

Influence of water management and fertilizer application on 137Cs and 133Cs uptake in paddy rice fields

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1	Influence of Water Management and Fertilizer Application on ¹³⁷ Cs and ¹³³ 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 ¹³⁷Cs and ¹³³Cs uptakes by rice plant.

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

Uptakes of ¹³⁷Cs and ¹³³Cs by rice became correlated over the elapsed year.

1 Abstract

Cesium-137 derived from the Tokyo Electric Power Company's Fukushima Dai-ichi $\mathbf{2}$ Nuclear Power Plant (FDNPP) accident contaminated large areas of agricultural land in 3 Eastern Japan. Previous studies before the accident have indicated that flooding enhances 4 radiocesium uptake in rice fields. We investigated the influence of water management in $\mathbf{5}$ combination with fertilizers on ¹³⁷Cs concentrations in rice plants at two fields in southern 6 Ibaraki Prefecture. Stable Cs (133Cs) in the plants was also determined as an analogue for 7 predicting ¹³⁷Cs behavior after long-term aging of soil ¹³⁷Cs. The experimental periods 8 comprised 3 y starting from 2012 in one field, and 2 y from 2013 in another field. These fields 9 were divided into three water management sections: a long-flooding section without 10 midsummer drainage, and medial-flooding, and short-flooding sections with one- or 11 12two-week midsummer drainage and earlier end of flooding than the long-flooding section. Six or four types of fertilizer subsections (most differing only in potassium application) were 13 nested in each water management section. Generally, the long-flooding treatment led to higher 14¹³⁷Cs and ¹³³Cs concentrations in both straw and brown rice than medial- and short-flooding 15treatments, although there were some notable exceptions in the first experimental year at each 16site. Effects of differing potassium fertilizer treatments were cumulative; the effects on ¹³⁷Cs 17and ¹³³Cs concentrations in rice plants were not obvious in 2012 and 2013, but in 2014, these 18 concentrations were highest where potassium fertilizer had been absent and lowest where 19 basal dressings of K had been tripled. The relationship between ¹³⁷Cs and ¹³³Cs in rice plants 20was not correlative in the first experimental year at each site, but correlation became evident 21in the subsequent year(s). This study demonstrates a novel finding that omitting midsummer 22drainage and/or delaying drainage during the grain-filling period enhances uptake of both 23¹³⁷Cs and ¹³³Cs. 24

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

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28 **1. Introduction**

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

Rice (Oryza sativa) cropping in flooded soil is the representative agricultural system in 38 Japan. Some previous studies before the FDNPP accident indicate that soil flooding enhances 39 radiocesium uptake by the rice plant. Tensho et al. (1961) demonstrated using pot-culture 40 experiments with artificial addition of radiocesium that the uptake of radiocesium by rice 41 plants is much greater from flooded soil than from unflooded soil. They suggested that 42ammonium (NH4⁺) as the main form of inorganic nitrogen in flooded soil might enhance 43availability of radiocesium to plants because NH4⁺ could exchange with radiocesium 44 selectively adsorbed in soil. Pot-culture experiments by D'Souza and Mistry (1980) generated 45results similar to those of Tensho et al. (1961). Verfaillie et al. (1967) demonstrated in an 46 actual paddy field in Northern Italy in 1964 that preventing flooding decreased ¹³⁷Cs 47concentration in rice grain. Although these studies compared the effects of flooding and 48 upland cultivation practices on radiocesium uptake by rice plants, it remains unknown 49 whether variations in the flooding period within a practical range for paddy rice cropping 50

affect ¹³⁷Cs uptake by rice plants. Smolders and Tsukada (2011) suggested that water management systems that suppress NH₄⁺ concentration might be a potential countermeasure against ¹³⁷Cs transfer to rice crops, and advocated factorial experiments focused on water management and nitrogen (N) fertilization.

