2013–2014 dataset showed significant influences of water management and year without any  
240 interaction (Table 5). The Tukey's post hoc test indicated that the differences between LF and  
241 the other two kinds of water management were significant.

242 Concentration of  $^{137}\text{Cs}$  in brown rice was also higher for LF than for MF and SF in most  
243 cases at YWR and in most 2014 cases at KND (Fig. 2b). At YWR, the annual mean values for  
244 LF were higher by 33%–57% than those for MF. The mean value for SF was lower than that  
245 for MF by 29% in 2012, but in 2013 and 2014 the mean values for SF and MF were almost  
246 equal. At KND in 2013, the annual mean value for brown rice  $^{137}\text{Cs}$  was higher for SF than  
247 for LF and MF, contrary to the straw  $^{137}\text{Cs}$  results. Among the different fertilizer treatments,  
248 the mean values in 2014 were highest for 0K at both fields and lowest for 3BK at KND, in  
249 accordance with the straw results. The annual grand mean (white bars in Fig. 2) for each field  
250 decreased from the first experimental year to the second year. The ANOVA results for the

251 2013–2014 dataset did not show significant influence of water management but revealed  
252 interactions of water management with year (fixed effect) and with field (random effect)  
253 (Table 5). It also showed significance or significant tendency ( $p < 0.10$ ) for fertilizer, year,  
254 and their interaction. The separate Tukey's post hoc test detected no significant difference in  
255 2013, but it showed that  $^{137}\text{Cs}$  in brown rice was significantly higher for 0K than for three  
256 other fertilizer treatments in 2014.

257 Concentration of  $^{133}\text{Cs}$  in straw was higher for LF than for MF and SF in most cases after  
258 2012 (Fig. 2c). At YWR, the differences in annual mean values for the different types of water  
259 management were negligible in 2012 but increased from year to year. Among the different  
260 fertilizer treatments, the values in 2014 were highest for 0K and lowest for 3BK at both fields.  
261 The ANOVA results for the 2013–2014 dataset showed significant tendencies for the effects  
262 of water management and fertilizer along with a significant interaction between fertilizer and  
263 year (Table 5). The separate Tukey's post hoc test for each year detected no significant  
264 difference for 2013, but it showed that  $^{133}\text{Cs}$  concentration in straw was significantly lower for  
265 3BK than for 0K and 3TK in 2014.

266 Concentration of  $^{133}\text{Cs}$  in brown rice exhibited trends similar to  $^{133}\text{Cs}$  concentration in straw  
267 (Fig. 2d). Additionally, the annual grand mean for each field was higher for 2014 than for  
268 earlier years. The increase from 2013 to 2014 is most evident in LF among water management  
269 treatments. At KND in 2013, the annual mean values were higher in LF than in MF and SF,  
270 which was consistent with both  $^{137}\text{Cs}$  and  $^{133}\text{Cs}$  in straw but not with  $^{137}\text{Cs}$  in brown rice. The  
271 ANOVA results for the 2013–2014 dataset showed significant influence of water management,  
272 fertilizer, and year (Table 5). However, it is not feasible to follow multiple comparison tests  
273 for any factor because there was also significant interaction within each pair of these three  
274 factors.

275 The relationship between  $^{133}\text{Cs}$  and  $^{137}\text{Cs}$  concentrations in each plant component (straw and

276 brown rice) are exhibited in Fig. 3, separately for each field and each year. There was no  
277 significant correlation in the first experimental year for each site but positive correlation was  
278 observed in the subsequent year(s).

279 The exchangeable  $\text{NH}_4^+$  and K contents of soil from selected subsections before and during  
280 rice cultivation in 2014 at YWR are exhibited in Fig. 4. Exchangeable  $\text{NH}_4^+$  content was  
281 higher for IN than for CR on the day before transplanting (Fig. 4a). After that, the  
282 exchangeable  $\text{NH}_4^+$  content became consistently low, regardless of the difference in  
283 application amount of N fertilizer. Midsummer drainage in the MF and SF sections did not  
284 change the values. During the grain-filling period, the  $\text{NH}_4^+$  content increased with no clearly  
285 evident difference between different types of water management or different fertilizer  
286 treatments. Exchangeable K content decreased from before transplanting to after the heading  
287 stage (Fig. 4b). After the end of flooding, exchangeable K values increased in SF sections.

288

## 289 **4. Discussion**

### 290 **4.1. Influence of water management**

291 Except the case of brown rice in 2013 at KND, the  $^{137}\text{Cs}$  concentrations of straw and brown  
292 rice showed highly significant correlation (YWR in 2012–2014,  $r=0.786$ ,  $p<0.001$ ; KND in  
293 2014,  $r=0.900$ ,  $p<0.001$ ) and average  $^{137}\text{Cs}$  concentrations for water management sections  
294 were ordered in the same way in the sense that the highest  $^{137}\text{Cs}$  concentrations were in the LF  
295 sections (Fig. 2a, b). The average  $^{133}\text{Cs}$  concentrations of straw and brown rice for the water  
296 management sections were in the same order as the average  $^{137}\text{Cs}$  concentrations (highest in  
297 the LF sections) except at YWR in 2012 (Fig. 2c, d). In these field experiments, the soil  $^{137}\text{Cs}$   
298 concentrations of the water management sections in the two fields were in about the same  
299 range (Table 2). The exchangeable K content was also closely similar among the sections of  
300 each field at the end of the first experimental year. Therefore, the relatively high  $^{137}\text{Cs}$

301 concentrations in rice plants in the LF sections are not attributable to spatial differences of soil  
302 quality in terms of either  $^{137}\text{Cs}$  concentration or K fertility. Variations of plant yields were  
303 small and insignificant (Table 3) indicating negligible effect of carbohydrate dilution. These  
304 results indicate as a whole that the enhanced rice plant Cs (both  $^{137}\text{Cs}$  and  $^{133}\text{Cs}$ )  
305 concentrations in LF sections (LF effect) was the outcome of the long-flooding treatment  
306 itself. The interaction between water management and fertilizer was not significant except for  
307 brown rice  $^{133}\text{Cs}$  (Table 5), and the LF effect was consistent across the different fertilizer  
308 treatments in most cases (for all cases in 2014, Fig. 2). Thus the LF treatment seems to have  
309 been influential regardless of fertilizer management and field. On the other hand, the effect of  
310 doubling the drainage period (i.e., the difference between the treatments in SF and MF) was  
311 not significant.

312 Possible causes of the LF effect on Cs uptake are discussed hereafter. Tensho et al. (1961)  
313 suggested that  $\text{NH}_4^+$  exchanges with radiocesium selectively absorbed in soil particles to  
314 enhance its availability to the plant under submerged conditions. Ammonium also has been  
315 reported to play an important role in radiocesium release from submerged sediments (Comans  
316 et al., 1989; Evans et al., 1983); also, liberal application of  $\text{NH}_4^+$  enhances plant uptake of  
317 radiocesium (Jackson et al., 1965; Lasat et al., 1997; Ohmori et al., 2014; Prister et al., 1992).  
318 It should be investigated whether or not  $\text{NH}_4^+$  derived from mineralization or a practical  
319 amount of fertilizer does contribute to Cs mobility, and whether oxidation by drainage  
320 treatment could suppress this  $\text{NH}_4^+$  contribution. In this study, application of 1.3 times more N  
321 to the IN subsections than to the control subsections produced no appreciable difference in Cs  
322 concentration in the plants (Fig. 2). The differences in exchangeable  $\text{NH}_4^+$  content among  
323 soils that experienced different water management treatments were small in the observation at  
324 YWR in 2014 (Fig. 4). Ammonium is not necessarily considered to be the cause of the LF  
325 effect at this moment.



