Radiocaesium bioavailability to flooded paddy rice is related to soil solution radiocaesium and potassium concentrations

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Abstract

Aims This study was set up to better understand the mechanisms of the variable radiocaesium bioavailability observed in flooded rice.

Methods Paddy topsoils ($n=9$) with contrasting soil properties were collected from the Fukushima-accident affected area, spiked with ^{134}Cs and cropped in the greenhouse with rice under flooded conditions. Soil solution was collected. As a reference to earlier work, we cropped the same paddy soils and some additional grassland soils ($n=22$ in total) with ryegrass under unsaturated conditions.

Results The rice shoot ^{134}Cs concentrations varied 110-fold among soils and were unrelated to soil solution ^{134}Cs concentrations. In contrast, the rice shoot ^{134}Cs concentrations clearly increased with the soil solution $^{134}\text{Cs}:\text{K}$ concentration ratio ($R^2=0.79$; $P<0.001$). The same trend was true for ryegrass shoot ^{134}Cs concentrations, which were, on average, 4.1-fold larger than those for flooded rice at an equal bulk soil solution $^{134}\text{Cs}:\text{K}$ ratio.

Conclusions Soil solution radiocaesium and K concentrations explain the radiocaesium availability to flooded rice, confirming established knowledge for ryegrass. Transport modelling suggests that the overall smaller ^{134}Cs bioavailability to rice in flooded soils than to ryegrass in unsaturated soils is related to the smaller K uptake rate of rice, yielding less K depletion in its rhizosphere with local higher K^+ , blocking the RCs^+ uptake by the roots.

Key words Soil-to-plant radiocaesium transfer; Flooded paddy rice; Fukushima accident affected area; Plant root uptake; Solid-liquid distribution; Soil solution

Introduction

The Fukushima accident in 2011 renewed our interest in the transfer of radiocaesium (RCs, ^{137}Cs and ^{134}Cs) in the food chain (Yamaguchi et al. 2016). Rice is a main staple food crop grown in the Fukushima accident affected area and is typically cultivated in intermittently flooded soils. The soil-to-plant transfer of RCs is traditionally expressed in terms of the transfer factor (TF) of RCs (WHO 2012), defined on dry weight (DW) basis (Uchida et al. 2009);

$$\text{TF} = \frac{c_{\text{plant}}}{c_{\text{soil}}} \quad [1]$$

in which c_{plant} (Bq kg^{-1} DW) is the RCs concentration in the plant and c_{soil} (Bq kg^{-1} DW) indicates the RCs concentration in the soil.

A large number of TFs of RCs for rice grown in Japan has been collected since the nuclear weapon testing and TFs range between 3.5×10^{-4} – 4.1×10^{-2} (IAEA 2009; Uchida et al. 2009). This range reflects the variable bioavailability of RCs among soils, rice varieties, climates, cropping years and land management (IAEA 2009). Soil properties such as clay content or pH only poorly explain the TF of RCs. The mechanisms of RCs bioavailability to cereal grains in grassland with aerobic soils are relatively well known and follow from the observation that crop RCs concentrations are related to the RCs:potassium (K) concentration ratio in the soil solution (Smolders et al. 1997; Absalom et al. 2001). This finding is because the non-essential trace element Cs is the analogue of the macronutrient K. Root uptake of RCs from solution largely increases when the solution K concentration decreases (Shaw et al. 1992). In soils, the role of K supply is more complex: crop RCs uptake from soil is a net effect of two subsequent reactions (Absalom et al. 2001; Uematsu et al. 2016);

where K_D (L kg^{-1}) is the solid-liquid distribution coefficient, defined with the RCs concentrations in the soil solid phase c_{solid} (Bq kg^{-1} DW) and in the soil solution c_{solution} (Bq L^{-1}) formulated as,

$$K_D = \frac{c_{\text{solid}}}{c_{\text{solution}}} \quad [3]$$

Potassium in soils plays a double but counteracting role on the RCs bioavailability to plants from soils. Increasing soil solution K concentration, on the one hand, mobilises RCs in soil solution (i.e. decreases K_D) by $\text{Cs}^+:\text{K}^+$ cation exchange in soils (Wauters et al. 1996b) while, on the other hand, reduces the plant uptake of RCs from soil solution by $\text{Cs}^+:\text{K}^+$ competition at plant roots (e.g. Waegeneers et al. 2001). Because the latter reaction is nonlinear and dominates the former linear reaction, the net effect is often a decrease in the TF of RCs with increasing K application to K-deficient soils. Along the same lines, it was found that the TFs are poorly related to soil clay content (Smolders et al. 1997). Increasing clay content, on the one hand, increases the K_D as RCs is specifically sorbed on clay minerals (e.g. Nakao et al. 2015; Uematsu et al. 2015, for Fukushima soils) while, on the other hand, reduces the soil solution K concentration (e.g. Absalom et al. 2001), thereby enhancing root RCs uptake. These contrasting effects obscure the net effect of soil clay content on soil-to-plant RCs transfer.