We conducted field experiments in paddy crops to investigate the influence of water 55management (flooding period) on ¹³⁷Cs concentration in rice plants. Additionally, the effects 56of different K application treatments were investigated. Stable cesium-133 (¹³³Cs) is regarded 57as a useful analogue for long-term assessment of ¹³⁷Cs in agricultural environments, because 5859 the fate of radionuclides in the environment follows the behavior of their stable isotopes (Tsukada et al., 2002; Uchida and Tagami, 2007). In this study, ¹³³Cs in rice plants was also 60 determined so the influence of water management and fertilizer treatment on ¹³⁷Cs uptake 61 long after the fallout event might be predicted. 62

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64 **2. Materials and Methods**

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66 **2.1. Experimental Fields**

The experimental area comprised two paddy fields in southern Ibaraki Prefecture. Site YWR 67 is located in the alluvial plain of the Kokaigawa River in Tsukubamirai City. Site KND is 68 located on the Hitachi tableland in Tsukuba City. According to the soil classification system in 69 Japan (Obara et al., 2011) or the World Reference Base for Soil Resources (International 70Union of Soil Sciences Working Group WRB, 2014) respectively, YWR soil is Gray Lowland 71soil or Glevic Fluvisol, and KND soil is Upland Reformed soil or Thaptandic Regosol 72(Transportic). These fields had no history of deep tillage after the FDNPP accident, therefore 73¹³⁷Cs was mainly distributed within topsoil. 74

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

76spatially uniform for more than a decade. A common practice in these areas, called "midsummer drainage," is aimed at enhancing root growth and reducing the number of 77unproductive tillers of rice plants. It also promotes subsurface drainage through formation of 7879 drying cracks in the soil and increases the soil's bearing capacity for machinery operation (Inoue and Tokunaga, 1995). In previous rice cropping at YWR, field flooding was started and 80 topsoil was puddled in early May, a few days before transplanting, as was customary for the 81 water management system. Basal-dressing fertilizer had been applied before flooding. In late 82 June, ponding water was temporarily drained for 2-3 weeks as midsummer drainage. Field 83 84 flooding ended in early September, a week before harvest. As was customary at KND, field flooding was started and topsoil was puddled in early May. A few days later, basal-dressing 85 fertilizer was applied to the flooded soil. Rice seedlings were transplanted a few days later 86 87 thereafter. The midsummer drainage was conducted for a week from early July. During drainage period in KND, drainpipe buried in the soil at a depth of 90 cm was opened to 88 promote underdrainage. Field flooding ended at the end of August, two weeks before harvest. 89 90 (Other details on field management before this experiment, such as fertilizer application, are summarized in Supplementary Tables.) 91

The experimental periods comprised 3 y from 2012 at the YWR field, and 2 y from 2013 at the KND field. The cultivar "Koshihikari" was transplanted in May and raised until harvest in September. The schedule of paddy field management during the experiment is shown in Table 1. During the season, air temperature ranged from 4.9 to 36.8°C (mean temperature of 23.4°C) and precipitation was recorded as 580, 380 and 760 mm in 2012, 2013 and 2014, 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

101three sections for different water management treatments using two plastic corrugated sheets (Nami-ita in Japanese). Each section was identified by the three type of water management as 102103 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 midsummer for 7 or 8 d and 7-12 d earlier at the end of flooding than the LF section; and a 105short-flooding (SF) section, which was drained in midsummer for 14-16 d and 15-20 d 106 earlier at the end of flooding than the LF section. In 2014, however, midsummer drainage for 107the MF and SF sections was increased to 20 d because of prolonged rainy weather. At KND, 108 109 only one water outlet existed for surface drainage; therefore, pumps were used to discharge standing water at that site. 110