326      Secondarily, the possibility of exogenous  $^{137}\text{Cs}$  entry from irrigation water might be  
327 considered. However, Suzuki et al. (2015) found that irrigation with water containing  
328 dissolved  $^{137}\text{Cs}$  at a concentration of  $0.10 \text{ Bq L}^{-1}$  did not or did only slightly (by less than  
329 20%) increase the  $^{137}\text{Cs}$  concentration of brown rice grown in pot culture using soil containing  
330  $^{137}\text{Cs}$  at  $200 \text{ Bq kg}^{-1}$ . Although  $^{137}\text{Cs}$  concentrations of irrigation water were not measured in  
331 our experiment, they were assumed to be lower than  $0.10 \text{ Bq L}^{-1}$ . In the investigation of Tsuji  
332 et al. (2014) in four rivers located in Fukushima Prefecture in 2012 and 2013, dissolved  $^{137}\text{Cs}$   
333 concentrations in river water were less than  $0.20 \text{ Bq L}^{-1}$  in the river where deposited  $^{137}\text{Cs}$  is  
334 highest, and these were less than  $0.05 \text{ Bq L}^{-1}$  in the other three rivers. Additionally, the  
335 differences in total duration of flooding among the water management sections were four  
336 weeks or less in this study. Thus the differences of  $^{137}\text{Cs}$  inflow load among the water  
337 management sections are considered not to be a main cause of the LF effect. Similar  
338 enhancements of radiocesium uptake by flooding were reported in pot experiments with no  
339 exogenous radiocesium entry (D'Souza and Mistry, 1980; Tensho et al., 1961).

340      Thirdly, D'Souza and Mistry (1980) suggested that shoot-base absorption of radiocesium in  
341 standing water can be a major means by which flooding increases radiocesium uptake, in  
342 reference to a report by Myttenaere (1972) that absorption of radiocesium by rice plants is  
343 greater through a shoot base dipped in water than through a root dipped in nutrient solution.  
344 However, the nutrient solution in the hydroponic experiment seems different from natural soil  
345 solution in the point of salt concentration. To estimate the realistic contribution of shoot-base  
346 radiocesium absorption to rice plants, the composition of the test solutions (K,  $\text{NH}_4^+$ ,  $^{133}\text{Cs}$   
347 etc.) should be modified according to that of soil solution and standing water in the actual  
348 field.

349      A fourth possibility is that K availability can be changed with longer flooding to influence  
350 Cs uptake. Reductive conditions reportedly decrease K release (Chen et al., 1987; Horikawa

351 and Kawaguchi, 1963), and oxidation of paddy soil by drainage decreases K-deficient  
352 symptoms in rice plants (Ogihara, 1960). At YWR in 2014, the exchangeable K content  
353 increased after the end of flooding where drainage had started earlier (Fig. 4b). This increase  
354 may have been caused by soil oxidation and related to lower Cs uptake in the SF section.  
355 Other causes are possible, but no clear mechanism for the LF effect is evident yet.

356 Results of  $^{133}\text{Cs}$  measurements for the different water management treatments at YWR show  
357 a trend not shared by  $^{137}\text{Cs}$  results; LF treatment did not change  $^{133}\text{Cs}$  concentrations in rice  
358 plants (both straw and brown rice) in 2012 but increased these in later years (Fig. 2c, d). We  
359 propose a two-fold hypothesis to explain the trend specific to  $^{133}\text{Cs}$ . First, the discordance in  
360 variation between  $^{133}\text{Cs}$  and  $^{137}\text{Cs}$  could have been caused by  $^{133}\text{Cs}$  uptake from the subsoil.  
361 Unlike  $^{137}\text{Cs}$ ,  $^{133}\text{Cs}$  uptake in rice plants comes from not only topsoil but also subsoil, where  
362 water and nutrient conditions are different from those. If  $^{133}\text{Cs}$  uptake in subsoil was  
363 substantial,  $^{133}\text{Cs}$  concentration in rice plant would not have reflected LF treatment. Second,  
364 repeated LF treatments might gradually have suppressed vertical growth of rice root into the  
365 subsoil. Consequently,  $^{133}\text{Cs}$  uptake would be enhanced by LF treatment in a manner similar  
366 to  $^{137}\text{Cs}$  uptake when root activity was predominant in topsoil. Straw samples in the LF  
367 section showed significant correlation between  $^{133}\text{Cs}$  and  $^{137}\text{Cs}$  concentrations among six  
368 subsections in 2013 ( $r = 0.905$ ,  $p < 0.01$ ) and 2014 ( $r = 0.802$ ,  $p < 0.05$ ), indicating that the  
369 uptake sources of both  $^{137}\text{Cs}$  and  $^{133}\text{Cs}$  were the same. In contrast, in the cases of MF and SF,  
370 there are no significant correlation in each year. Midsummer drainage reportedly promotes  
371 vertical growth of rice root (Kawata and Katano, 1977) and nutrient uptake from subsoil  
372 (Kaneda, 1995). In the plow sole and subsoil, rice roots grow along with cracks and tubular  
373 macro-pores (Kaneda, 1995; Kawata et al., 1980). These macro-pores are formed by drying  
374 and previous root activity, whereas they are clogged by clay particles deposited during  
375 puddling (Inoue and Tokunaga, 1995). Therefore, repeatedly skipping midsummer drainage

376 should decrease macro-pores in plow soles and subsoil and suppress vertical growth of rice  
377 root. Compared with MF and SF sections, the LF section at YWR had a low yield of rice  
378 plants in 2014 (Table 3), and became lower in exchangeable K contents in topsoil year by year  
379 (Table 2). These results are consistent with the hypothesis that repeated LF treatment  
380 concentrated root activity within topsoil. At KND, the trend specific to  $^{133}\text{Cs}$  uptake discussed  
381 above was not obvious. However, the increase in  $^{133}\text{Cs}$  concentrations in brown rice grown in  
382 the LF section from 2013 to 2014 (Fig. 2d) can be explained by this hypothesis.