The concept of Eq.[2] and the double role of K on RCs bioavailability had been shown by Smolders et al. (1997) in a pot trial. This concept was later included in semi-mechanistic models (Absalom et al. 2001; Tarsitano et al. 2011) to predict the TF of RCs with soil and plant properties for temperate European soils and for grasses and cereal grains, all crops grown in unsaturated soils (Uematsu et al. 2016). The mechanisms behind these semi-mechanistic models (Absalom et al. 2001; Tarsitano et al. 2011) explain why K deficiency in soils enhances the TF of RCs (Waegeneers et al. 2001) and demonstrate that basically the TF can be predicted by the soil radiocaesium interception potential (RIP) and exchangeable K (Absalom et al. 2001; Uematsu et al. 2016). The RIP is a parameter to quantify the selective sorption of RCs in soils which was developed by Cremers et al. (1988) and Wauters et al. (1996b) for temperate European soils and which has been subsequently applied to soils collected from the Fukushima affected region (Nakao et al. 2015; Uematsu et al. 2015).

It is still unknown to what extent this established concept also applies to flooded soils in which rice is grown. Increasing water content by soil flooding lowers the soil redox potential that is expected to influence RCs transfer in different ways.

The first is the effect due to the accumulation of the ammonium cation (NH_4^+) in reduced soils because of limited nitrification (Fageria et al. 2011). The NH_4^+ cation is approximately fivefold more effective than K^+ to compete with RCs^+ for sorption on highly selective sites in soils (Cremers et al. 1988; Wauters et al. 1996b). Soil flooding has been shown to mobilise

RCs (i.e. decrease the K_D of RCs) in soils (Wang and Staunton 2010). These authors ascribed this RCs mobility to a higher soil solution NH_4^+ concentration measured under the saturated conditions than under the unsaturated conditions (Wang and Staunton 2010). By contrast, NH_4^+ shows less effect than K^+ on reducing the RCs uptake by plant roots from soil solution (Sanchez et al. 1999). Such reactions suggest that NH_4^+ accumulation in anaerobic flooded soils may enhance the RCs TF compared to unsaturated soils.

The second potential effect of soil flooding is simply related to dilution of not only RCs but also K in soil solution, with unknown consequences on RCs bioavailability.

The third potential effect is the increase in the effective diffusion coefficients of K and RCs in soil with increasing water-filled pores (Barracough and Tinker 1981), yielding a decrease in depletion of these cations in the rhizosphere. This depletion is more pronounced for K than for RCs (Waegeneers et al. 2001). It might suggest that soil flooding reduces K depletion, which leads to increasing competitive effects of K^+ at roots on blocking RCs⁺ uptake. As a consequence, this process is expected to decrease the TF of RCs upon flooding.

Finally, we may consider the potential long-term effects of soil flooding on the TF. The gradual fixation (Sawhney 1972) or aging of RCs in soil interlayer sites might be affected by soil flooding because of differences in soil solution K and NH_4^+ concentrations (Wauters et al. 1996b) and of the increase in the frayed edge sites due to the reductive dissolution of Fe(III) coating on the clay minerals (Zhang and Scherer 2002; Matsuoka and Moritsuka 2015).

It is not fully understood if the properties of RCs uptake by flooded rice are similar to those by ryegrass. A semi-mechanistic model has been developed predicting ^{137}Cs transfer to flooded rice (Keum et al. 2007); however, the model parameters for ^{137}Cs uptake to flooded rice in that model had been calibrated for ryegrass data. The inter-species differences in RCs bioavailability were suggested by pot trials with soils (Waegeneers et al. 2001) and by hydroponics (Broadley and Willey 1999). Better understanding mechanisms of RCs bioavailability to rice is beneficial to predicting soil-to-plant RCs transfer to rice.

A compilation of TF values for Cs from field observations in Japan indicated that the TF for stable caesium (Cs) does not significantly differ ($P > 0.05$) between brown rice grown in paddy fields and other grain cereals grown in unsaturated soils (Uchida et al. 2007). In addition, a worldwide collection of field TF data revealed that the RCs TF for paddy rice (brown rice) was

not significantly different ($P > 0.05$) from that for grain cereals grown in unsaturated soils (Uchida et al. 2009). In these comparisons, rice (paddy) and cereals (unsaturated soils) had not been collected from identical soils, i.e. unpaired comparisons in terms of soil types and plant species. Since soil properties such as K content and RIP have a large effect on the RCs TF, it is likely here that any effect of soil flooding on the RCs TF is obscured in the background variability.