111 Each water management section was divided into six (at YWR) or four (at KND) subsections for different fertilizer treatments. Control subsections (CR) at each site received 1123.0 g m⁻² of N as ammonium sulfate ((NH₄)₂SO₄), 2.6 g m⁻² of P as calcium superphosphate 113(mainly Ca(H₂PO₄)₂·H₂O), and 5.0 g m⁻² of K as potassium chloride (KCl) as basal dressing, 114and 3.0 g m⁻² of N as $(NH_4)_2SO_4$ and 2.5 g m⁻² of K as KCl as top-dressing in late July. Three 115types of subsections at each site were defined according to the KCl application strategy; no K 116fertilizer application (0K: no K), triple basal dressing without top-dressing (3BK: totally 15 g 117 m⁻² K), and triple top-dressing without basal dressing (3TK: totally 7.5 g K m⁻² K). Of the 118remaining subsections at YWR, one type received 7.5 g m^{-2} of K as potassium silicate 119 (K₂SiO₃), which is a release fertilizer, as basal dressing instead of KCl (KS). The other type 120 121differs from the control subsections in having had no top-dressing of KCl (0TK) in 2012 and increased N fertilizer application in 2013 and 2014 (IN: 4 g m⁻² of N as (NH₄)₂SO₄ and the 122same amount of N as controlled-release coated urea, LPSS100) as basal dressing. 123

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.

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

In mid-September, the aerial parts of matured rice were harvested at about 2 cm above ground from a 5.0 or 6.5 m² area of each subsection. A sample of the underlying topsoil, which was 17 cm thick on average, was also collected from a harvest area of each subsection with a shovel or a root auger (4 cm diameter; DIK-102A, Daiki Rika Kogyo Co., Ltd., Japan). Brown rice samples were obtained after air-drying, husking, and sieving through 1.8 mm sieves. Straw samples were dried at 70 °C, brushed to remove surface tissue stained with soil remnants, and milled. Soil samples were air-dried and sieved through 2.0 mm sieves.

Concentrations of ¹³⁷Cs in straw, brown rice, and soil samples were determined with 136 germanium (Ge) gamma-ray detectors (GEM55P, GEM20-70, SEIKO EG&G, Co., Ltd., 137Japan; GC2020, GC2520, GC4020-7500SL-2002SCL, Canberra, USA) using 2.0 L of plant 138samples or 100 mL of soil samples. Before measurement, each sample was mixed to 139140 homogenize the material and was then uniformly packed into a plastic container. The counting efficiency of the Ge gamma-ray detectors was calibrated using gamma-ray reference source 141 (MX033MR and MX033U8PP, Japan Radioisotope Association, Tokyo, Japan). The decay 142corrections were made to the harvest day in each year. 143

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

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

To investigate the influence of water management and increasing N fertilizer application on 158soil NH4⁺ concentration, the exchangeable NH4⁺ content of soils in the CR and IN subsections 159was determined before and during cultivation in 2014 at the YWR field. Soil cores (3 cm 160 161 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 the MF and SF sections (16 July), after heading stage (6 August), and after drainage in the LF 163section (5 September). Each wet core sample was stirred to homogenize, and a part of it was 164used for 1-h extraction with 2 mol L^{-1} KCl solution (solution/soil ratio of 10 mL g^{-1}) within 165the sampling day. After centrifugation and filtration, the NH4⁺ concentration of the 166 supernatant solution was determined using an Autoanalyzer (QuAAtro 2-HR, BL-TEC, Japan). 167The remaining wet samples were used to estimate exchangeable K contents during cultivation. 168 Similar to KCl extractions, 1-h extractions with 1 mol L⁻¹ ammonium acetate solution were 169 conducted. The K concentration of the supernatant solutions was determined using atomic 170171absorption spectrometry.

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

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175 **2.4. Statistical analysis**

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

The data of four subsections (CR, 0K, 3BK, and 3TK) for 2013-2014, which have 181counterparts in all sections of both fields, were subjected to mixed model analysis of variance 182(ANOVA) with the SAS Add-In for Microsoft office version 6.1 M1 (SAS Institute Inc., 183 USA). This dataset is structured as a split-plot design with site as the block, water 184 management as the primary factor, fertilizer as the secondary factor, and experimental year as 185the tertiary factor. In this paper, the statistical term "significant" refers to p < 0.050 and 186"significant tendency" to 0.050 . When the effects of water management or187fertilizer were significant without any interaction in ANOVA, Tukey's post hoc multiple 188comparison test was performed to determine significant difference among sections or 189 190 subsections. When significant interactions between experimental year and water management or fertilizer were revealed by ANOVA, a post-hoc Tukey's test was performed separately for 191both 2013 and 2014 datasets at an adjusted significance level (p < 0.050 / 2 = 0.025). 192

Correlations between ¹³⁷Cs and ¹³³Cs concentrations in straw and brown rice were calculated
by linear regression using Microsoft Excel 2013.