383

#### 384 **4.2. Influence of K fertilizer application**

385 The effect of K fertilizer is significant or has a significant tendency for brown rice and straw  
386  $^{133}\text{Cs}$ , and there are significant interactions with year in all cases (Table 5). In 2014, plants  
387 under 0K treatments had the highest Cs concentrations in both straw and brown rice of all  
388 fertilizer treatments (Fig. 2), and Tukey's post-hoc test indicated some significant differences  
389 from other subsections (Table 5). Conversely, the lowest values for Cs concentrations were  
390 commonly observed in plants under the 3BK treatment, especially at KND. However, neither  
391 treatment significantly changed K concentration in the rice plants (Table 4). These results  
392 indicate that K availability was sufficient for rice plant requirements in all subsections, but its  
393 variation within this range affected Cs uptake by rice plant. Increase of the K basal dressing  
394 would more effectively enhance K availability at KND than YWR because the former site was  
395 poorer in exchangeable K (Table 2). However, the order of Cs uptake for different K  
396 application treatments (3BK < other subsections < 0K) was not observed before 2013 even at  
397 KND. The temporally broadening range of exchangeable K content across the subsections  
398 suggests that K application cumulatively affected soil K availability over the years of the  
399 experiment (Table 2). This accumulation would explain the changes in response to K fertilizer  
400 treatments. Although application timing of K fertilizer (i.e., basal dressing or top-dressing) is

401 reportedly important to suppress radiocesium concentration in rice plants (Nobori et al., 2014;  
402 Saito et al., 2015), the effect of the 3BK treatment on Cs uptake observed in this study is  
403 mainly attributed to cumulative enhancement of soil K fertility by application of high  
404 amounts of K.

405

#### 406 **4.3. Influence of time**

407 Plant  $^{137}\text{Cs}$  values were expected to decrease over the years of the experiment along with  
408 aging of soil  $^{137}\text{Cs}$  (Rigol et al., 1999; Roig et al., 2007). Accordingly, these decreases were  
409 observed (Fig. 2a, b, Table 5), but the decrease from 2013 to 2014 at YWR was very small. In  
410 this study, however, soil  $^{137}\text{Cs}$  aging might not be the only cause of these decreases. The grand  
411 means of exchangeable K content increased by 9% at YWR and by 11% at KND from the first  
412 year to the second year. The increasing K availability in the entire field also might have  
413 contributed to the difference of  $^{137}\text{Cs}$  uptake between experimental years.

414 The  $^{137}\text{Cs}$  and  $^{133}\text{Cs}$  concentrations in rice plants over the elapsed years became correlated  
415 during the experimental period (Fig. 3). The observation that  $^{137}\text{Cs}$  concentrations gradually  
416 began to follow those of  $^{133}\text{Cs}$  over time can be explained by the aging of soil  $^{137}\text{Cs}$ . As shown  
417 in Fig. 3, however, the variation range of stable  $^{133}\text{Cs}$  becomes wider to fit that of  $^{137}\text{Cs}$ ,  
418 except in the case of straw at KND. The widening variation of  $^{133}\text{Cs}$  was caused by the  
419 repetition of water management methods and fertilizer treatments. Repeated LF treatments  
420 increased  $^{133}\text{Cs}$  uptake by rice plant, which might be attributed to concentration of root  
421 activity within topsoil (Section 4.1). Repeated K fertilizer treatments cumulatively affected K  
422 availability, and resulted in a significant difference in  $^{133}\text{Cs}$  uptake by rice plants between the  
423 0K and 3BK treatments (Section 4.2). Although  $^{133}\text{Cs}$  uptake from subsoil might reflect K  
424 availability not in topsoil but in subsoil, repeated K fertilization might promote leaching from  
425 topsoil, and thereby enhance K availability in subsoil. The convergence of  $^{137}\text{Cs}$  and  $^{133}\text{Cs}$

426 concentrations over the elapsed years can be attributed to both aging of soil  $^{137}\text{Cs}$  and the  
427 cumulative effects of repetitive water management practices and fertilizer treatments.

428

#### 429 **4.4. Effectiveness of water management and K fertilization treatments in reducing** 430 **$^{137}\text{Cs}$ transfer to rice**

431 The enhancement of  $^{137}\text{Cs}$  uptake by the LF treatment was observed in both of the  
432 experimental fields, which had differing K status, site location, and soil taxa. The similar  
433 enhancement of natural  $^{133}\text{Cs}$  concentration in rice plants after 2012 implies that LF treatment  
434 could be influential on  $^{137}\text{Cs}$  transfer to rice plants even after long-term aging of soil  $^{137}\text{Cs}$ ,  
435 but the absence of an LF effect on  $^{133}\text{Cs}$  in 2012 needs explanation. That is, the changing  
436 water management from LF to MF could be an effective, long-term method to reduce  $^{137}\text{Cs}$   
437 transfer to rice. On the other hand, extending the drainage period in midsummer and  
438 grain-filling period to more than that of MF appears to be fruitless. In rice cropping systems  
439 in Japan, midsummer drainage is commonly practiced, and the total flooding period is shorter  
440 than that in the LF section of this study. Therefore, changing the practice of LF might be  
441 applicable only in fields where drainage treatment is skipped or imperfectly practiced due to  
442 low water permeability, inflow of mountain runoff, or other reasons. In such fields, efforts to  
443 practice midsummer drainage and/or to terminate flooding earlier in the grain-filling period  
444 could decrease  $^{137}\text{Cs}$  transfer to rice. However, the effects of these two practices were not  
445 individually investigated in this study. Additionally, the cause and the extent of generalization  
446 of the phenomenon should be examined in subsequent studies.

447 Increased K fertilization (3BK) treatment significantly decreased Cs concentration in rice  
448 plants, but the effect was not immediately observed, especially at YWR. Kato et al. (2015)  
449 reports a negative correlation between exchangeable K content after harvest and transfer  
450 factor (the ratio of radiocesium concentration in brown rice to that in soil), but it was not clear

451 when exchangeable K content was higher than 3.8 mmol kg<sup>-1</sup>. In previous experiments to  
452 investigate the effect of K fertilizer on radiocesium uptake by rice plants, exchangeable K  
453 contents in used soil were often low (for example, <1 mmol kg<sup>-1</sup>, Fujimura et al. (2014) and  
454 Saito et al. (2012)). It is unclear whether K fertilization is effective in reducing Cs uptake by  
455 rice plants in fields where exchangeable K content is moderate or rich. The exchangeable K  
456 content in the first year was, on average, 4.7 mmol kg<sup>-1</sup> at YWR and 2.6 mmol kg<sup>-1</sup> at KND.  
457 This experiment shows that regular application of high amount of K is effective in reducing  
458 Cs uptake by rice plants, even where exchangeable K content is not low and the first K  
459 fertilization seems to have no effect.

460

## 461 **5. Conclusion**

462 The rice cultivation experiments in two fields in southern Ibaraki Prefecture exhibited the  
463 following results.

464 1. Cs uptake in rice plants was significantly affected in many cases by water management  
465 treatments; omitting midsummer drainage and/or delaying drainage during the grain filling  
466 period (LF) mostly enhanced Cs concentration. The phenomenon was observed to be  
467 independent of fertilizer application treatment type.

468 2. High K fertilization over successive years had a cumulative effect on Cs uptake by rice  
469 plants. The difference of K fertilizing was not recognized in plant Cs concentrations in 2012  
470 and 2013 but exhibited a significant influence in 2014. By 2014, the cumulative effect of  
471 applying no K clearly enhanced plant Cs concentrations, while the cumulative effect of  
472 tripling the K basal dressing had the opposite effect, especially in a field with lower K status.

473 3. The correlation between <sup>137</sup>Cs and <sup>133</sup>Cs uptakes was insignificant at first and became close  
474 and significant with elapsed years. The convergence between <sup>137</sup>Cs and <sup>133</sup>Cs is attributed to  
475 the aging of soil <sup>137</sup>Cs and the cumulative effects of repeated water management methods and

476 fertilizer treatments.