The transfer of Cs to rice has been compared for the same soils using different water management approaches, allowing a pairwise comparison of the TF. The observation made in those studies is, however, not consistent. Continuous soil flooding in a field did not significantly increase the soil-to-brown-rice TF of stable Cs compared to intermittent irrigation ($P > 0.05$; Moreno-Jiménez et al. 2014). In contrast, a field trial in Japan revealed that continuous soil flooding significantly enhanced the TF of ^{137}Cs (Fukushima accident-derived) to brown rice compared to intermittent irrigation ($P < 0.05$; Wakabayashi et al. 2016). In contrast, a field trial with rice in a Japanese soil showed that the TF of Cs to brown rice under flooded conditions was only a factor 1.2 larger than that under unsaturated conditions (Nishiyama et al. 2016).

Taken together, this field evidence suggests that soil flooding might impact RCs bioavailability but the effects are likely small and inconsistent. As far as is known, no studies with flooded soils included soil solution data to identify soil factors affecting the bioavailability in such soils (Ohmori et al. 2014; Nishiyama et al. 2016; Wakabayashi et al. 2016).

Soil solution concentrations are useful to better understand the mobility of RCs and the competing effects of K^+ and NH_4^+ on mobilising RCs^+ (Smolders et al. 1997; Sanchez et al. 1999; Absalom et al. 2001; Uematsu et al. 2016). Multisite trials with flooded rice have revealed that increasing soil exchangeable K content reduces the TF of flooded rice, but that study was limited to four sites only (Kato et al. 2015).

Against this background, this study was set up to reveal the mechanisms of RCs bioavailability in flooded soils, thereby speculating the hypothesis that the soil solution composition (i.e. RCs, K and NH_4^+) may explain the variable RCs uptake from contrasting soil samples. Paddy soil samples with contrasting soil properties were collected from the Fukushima affected area and were cropped in the greenhouse with rice under flooded conditions. Soil solution was collected, and then the soil solution composition in the

rhizosphere was estimated by modelling with diffusion and mass flow in soils. As a reference to earlier work (e.g. Absalom et al. 2001), we cropped the same soils and some additional grassland soils with ryegrass under unsaturated conditions.

Materials and Methods

Experimental design

The soil-to-shoot transfer factor (TF) of ^{134}Cs was determined for rice grown in pots under flooded conditions with soils collected from paddy fields (“paddy soils”, $n = 9$, Table 1) in the Fukushima affected area. These soils were spiked with ^{134}Cs to obtain sufficient ^{134}Cs activity concentrations in soil solution. The same paddy soil samples and 13 other grassland soil samples from the Fukushima affected area (Table 1) ($n = 22$ in total) were additionally cropped with ryegrass under unsaturated conditions. These ryegrass experiments were included as a reference to earlier observations (Smolders et al. 1997; Sanchez et al. 1999) and the details of the experiments are given in Method A1 in Online Resource.

Soil sampling and physicochemical analysis

Topsoils (0-15 cm) were collected in 2013 from 9 paddy fields (Fig. A1 in Online Resource). These soils included typical Gleysols and Andosols (National Institute for Agro-Environmental Sciences 2009). The soils were air-dried and passed through a 2 mm sieve. The radiocaesium interception potential (RIP) of the soil was determined by the established method of pre-equilibrium of the soil with solution containing 100 mM Ca and 0.5 mM K (Wauters et al. 1996b; Uematsu et al. 2015), and the soil clay content was measured by the international standardised method (ISO 1998; Uematsu et al. 2015) (Table 1).

Soil spike with ^{134}Cs

In April 9, 2015, approximately 3 kg of each air-dried soil was transferred to a 10-L bucket. The paddy soil was moistened using a solution containing $^{134}\text{CsCl}$, $\text{Ca}(\text{NO}_3)_2$ and $\text{Ca}(\text{H}_2\text{PO}_4)_2$, and then homogenised by using a spatula. The amount of solution added was set to obtain saturated water contents of each soil. To make this solution, the reagents equivalent to 3.6 mmol N, 1.6 mmol P and 1800 kBq ^{134}Cs per kg dry soil were added to distilled water and

subsequently mixed to the original air-dried soils. These N and P doses were recommended values for rice paddies by the local Fukushima Prefectural Government (2006) but without K. The moist soils were stored in the buckets closed with a lid to reduce the evaporation and incubated for 28 d at room temperature. Every third day, the soils were weighed and the water loss was adjusted by adding distilled water. After the incubation, a subsample of the spiked soil was air-dried. The total concentration of ^{137}Cs (accident-derived) and that of ^{134}Cs in the spiked soils were measured by a NaI(Tl) gamma counter (1480 Wizard 3", PerkinElmer).