195

196 **3. Results**

The soil ¹³⁷Cs concentrations were closely similar in the two fields (Table 2). For each field in each year, the average soil ¹³⁷Cs concentration for each type of water management or fertilizer treatment ranged within a narrow interval (for example, 153–177 Bq kg⁻¹ at YWR and 152–185 Bq kg⁻¹ at KND in 2014). The ANOVA results for the 2013–2014 dataset

revealed no significant differences in ¹³⁷Cs among soils that experienced different water 201management or fertilizer treatment, but there were significant differences for the different 202experimental years. The values were lower in 2013 than in other years, which could be 203204 attributed to a sampling error caused by using a different tool in 2013 (root auger) than in the other years (shovels). When root augers were vertically inserted into the soil during sampling, 205the penetration resistance appeared to be lower than when a shovel was used, indicating that 206the sampling with the auger may have been deeper. This could have allowed accidental 207 208inclusion 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 values for 0K and 3BK were the lowest and the highest, respectively, among the values for 210211different fertilizer treatments, corresponding to their ranking in respect to K application 212amounts. The average exchangeable K content for 3BK increased annually in each field, in contrast to stable or decreasing values for 0K, so the differences widened over years. Water 213214management sections exhibited small exchangeable K differences at KND. At YWR, the 215difference was negligible in 2012, but in the subsequent years, the exchangeable K content was highest in the soil that had experienced longer drainage treatment (SF). The ANOVA 216 results for the 2013-2014 dataset revealed significant fixed effects of fertilizer and 217experimental year and their interaction (data not shown). 218

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

The K concentration in straw and brown rice varied more between experimental years than between experimental treatments (Table 4). Water management practices and fertilizer treatments did not cause the value to vary by more than 10% from the grand mean in each year at each field. The ANOVA results for the 2013–2014 dataset revealed a significant difference only between experimental years (data not shown).

The ¹³⁷Cs and ¹³³Cs concentrations found in straw and brown rice are presented in Fig. 2. The ANOVA results for the 2013–2014 dataset are summarized in Table 5.

Concentration of ¹³⁷Cs in straw was higher for LF than for MF and SF in each case, except 231for 0K at KND in 2013 (Fig. 2a). The annual mean values for each water management are 232shown in Fig. 2 together with the annual grand mean (white bars in Fig. 2). Compared to MF, 233the annual mean values for LF were higher by 28%-52%, while those of SF were almost the 234same, except in each field's first experimental year when annual mean values for SF were 235lower by 12% (YWR) and 32% (KND). Among the different fertilizer treatments, the mean 236237values in 2014 were highest for 0K at both fields and lowest for in 3BK at KND. The values in each section and subsection mostly decreased from year to year. The ANOVA results for the 2382013–2014 dataset showed significant influences of water management and year without any 239240interaction (Table 5). The Tukey's post hoc test indicated that the differences between LF and the other two kinds of water management were significant. 241

Concentration of ¹³⁷Cs in brown rice was also higher for LF than for MF and SF in most 242cases at YWR and in most 2014 cases at KND (Fig. 2b). At YWR, the annual mean values for 243LF were higher by 33%–57% than those for MF. The mean value for SF was lower than that 244245for MF by 29% in 2012, but in 2013 and 2014 the mean values for SF and MF were almost equal. At KND in 2013, the annual mean value for brown rice ¹³⁷Cs was higher for SF than 246 for LF and MF, contrary to the straw ¹³⁷Cs results. Among the different fertilizer treatments, 247the mean values in 2014 were highest for 0K at both fields and lowest for 3BK at KND, in 248accordance with the straw results. The annual grand mean (white bars in Fig. 2) for each field 249decreased from the first experimental year to the second year. The ANOVA results for the 250

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 ¹³⁷Cs in brown rice was significantly higher for 0K than for three 256 other fertilizer treatments in 2014.