477 4. Efforts to practice midsummer drainage and/or terminate flooding earlier in the grain-filling  
478 period could be an effective countermeasure to suppress  $^{137}\text{Cs}$  transfer to rice plants in fields  
479 where drainage is skipped or imperfectly practiced. The effect would be promising even after  
480 long-term aging of  $^{137}\text{Cs}$ .

481

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492

## 493 **References**

- 494 Chen, S.Z., Low, P.F., Roth, C.B., 1987. Relation between potassium fixation and the  
495 oxidation state of octahedral iron. *Soil Sci. Soc. Am. J.* 51, 82–86.  
496 doi:10.2136/sssaj1987.03615995005100010017x
- 497 Comans, R.N.J., Middelburg, J.J., Zonderhuis, J., Woittiez, J.R.W., De Lange, G.J., Das, H.A.,  
498 Van Der Weijden, C.H., 1989. Mobilization of radiocaesium in pore water of lake  
499 sediments. *Nature* 339, 367–369.
- 500 D’Souza, T.J., Mistry, K.B., 1980. Absorption of gamma-emitting fission products and

501 activation products by rice under flooded and unflooded conditions from two tropical soils.  
502 Plant Soil 55, 189–198. doi:10.1007/BF02181798

503 Evans, D.W., Alberts, J.J., Clark III, R.A., 1983. Reversible ion-exchange fixation of  
504 cesium-137 leading to mobilization from reservoir sediments. Geochim. Cosmochim. Acta  
505 47, 1041–1049.

506 Fujimura, S., Ishikawa, J., Sakuma, Y., Saito, T., Sato, M., Yoshioka, K., 2014. Theoretical  
507 model of the effect of potassium on the uptake of radiocesium by rice. J. Environ.  
508 Radioact. 138, 122–31. doi:10.1016/j.jenvrad.2014.08.017

509 Horikawa, Y., Kawaguchi, K., 1963. Studies on the potassium-adsorption caused by  
510 soil-reduction part I. Soil Sci. Plant Nutr. 9, 15–22. doi:10.1080/00380768.1963.10431050

511 Inoue, H, Tokunaga, K, 1995. Soil and water management, in: Tabuchi, T., Hasegawa, S.  
512 (Eds.), Paddy Fields in the World. The Japanese Society of Irrigation, Drainage and  
513 Reclamation Engineering, Tokyo, pp. 303–325.

514 International Union of Soil Sciences Working Group WRB, 2014. World reference base for  
515 soil resources 2014. World Soil Resources Reports 106, Food and Agriculture  
516 Organization of the United Nations, Rome.

517 Jackson, W.A., Craig, D., Lugo, H.M., 1965. Effect of various cations on cesium uptake from  
518 soils and clay suspensions. Soil Sci. 99, 345–353.

519 Japan Meteorological Agency, 2015. Past meteorological data search, Tsukuba (Tateno)  
520 [http://www.data.jma.go.jp/obd/stats/etrn/index.php?prec\\_no=40&block\\_no=47646](http://www.data.jma.go.jp/obd/stats/etrn/index.php?prec_no=40&block_no=47646) (June,  
521 2015).

522 Kaneda, Y., 1995. Soil management and paddy rice root by non-tilled farming. Root research  
523 4, 47–51. (in Japanese)

524 Kato, N., Kihou, N., Fujimura, S., Ikeba, M., Miyazaki, N., Saito, Y., Eguchi, T., Itoh, S.,  
525 2015. Potassium fertilizer and other materials as countermeasures to reduce radiocesium



526 levels in rice: Results of urgent experiments in 2011 responding to the Fukushima Daiichi  
527 Nuclear Power Plant accident. *Soil Sci. Plant Nutr.* 61, 179–190.  
528 doi:10.1080/00380768.2014.995584

529 Kawata, S., Katano, M., 1977. Effect of water management of paddy fields on the direction of  
530 crown root growth and the lateral root formation of rice plants. *Japanese Journal of Crop*  
531 *Science* 46, 543–557. (in Japanese with English summary)

532 Kawata, S., Katano, M., Yamazaki, K., 1980. The root system development of rice plants in  
533 the worked- and sub-soils of actual paddy field. *Japanese Journal of Crop Science* 49,  
534 311–316. (in Japanese with English summary)

535 Lasat, M.M., Norvell, W.A., Kochian, L.V., 1997. Potential for phytoextraction of <sup>137</sup>Cs from  
536 a contaminated soil. *Plant Soil* 195, 99–106. doi:10.1023/A:1004210110855

537 Nobori, T., Kobayashi, N.I., Tanoi, K., Nakanishi, T.M., 2014. Effects of potassium in  
538 reducing the radiocesium translocation to grain in rice. *Soil Sci. Plant Nutr.* 60, 772–781.  
539 doi:10.1080/00380768.2014.947617

540 Myttenaere, C., 1972. Absorption of radiocaesium by flooded rice: relative importance of  
541 roots and shoot base in the transfer of radioactivity. *Plant Soil* 36, 215–218.  
542 doi:10.1007/BF01373473

543 Obara, H., Ohkura, T., Takata, Y., Kohyama, K., Maejima, Y., Hamazaki, T., 2011.  
544 Comprehensive Soil Classification System of Japan—First Approximation. *Bulletion of*  
545 *National Institute for Agro-environmental Sciences* 29. (in Japanese with English  
546 summary)

547 Ogihara, T., 1960. Studies on the potassium deficiency of rice plant. *Special Report in*  
548 *Fukuoka Prefecture Agricultural Experimental Station* 15. (in Japanese)

549 Ohmori, Y., Kajikawa, M., Nishida, S., Tanaka, N., Kobayashi, N.I., Tanoi, K., Furukawa, J.,  
550 Fujiwara, T., 2014. The effect of fertilization on cesium concentration of rice grown in a

551 paddy field in Fukushima Prefecture in 2011 and 2012. *J. Plant Res.* 127, 67–71.  
552 doi:10.1007/s10265-013-0618-7

553 Prister, B., Loshchilov, N., Perepelyatnikova, L., Perepelyatnikov, G., Bondar, P., 1992.  
554 Efficiency of measures aimed at decreasing the contamination of agricultural products in  
555 areas contaminated by the Chernobyl NPP accident. *Sci. Total Environ.* 112, 79–87.  
556 doi:10.1016/0048-9697(92)90240-S

557 Rigol, A., Roig, M., Vidal, M., Rauret, G., 1999. Sequential extractions for the study of  
558 radiocesium and radiostrontium dynamics in mineral and organic soils from Western  
559 Europe and Chernobyl areas. *Environ. Sci. Technol.* 33, 887–895. doi:10.1021/es980720u

560 Roig, M., Vidal, M., Rauret, G., Rigol, A., 2007. Prediction of radionuclide aging in soils  
561 from the Chernobyl and Mediterranean areas. *J. Environ. Qual.* 36, 943–952.  
562 doi:10.2134/jeq2006.0402

563 Saito, T., Ohkoshi, S., Fujimura, S., 2012. Effect of potassium application on root uptake of  
564 radiocesium in rice. in: *Proceedings of international symposium on environmental  
565 monitoring and dose estimation of residents after accident of TEPCO's Fukushima Daiichi  
566 Nuclear Power Station.* Kyoto University Research Reactor Institute Press, Kyoto, pp.  
567 165–169. doi:10.1007/s10967-014-3609-9

568 Saito, T., Takahashi, K., Makino, T., Tsukada, H., Sato, M., Yoshioka, K., 2015. Effect of  
569 application timing of potassium fertilizer on root uptake of <sup>137</sup>Cs in brown rice. *J.  
570 Radioanal. Nucl. Chem.* 303, 1585–1587. doi:10.1007/s10967-014-3609-9

571 Shaw, G., Bell, J.N.B., 1991. Competitive effects of potassium and ammonium on caesium  
572 uptake kinetics in wheat. *J. Environ. Radioact.* 13, 283–296.  
573 doi:10.1016/0265-931X(91)90002-W

574 Smolders, E., Van den Brande, K., Merckx, R., 1997. Concentration of <sup>137</sup>Cs and K in soil  
575 solution predict the plant availability of <sup>137</sup>Cs in soils. *Environ. Sci. Technol.* 31, 3432–

576 3438. doi:10.1021/es970113r

577 Smolders, E., Tsukada, H., 2011. The transfer of radiocesium from soil to plants: Mechanisms,  
578 data, and perspectives for potential countermeasures in Japan. *Integr. Environ. Assess.*  
579 *Manag.* 7, 379–81. doi:10.1002/ieam.236

580 Suzuki, Y., Yasutaka, T., Fujimura, S., Yabuki, T., Sato, M., Yoshioka, K., Inubushi, K., 2015.  
581 Effect of the concentration of radiocesium dissolved in irrigation water on the  
582 concentration of radiocesium in brown rice. *Soil Sci. Plant Nutr.* 61, 191–199.  
583 doi:10.1080/00380768.2014.1003192

584 Tensho, K., Yeh, K.-L., Mitsui, S., 1961. The uptake of strontium and cesium by plants from  
585 soil with special reference to the unusual cesium uptake by lowland rice and its  
586 mechanism. *Soil and Plant Food* 6, 176–183. doi:10.1080/00380768.1961.10430944

587 Tsuji, H., Yasutaka, T., Kawabe, Y., Onishi, T., Komai, T., 2014. Distribution of dissolved and  
588 particulate radiocesium concentrations along rivers and the relations between radiocesium  
589 concentration and deposition after the nuclear power plant accident in Fukushima. *Water*  
590 *Res.* 60, 15–27. doi:10.1016/j.watres.2014.04.024

591 Tsukada, H., Hasegawa, H., Hisamatsu, S., Yamasaki, S., 2002. Rice uptake and distributions  
592 of radioactive  $^{137}\text{Cs}$ , stable  $^{133}\text{Cs}$  and K from soil. *Environ. Pollut.* 117, 403–409.  
593 doi:10.1016/S0269-7491(01)00199-3

594 Uchida, S., Tagami, K., 2007. Soil-to-plant transfer factors of fallout  $^{137}\text{Cs}$  and native  $^{133}\text{Cs}$  in  
595 various crops collected in Japan. *J. Radioanal. Nucl. Chem.* 273, 205–210.  
596 doi:10.1007/s10967-007-0737-5

597 Verfaillie, G., Myttenaere, C., Bourdeau, P., 1967. Factors involved in the accumulation of  
598 fallout radionuclides in irrigated rice and meadow plants, in: Aberg, B., Hungate, F.P.  
599 (Eds.), *Radioecological Concentration Processes: Proceedings of an International*  
600 *Symposium Held in Stockholm, April, 1966.* Pergamon Press, NewYork, pp. 429–436.

601 Figure captions

602

603 Figure 1. Field experimental designs. CR, control; 0K, no K fertilizer application; 3BK, triple  
604 basal dressing of KCl; 3TK, triple top-dressing of KCl; KS, K<sub>2</sub>SiO<sub>3</sub> application instead of  
605 KCl; 0TK/IN, no K top-dressing in 2012 and increased N fertilizer application in 2013–2014.

606

607 Figure 2. <sup>137</sup>Cs concentrations in straw (a) and brown rice (b) and <sup>133</sup>Cs concentrations in  
608 straw (c) and brown rice (d). Fertilizer treatments: CR, control; 0K, no K fertilizer  
609 application; 3BK, triple basal dressing of KCl; 3TK, triple top-dressing of KCl; KS, K<sub>2</sub>SiO<sub>3</sub>  
610 application instead of KCl; 0TK, no K top-dressing; IN; increased N fertilizer application.  
611 Gray and white rectangle bars represent the averages between water management treatments  
612 for each fertilizer subsection and overall, respectively. Error bars in (a) and (b) represent  
613 counting errors. The short-flooding, 3TK value for brown rice <sup>137</sup>Cs (b) at KND in 2013 (2.03  
614 Bq kg<sup>-1</sup>) was an outlier and hence excluded from data.

615

616 Figure 3. Relationships between <sup>137</sup>Cs and <sup>133</sup>Cs concentrations in straw (upper panels) and  
617 brown rice (lower panels). The significances of the correlation coefficients (*r*) by *t*-test is  
618 indicated with asterisks: \*\**p* < 0.010, \*\*\**p* < 0.001. Regression lines are shown when  
619 significant correlation was observed at *p* < 0.050.

620

621 Figure 4. Exchangeable NH<sub>4</sub><sup>+</sup> (a) and K (b) contents in control (CR) and increased N fertilizer  
622 (IN) subsections at the YWR field for samples collected five times in 2014: the days before  
623 transplanting (15 May), before midsummer drainage (23 Jun), after midsummer drainage in  
624 the medial- and short-flooding sections (16 Jul), after heading stage (6 Aug), and after  
625 drainage in the long-flooding section (5 Sep). Rectangles and error bars respectively represent

626 the averages and standard deviations of triplicates.

Table 1. Schedule of paddy field management during the experiment

Field management	2012		2013				2014			
	YWR		YWR		KND		YWR		KND	
Basal dressing fertilizer application and soil mixing	1	May	7	May	13	May	7	May	12	May
Start of flooding and puddling	9		13		10–12 <sup>a</sup>		14		14	
Transplanting	11		15		15		16		19	
Starting midsummer drainage in MF and SF sections	29	June	24	June	24	June	25	June	25	June
Re-flooding in MF section	6	July	2	July	2	July	15	July	15	July
Re-flooding in SF section	13		9		10		15		15	
Top-dressing fertilizer application	18		18		18		22		25	
Drainage in SF section	20	August	15	August	15	August	15	August	15	August
Drainage in MF section	27		23		22		22		22	
Drainage in LF section	5	September	30		30		4	September	3	September
Harvesting	26		10	September	11	September	19		16	

<sup>a</sup> In the case of 2013 in KND field, flooding started before basal dressing although the exact starting date was unfortunately not recorded. LF, MF and SF in the table mean as long-, medial- and short-flooding, respectively.

Table 2. Annual mean of soil  $^{137}\text{Cs}$  and exchangeable K contents after harvest in experimental sections and subsections.