Concentration of ¹³³Cs in straw was higher for LF than for MF and SF in most cases after 2572012 (Fig. 2c). At YWR, the differences in annual mean values for the different types of water 258259management were negligible in 2012 but increased from year to year. Among the different fertilizer treatments, the values in 2014 were highest for 0K and lowest for 3BK at both fields. 260The ANOVA results for the 2013–2014 dataset showed significant tendencies for the effects 261262of water management and fertilizer along with a significant interaction between fertilizer and year (Table 5). The separate Tukey's post hoc test for each year detected no significant 263difference for 2013, but it showed that ¹³³Cs concentration in straw was significantly lower for 2642653BK than for 0K and 3TK in 2014.

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

275 The relationship between ¹³³Cs and ¹³⁷Cs concentrations in each plant component (straw and

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

The exchangeable NH₄⁺ and K contents of soil from selected subsections before and during 279rice cultivation in 2014 at YWR are exhibited in Fig. 4. Exchangeable NH4⁺ content was 280higher for IN than for CR on the day before transplanting (Fig. 4a). After that, the 281exchangeable NH4⁺ content became consistently low, regardless of the difference in 282application amount of N fertilizer. Midsummer drainage in the MF and SF sections did not 283change the values. During the grain-filling period, the NH4⁺ content increased with no clearly 284evident difference between different types of water management or different fertilizer 285treatments. Exchangeable K content decreased from before transplanting to after the heading 286287 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**

Except the case of brown rice in 2013 at KND, the ¹³⁷Cs concentrations of straw and brown 291rice showed highly significant correlation (YWR in 2012–2014, r=0.786, p<0.001; KND in 2922014, r=0.900, p < 0.001) and average ¹³⁷Cs concentrations for water management sections 293were ordered in the same way in the sense that the highest ¹³⁷Cs concentrations were in the LF 294sections (Fig. 2a, b). The average ¹³³Cs concentrations of straw and brown rice for the water 295management sections were in the same order as the average ¹³⁷Cs concentrations (highest in 296 the LF sections) except at YWR in 2012 (Fig. 2c, d). In these field experiments, the soil ¹³⁷Cs 297298concentrations of the water management sections in the two fields were in about the same range (Table 2). The exchangeable K content was also closely similar among the sections of 299each field at the end of the first experimental year. Therefore, the relatively high ¹³⁷Cs 300

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

312Possible causes of the LF effect on Cs uptake are discussed hereafter. Tensho et al. (1961) suggested that NH4⁺ exchanges with radiocesium selectively absorbed in soil particles to 313enhance its availability to the plant under submerged conditions. Ammonium also has been 314 315reported to play an important role in radiocesium release from submerged sediments (Comans et al., 1989; Evans et al., 1983); also, liberal application of NH₄⁺ enhances plant uptake of 316 radiocesium (Jackson et al., 1965; Lasat et al., 1997; Ohmori et al., 2014; Prister et al., 1992). 317 It should be investigated whether or not NH₄⁺ derived from mineralization or a practical 318 amount of fertilizer does contribute to Cs mobility, and whether oxidation by drainage 319 treatment could suppress this NH₄⁺ contribution. In this study, application of 1.3 times more N 320to the IN subsections than to the control subsections produced no appreciable difference in Cs 321concentration in the plants (Fig. 2). The differences in exchangeable NH_4^+ content among 322323soils that experienced different water management treatments were small in the observation at YWR in 2014 (Fig. 4). Ammonium is not necessarily considered to be the cause of the LF 324325effect at this moment.