Sections or subsections		Soil $^{137}\text{Cs}$ (Bq kg $^{-1}$ )					Exchangeable K (mmol kg $^{-1}$ )				
		YWR			KND		YWR			KND	
		2012	2013	2014	2013	2014	2012	2013	2014	2013	2014
Water management	LF	160	136	167	167	173	4.7	4.4	4.6	2.5	2.9
	MF	164	135	154	148	183	4.8	5.3	5.2	2.7	2.9
	SF	174	147	161	147	169	4.8	5.5	5.8	2.7	3.0
Fertilizer	CR	164	133	155	151	152	4.9	5.4	5.3	2.5	2.7
	0K	157	156	161	138	185	4.4	4.5	4.3	2.5	2.2
	3BK	160	139	166	161	182	5.3	5.7	6.5	2.9	3.8
	3TK	171	122	153	166	181	4.8	4.7	4.7	2.6	3.0
	KS	174	138	177			4.4	5.0	5.1		
	0TK/IN	171	147	154			4.9	5.0	5.2		

Water management sections: LF, long flooding; MF, medial flooding; SF, short flooding. Fertilizer subsections; CR, control; 0K, no K fertilizer application; 3BK, triple basal dressing of KCl; 3TK, triple top-dressing of KCl; KS,  $\text{K}_2\text{SiO}_3$  application instead of KCl; 0TK/IN, no K top-dressing in 2012 and increased N fertilizer application in 2013 and 2014.

Table 3. Annual mean yields of straw and brown rice in experimental sections and subsections.

Sections or subsections		Straw yield (g m <sup>-2</sup> )					Brown rice yield (g m <sup>-2</sup> )				
		YWR			KND		YWR			KND	
		2012	2013	2014	2013	2014	2012	2013	2014	2013	2014
Water management	LF	685	792	664	551	694	438	460	388	417	423
	MF	661	801	759	556	682	432	471	470	406	401
	SF	685	817	809	510	717	463	468	479	393	427
Fertilizer	CR	651	814	732	552	708	437	477	431	409	418
	0K	681	786	720	562	700	443	466	451	411	418
	3BK	717	788	729	502	693	461	485	437	395	411
	3TK	608	762	680	540	689	402	429	425	407	421
	KS	743	834	811			482	497	485		
	0TK/IN	662	834	792			440	444	447		

Values are based on dry matter mass. Water management sections: LF, long flooding; MF, middle flooding; SF, short flooding. Fertilizer subsections; CR, control; 0K, no K fertilizer application; 3BK, triple basal dressing of KCl; 3TK, triple top-dressing of KCl; KS, K<sub>2</sub>SiO<sub>3</sub> application instead of KCl; 0TK/IN, no K top-dressing in 2012 and increased N fertilizer application in 2013 and 2014.



Table 4. Annual means of K concentrations in straw and brown rice in experimental sections and subsections.

Sections or subsections		Straw K (g kg <sup>-1</sup> )					Brown rice K (g kg <sup>-1</sup> )				
		YWR			KND		YWR			KND	
		2012	2013	2014	2013	2014	2012	2013	2014	2013	2014
Water management	LF	16.0	19.4	18.4	17.3	18.5	2.87	2.82	2.70	2.91	2.64
	MF	16.1	19.1	19.8	16.4	17.3	2.89	2.82	2.74	2.95	2.69
	SF	16.7	18.4	19.3	15.8	17.0	2.98	2.92	2.71	2.95	2.61
Fertilizer	CR	16.1	20.1	19.4	16.3	17.9	2.87	2.90	2.71	2.95	2.66
	0K	16.4	18.8	18.8	16.4	17.1	2.99	2.90	2.72	2.90	2.67
	3BK	16.3	19.4	19.5	16.6	16.9	2.78	2.99	2.70	2.98	2.60
	3TK	16.0	17.2	18.2	16.6	18.7	2.97	2.76	2.80	2.92	2.66
	KS	16.7	20.6	20.3			2.91	2.97	2.68		
	0TK/IN	16.2	17.9	18.7			2.98	2.59	2.68		

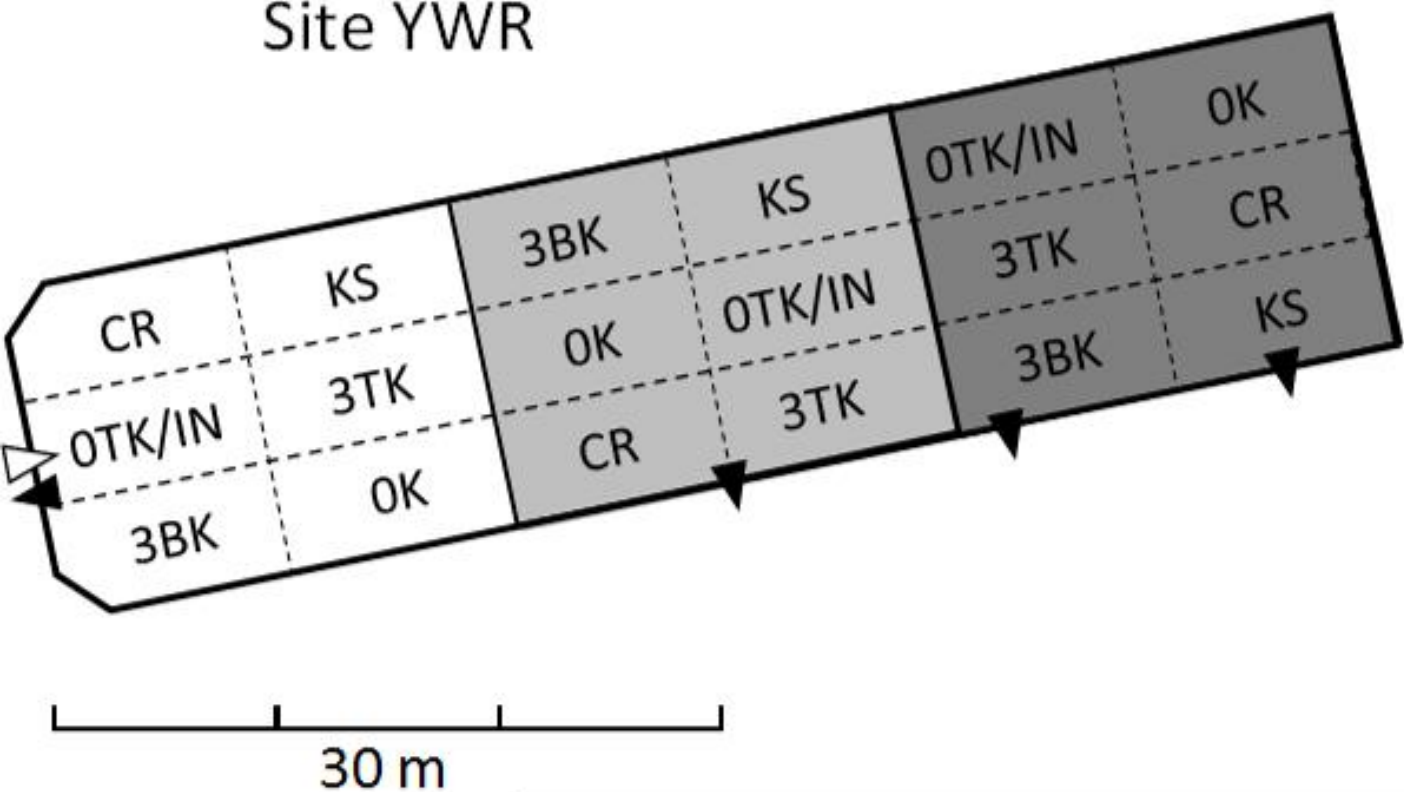
Water management sections: LF, long flooding; MF, middle flooding; SF, short flooding. Fertilizer subsections; CR, control; 0K, no K fertilizer application; 3BK, triple basal dressing of KCl; 3TK, triple top-dressing of KCl; KS, K<sub>2</sub>SiO<sub>3</sub> application instead of KCl; 0TK/IN, no K top-dressing in 2012 and increased N fertilizer application in 2013 and 2014.