Secondarily, the possibility of exogenous ¹³⁷Cs entry from irrigation water might be 326 considered. However, Suzuki et al. (2015) found that irrigation with water containing 327 dissolved ¹³⁷Cs at a concentration of 0.10 Bq L^{-1} did not or did only slightly (by less than 32820%) increase the ¹³⁷Cs concentration of brown rice grown in pot culture using soil containing 329 ¹³⁷Cs at 200 Bq kg⁻¹. Although ¹³⁷Cs concentrations of irrigation water were not measured in 330our experiment, they were assumed to be lower than $0.10 \text{ Bg } \text{L}^{-1}$. In the investigation of Tsuji 331et al. (2014) in four rivers located in Fukushima Prefecture in 2012 and 2013, dissolved ¹³⁷Cs 332concentrations in river water were less than 0.20 Bg L^{-1} in the river where deposited ¹³⁷Cs is 333 highest, and these were less than 0.05 Bq L^{-1} in the other three revers. Additionally, the 334differences in total duration of flooding among the water management sections were four 335 weeks or less in this study. Thus the differences of ¹³⁷Cs inflow load among the water 336 management sections are considered not to be a main cause of the LF effect. Similar 337 enhancements of radiocesium uptake by flooding were reported in pot experiments with no 338exogenous radiocesium entry (D'Souza and Mistry, 1980; Tensho et al., 1961). 339

340 Thirdly, D'Souza and Mistry (1980) suggested that shoot-base absorption of radiocesium in standing water can be a major means by which flooding increases radiocesium uptake, in 341 reference to a report by Myttenaere (1972) that absorption of radiocesium by rice plants is 342greater through a shoot base dipped in water than through a root dipped in nutrient solution. 343 However, the nutrient solution in the hydroponic experiment seems different from natural soil 344345solution in the point of salt concentration. To estimate the realistic contribution of shoot-base radiocesium absorption to rice plants, the composition of the test solutions (K, NH₄⁺, ¹³³Cs 346 etc.) should be modified according to that of soil solution and standing water in the actual 347 field. 348

A fourth possibility is that K availability can be changed with longer flooding to influence Cs uptake. Reductive conditions reportedly decrease K release (Chen et al., 1987; Horikawa and Kawaguchi, 1963), and oxidation of paddy soil by drainage decreases K-deficient symptoms in rice plants (Ogihara, 1960). At YWR in 2014, the exchangeable K content increased after the end of flooding where drainage had started earlier (Fig. 4b). This increase may have been caused by soil oxidation and related to lower Cs uptake in the SF section. Other causes are possible, but no clear mechanism for the LF effect is evident yet.

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

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

383

4.2. Influence of K fertilizer application

The effect of K fertilizer is significant or has a significant tendency for brown rice and straw 385386 ¹³³Cs, and there are significant interactions with year in all cases (Table 5). In 2014, plants under 0K treatments had the highest Cs concentrations in both straw and brown rice of all 387 fertilizer treatments (Fig. 2), and Tukey's post-hoc test indicated some significant differences 388389 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 treatment significantly changed K concentration in the rice plants (Table 4). These results 391indicate that K availability was sufficient for rice plant requirements in all subsections, but its 392 variation within this range affected Cs uptake by rice plant. Increase of the K basal dressing 393 would more effectively enhance K availability at KND than YWR because the former site was 394 poorer in exchangeable K (Table 2). However, the order of Cs uptake for different K 395 application treatments (3BK < other subsections < 0K) was not observed before 2013 even at 396 KND. The temporally broadening range of exchangeable K content across the subsections 397 suggests that K application cumulatively affected soil K availability over the years of the 398 experiment (Table 2). This accumulation would explain the changes in response to K fertilizer 399 treatments. Although application timing of K fertilizer (i.e., basal dressing or top-dressing) is 400

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

405

406 **4.3. Influence of time**

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

The ¹³⁷Cs and ¹³³Cs concentrations in rice plants over the elapsed years became correlated 414 during the experimental period (Fig. 3). The observation that ¹³⁷Cs concentrations gradually 415began to follow those of ¹³³Cs over time can be explained by the aging of soil ¹³⁷Cs. As shown 416 in Fig. 3, however, the variation range of stable ¹³³Cs becomes wider to fit that of ¹³⁷Cs, 417 except in the case of straw at KND. The widening variation of ¹³³Cs was caused by the 418 repetition of water management methods and fertilizer treatments. Repeated LF treatments 419 increased ¹³³Cs uptake by rice plant, which might be attributed to concentration of root 420activity within topsoil (Section 4.1). Repeated K fertilizer treatments cumulatively affected K 421availability, and resulted in a significant difference in ¹³³Cs uptake by rice plants between the 4220K and 3BK treatments (Section 4.2). Although ¹³³Cs uptake from subsoil might reflect K 423availability not in topsoil but in subsoil, repeated K fertilization might promote leaching from 424topsoil, and thereby enhance K availability in subsoil. The convergence of ¹³⁷Cs and ¹³³Cs 425

- 426 concentrations over the elapsed years can be attributed to both aging of soil ¹³⁷Cs 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 ¹³⁷Cs transfer to rice

The enhancement of ¹³⁷Cs uptake by the LF treatment was observed in both of the 431experimental fields, which had differing K status, site location, and soil taxa. The similar 432enhancement of natural ¹³³Cs concentration in rice plants after 2012 implies that LF treatment 433could be influential on ¹³⁷Cs transfer to rice plants even after long-term aging of soil ¹³⁷Cs, 434but the absence of an LF effect on ¹³³Cs in 2012 needs explanation. That is, the changing 435water management from LF to MF could be an effective, long-term method to reduce ¹³⁷Cs 436 437 transfer to rice. On the other hand, extending the drainage period in midsummer and grain-filling period to more than that of MF appears to be fruitless. In rice cropping systems 438in Japan, midsummer drainage is commonly practiced, and the total flooding period is shorter 439440 than that in the LF section of this study. Therefore, changing the practice of LF might be applicable only in fields where drainage treatment is skipped or imperfectly practiced due to 441 low water permeability, inflow of mountain runoff, or other reasons. In such fields, efforts to 442practice midsummer drainage and/or to terminate flooding earlier in the grain-filling period 443 could decrease ¹³⁷Cs transfer to rice. However, the effects of these two practices were not 444 individually investigated in this study. Additionally, the cause and the extent of generalization 445of the phenomenon should be examined in subsequent studies. 446

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

when exchangeable K content was higher than 3.8 mmol kg⁻¹. In previous experiments to 451investigate the effect of K fertilizer on radiocesium uptake by rice plants, exchangeable K 452contents in used soil were often low (for example, <1 mmol kg⁻¹, Fujimura et al. (2014) and 453Saito et al. (2012)). It is unclear whether K fertilization is effective in reducing Cs uptake by 454rice plants in fields where exchangeable K content is moderate or rich. The exchangeable K 455content in the first year was, on average, 4.7 mmol kg^{-1} at YWR and 2.6 mmol kg^{-1} at KND. 456This experiment shows that regular application of high amount of K is effective in reducing 457Cs uptake by rice plants, even where exchangeable K content is not low and the first K 458459fertilization seems to have no effect.

460

461 **5. Conclusion**

462 The rice cultivation experiments in two fields in southern Ibaraki Prefecture exhibited the463 following results.

1. Cs uptake in rice plants was significantly affected in many cases by water management treatments; omitting midsummer drainage and/or delaying drainage during the grain filling period (LF) mostly enhanced Cs concentration. The phenomenon was observed to be 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 137 Cs and 133 Cs uptakes was insignificant at first and became close

474 and significant with elapsed years. The convergence between ¹³⁷Cs and ¹³³Cs is attributed to

the aging of soil ¹³⁷Cs and the cumulative effects of repeated water management methods and

476 fertilizer treatments.

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

481

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Figure 1. Field experimental designs. CR, control; 0K, no K fertilizer application; 3BK, triple
basal dressing of KCl; 3TK, triple top-dressing of KCl; KS, K₂SiO₃ application instead of
KCl; 0TK/IN, no K top-dressing in 2012 and increased N fertilizer application in 2013–2014.