Table 5. Analysis-of-variance table for <sup>137</sup>Cs activity and <sup>133</sup>Cs concentration in straw and brown rice with results of Tukey's post-hoc multiple comparisons.

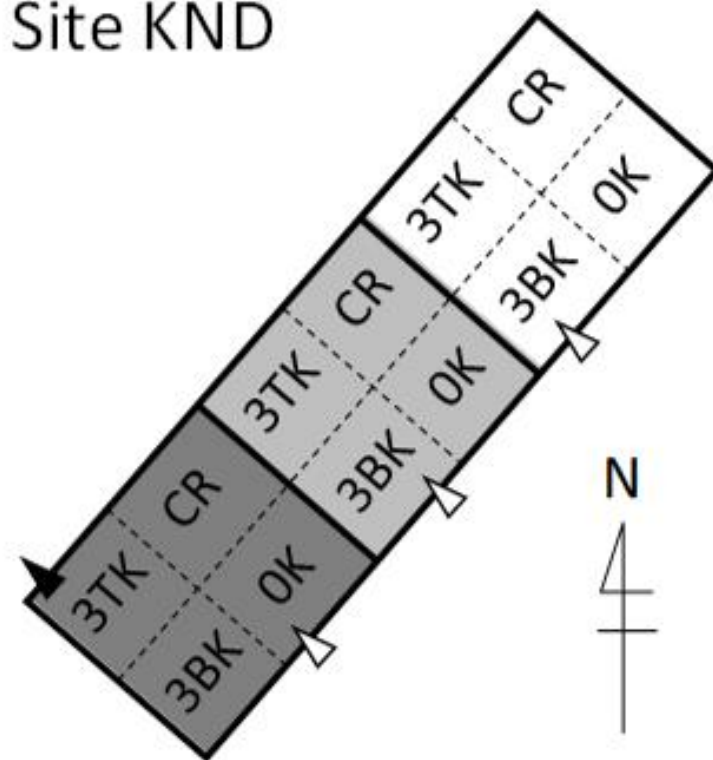
Factor	DF <sup>a</sup>	Error term	Straw <sup>137</sup> Cs			Brown rice <sup>137</sup> Cs			Straw <sup>133</sup> Cs			Brown rice <sup>133</sup> Cs		
			MS <sup>b</sup>	F	<i>p</i>	MS	F	<i>p</i>	MS	F	<i>p</i>	MS	F	<i>p</i>
<i>Fixed effects</i>														
Water management	2	s w	5.61	107.6	0.0092	0.171	2.0	0.3370	98.5	9.6	0.0942	10.2	34.9	0.0279
Fertilizer	3	s w f	0.75	3.0	0.0866	0.045	3.7	0.0572	10.1	3.3	0.0732	4.0	17.7	0.0004
Year	1	Residual	2.11	8.9	0.0114	0.079	6.0	0.0320	4.2	1.9	0.1958	14.2	37.2	0.0001
w f	6	s w f	0.13	0.5	0.7862	0.027	2.2	0.1369	5.0	1.6	0.2455	0.8	3.5	0.0443
w y	2	Residual	0.65	2.7	0.1058	0.057	4.3	0.0414	0.3	0.1	0.8810	1.8	4.7	0.0312
f y	3	Residual	0.41	1.7	0.2179	0.051	3.8	0.0399	10.3	4.6	0.0230	2.4	6.2	0.0087
w f y	6	Residual	0.18	0.8	0.6160	0.023	1.7	0.1946	3.6	1.6	0.2280	0.5	1.2	0.3716
<i>Random effects</i>														
Site	1	s w	0.57	10.8	0.0812	0.068	0.8	0.4672	91.8	9.0	0.0957	41.3	141.2	0.0070
s w	2	s w f	0.05	0.2	0.8154	0.085	7.1	0.0155	10.2	3.3	0.0835	0.3	1.3	0.3235
s w f	9	Residual	0.25	1.1	0.4564	0.012	0.9	0.5462	3.1	1.4	0.2945	0.2	0.6	0.7761
Residual	12		0.24			0.013			2.2			0.4		
Tukey's post-hoc multiple comparisons test	In 2013–2014		LF > MF, SF				-			-			-	
	In 2013		-				no significance				no significance			
	In 2014		-				0K > CR, 3BK, 3TK				0K, 3TK > 3BK			

<sup>a</sup> Degrees of freedom. <sup>b</sup> Mean square. Interactions between factors were shown as combinations of capital letter for each factor (w: water management, f: fertilizer, y: year, s: site). Data from four subsections (CR, 0K, 3BK and 3TK) in 2013–2014 were used for the analysis. When effects of water management or fertilizer were significant ( $p < 0.050$ ) without any interaction, Tukey's post-hoc multiple comparisons test was used to determine significant differences among water management sections (LF, long flooding; MF, middle flooding; SF, short flooding) or fertilizer subsections (CR, control; 0K, no K fertilizer; 3BK, triple basal dressing of KCl; 3TK, triple top-dressing of KCl) at  $p < 0.050$ . When significant interactions of year with water management or fertilizer were revealed by ANOVA, the post-hoc test was performed separately for 2013 and 2014 data at  $p < 0.025$ .