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Figure 2. ¹³⁷Cs concentrations in straw (a) and brown rice (b) and ¹³³Cs concentrations in 607 straw (c) and brown rice (d). Fertilizer treatments: CR, control; 0K, no K fertilizer 608 609 application; 3BK, triple basal dressing of KCl; 3TK, triple top-dressing of KCl; KS, K₂SiO₃ application instead of KCl; 0TK, no K top-dressing; IN; increased N fertilizer application. 610 611 Gray and white rectangle bars represent the averages between water management treatments for each fertilizer subsection and overall, respectively. Error bars in (a) and (b) represent 612 counting errors. The short-flooding, 3TK value for brown rice ¹³⁷Cs (b) at KND in 2013 (2.03 613 Bq kg^{-1}) was an outlier and hence excluded from data. 614

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

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Figure 4. Exchangeable NH₄⁺ (a) and K (b) contents in control (CR) and increased N fertilizer (IN) subsections at the YWR field for samples collected five times in 2014: the days before transplanting (15 May), before midsummer drainage (23 Jun), after midsummer drainage in the medial- and short-flooding sections (16 Jul), after heading stage (6 Aug), and after drainage in the long-flooding section (5 Sep). Rectangles and error bars respectively represent 626 the averages and standard deviations of triplicates.

Table	1.	Schedul	e of	paddy	field	managemen	t during	the	experin	nent

		2012	2013				_			
Field management		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 ^a		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	Septembe	er 11	September	19		16	

^a 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.

			Soil	¹³⁷ Cs (B	$q kg^{-1}$)		Exchangeable K (mmol kg ⁻¹)						
Sections		YWR			KN	JD		YWR	KND				
or subsections		2012	2013	2014	2013	2014	2012	2013	2014	2013	2014		
Water	LF	160	136	167	167	173	4.7	4.4	4.6	2.5	2.9		
management	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				

Table 2. Annual mean of soil ¹³⁷Cs and exchangeable K contents after harvest in experimental sections and subsections.

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, K₂SiO₃ application instead of KCl; 0TK/IN, no K top-dressing in 2012 and increased N fertilizer application in 2013 and 2014.

			Stra	w yield ($(g m^{-2})$		Brown rice yield (g m^{-2})						
Sections			YWR		KN	١D		YWR	KI	KND			
or subsections		2012	2013	2014	2013	2014	2012	2013	2014	2013	2014		
Water	LF	685	792	664	551	694	438	460	388	417	423		
management	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				

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

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, K₂SiO₃ application instead of KCl; 0TK/IN, no K top-dressing in 2012 and increased N fertilizer application in 2013 and 2014.

		Straw K (g kg ⁻¹)					Brown rice K (g kg ⁻¹)						
Sections		YWR			KN	ID		YWR		KND			
or subsections		2012	2013	2014	2013	2014	2012	2013	2014	2013	2014		
Water	LF	16.0	19.4	18.4	17.3	18.5	2.87	2.82	2.70	2.91	2.64		
management	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				

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

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, K2SiO3 application instead of KCl; 0TK/IN, no K top-dressing in 2012 and increased N fertilizer application in 2013 and 2014.

Factor	DEa	Error		Straw ¹²	³⁷ Cs	Brow	vn rice	¹³⁷ Cs	St	raw ¹³³	Cs	Bro	wn rice	¹³³ Cs
	DF	term	MS^{b}	F	р	MS	F	р	MS	F	р	MS	F	р
Fixed effects														
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
fy	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
Random effects														
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	In 2	013–2014	L	F > MF,	SF		-			-			-	
multiple	Iı	n 2013		-		no s	significa	ance	no s	ignifica	ance		-	
comparisons test	Iı	n 2014		-		0K > CR, 3BK, 3TK		0K, 3TK > 3BK			-			

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

^a Degrees of freedom. ^b 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.