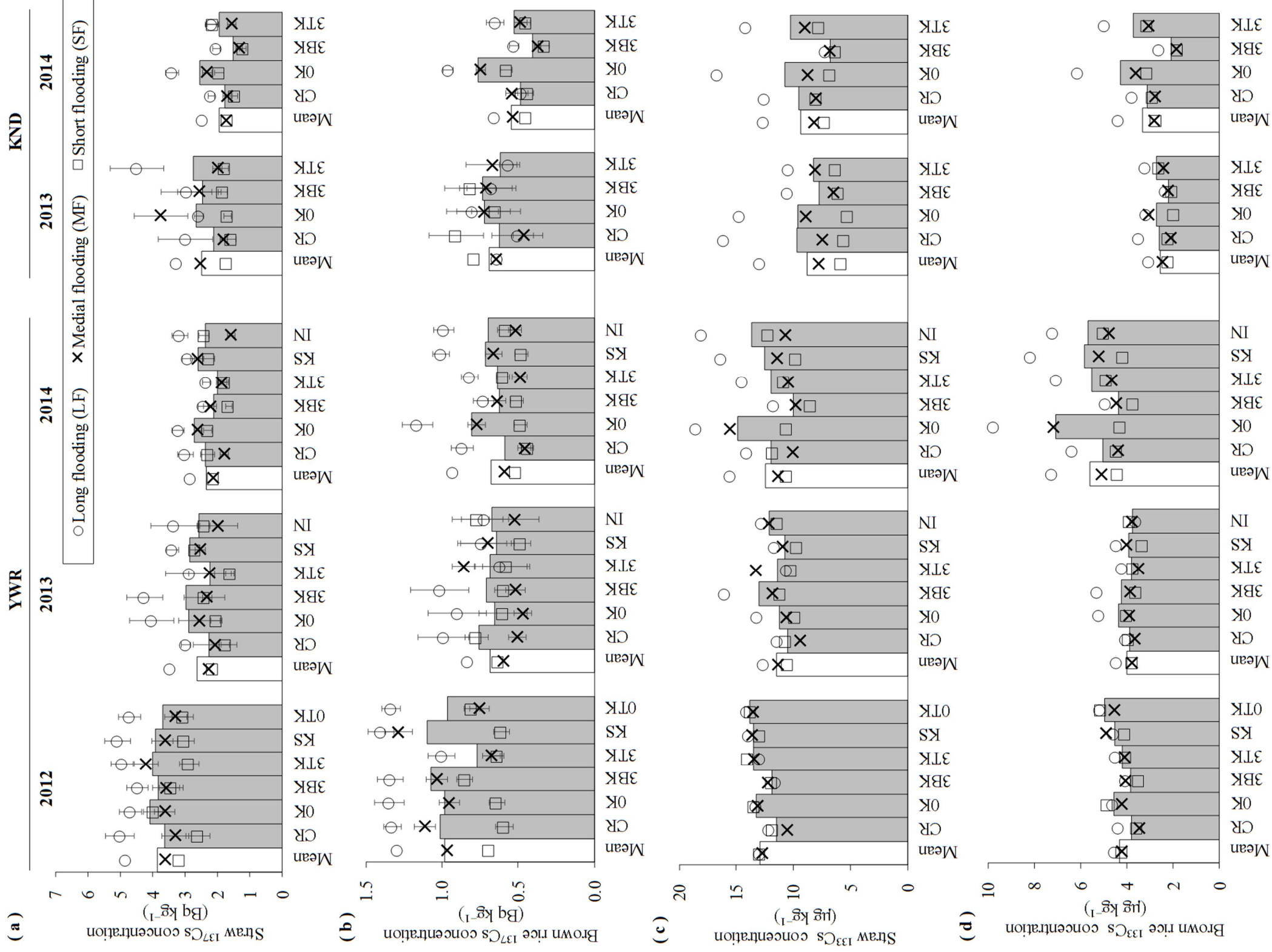
Site YWR



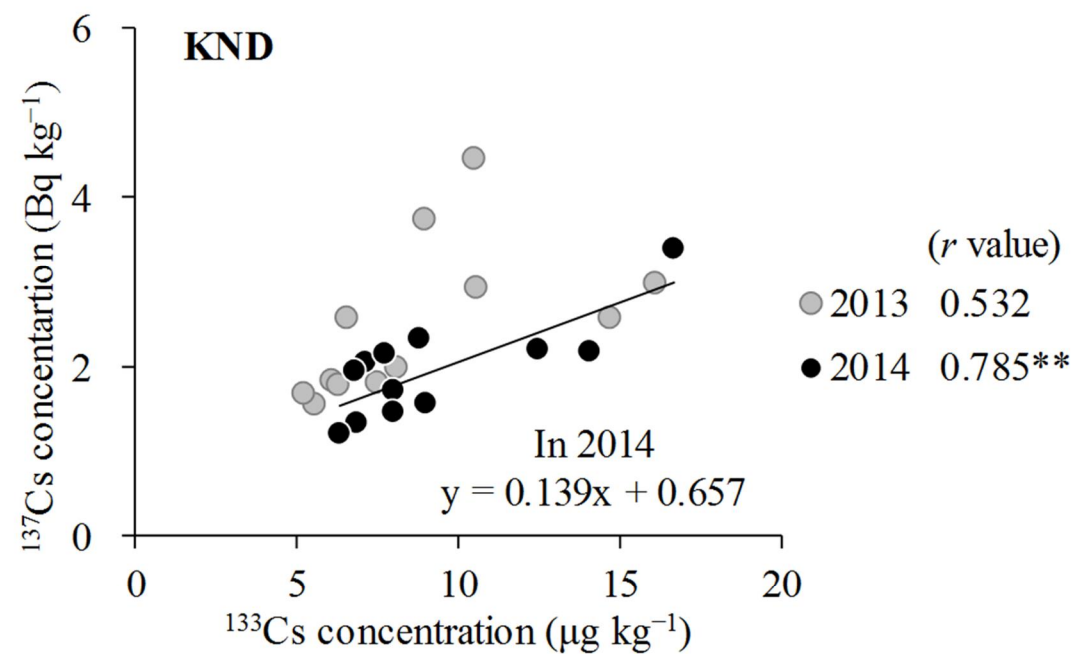
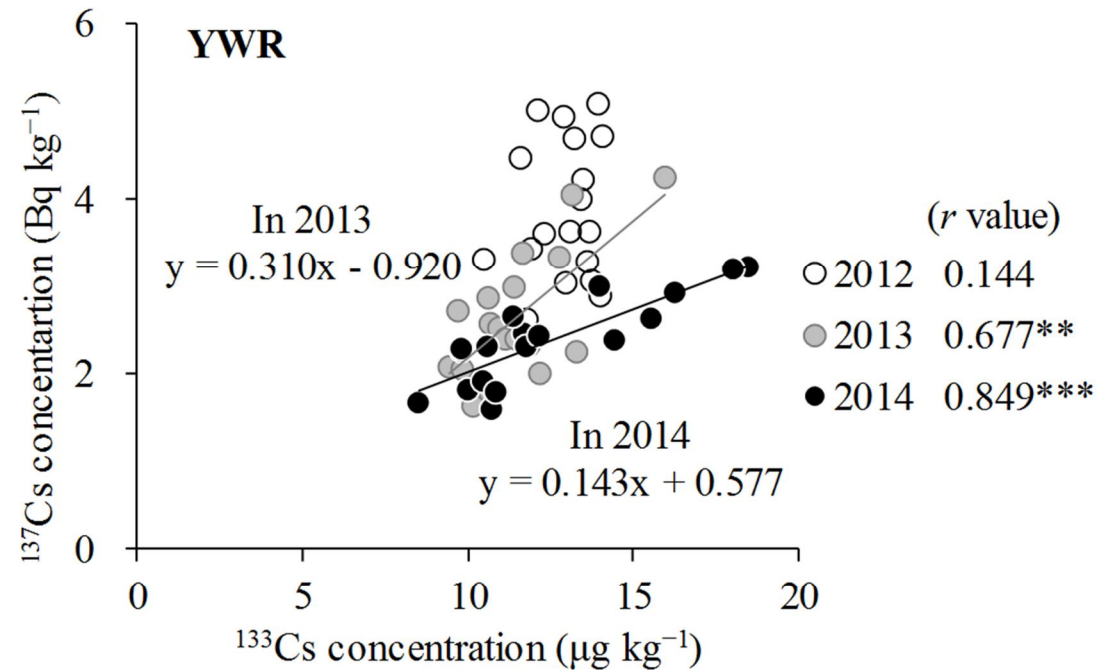
Site KND



	LF: long-flooding management		water inlet
	MF: medial-flooding management		outlet
	SF: short-flooding management		



( Straw )



( Brown rice )