Growth of flooded rice

Seeds of typical Japanese paddy rice cultivar (*Oryza sativa* L., cv. Koshihikari) were germinated on a wet paper at 30°C for 48 h. The saturated soil, after 28-d incubation, was transferred to triplicate 1-L pots, filled with approximately 0.7 L of the soil. The exact volume of each potted soil was measured. Five germinated seeds per pot were then embedded at 5 mm depth from the soil surface. The rice plants were grown in a greenhouse for 32 d. The seedlings were thinned to three at 7 d after sowing. The daily maximum temperature was 35±6°C, minimum 16±2°C and relative humidity 42±10% (daily average ± standard deviation) during the growth. During the first 7 d, no water was added to promote rice root growth. From day 8 after sowing, water loss from each pot was recorded and at least 2 cm depth of water above the soil surface was maintained by adding distilled water.

Plant harvest and analysis

The shoots of flooded rice were cut at 3 cm above the soil surface. The shoots were dried at 70°C for 7 d followed by recording the shoot DW per pot. The dried shoot was then cut into short pieces and homogenised. The shoot was calcined at 450 °C for 6 h, dissolved in 1 mL of 12 M HCl and then diluted with 19 mL of distilled water. The ^{134}Cs and ^{137}Cs concentrations in these solutions were determined by a high purity germanium detector (SEGe, Canberra).

Soil solution

The soil solution was isolated from the wet soil, as outlined below. Both flooded soils (before rice growth) and unsaturated soils (before ryegrass growth), i.e. at 28 d after spiking, were homogenised. A syringe without a plunger was filled with a nylon mesh and approximately 50 g of the soil (one sample per soil type and water condition), then the syringe

was spun at $4000 \times g$ for 30 min. The soil solution was collected also after plant harvest (one sample per pot). For this collection, plant roots were carefully hand-picked from the potted soil, water content of the remaining soil was adjusted to the water content before sowing. The soil was then homogenised to analyse the soil solution averaged for the potted soil. Subsequently, the soil was centrifuged as mentioned above. The retrieved solution was passed through a $0.45 \mu\text{m}$ filter, and then analysed for the ^{134}Cs concentration by a NaI(Tl) gamma counter, for the K concentration by atomic absorption spectrometry (AAS; 932AA, GBC), and for the NH_4^+ concentration by the indophenol blue method (Searle 1984) at wavelength of 640 nm. The ^{134}Cs concentrations in soil, soil solution and plant samples were decay-corrected to the date of each plant harvest. The K_D of ^{134}Cs was computed as defined in Eq.[3]. The exchangeable K content in the spiked soils was determined before sowing by extracting the soils (air-dried) with 1 M $\text{CH}_3\text{COONH}_4$ for 24 h and by analysing the K concentration in the soil extracts by AAS (Table 1).

The labile solid-liquid distribution coefficient of ^{134}Cs (K_{DL} , L kg^{-1}) was predicted based on Eq.[4] established by Wauters et al. (1996a) which accounts for the competitive sorption of $^{134}\text{Cs}^+$ by K^+ and NH_4^+ cations on frayed edge sites considering a contact time of 24 h. The K_{DL} was calculated using Eq.[4] by substituting the RIP (mmol kg^{-1}) of the soils and the soil solution concentrations of K (m_K , mM) and NH_4^+ (m_{NH_4} , mM), all measured in this experiment, for Eq.[4].

$$K_{DL} = \frac{\text{RIP}}{m_K + K_C(\text{NH}_4/\text{K}) \cdot m_{\text{NH}_4}} \quad [4]$$

where we used $K_C(\text{NH}_4/\text{K})$, the selectivity coefficient for NH_4^+ in reference to K^+ at the frayed edge sites, of 5.3 (Wauters et al. 1996a).

The ^{134}Cs and K concentrations in the rhizosphere soil solution of flooded rice and ryegrass were estimated from the plant uptake of these elements and from estimated root surface area using an existing solute transport model of Baldwin et al. (1973). This model assumes that the uptake of solutes occurs by randomly dispersed roots and that solutes are supplied via diffusion and mass flow (Baldwin et al. 1973). The model assumptions and calculations are described in Online Resource (Method A2).

A pairwise comparison was performed by the paired *t*-test for the TF of ^{134}Cs between flooded rice and ryegrass and for the K_D of ^{134}Cs between soils under flooded conditions and those under unsaturated conditions (each TF and K_D log-transformed) by using program JMP (SAS Institute Inc. 2013). Correlation was tested between the shoot ^{134}Cs concentration and the concentration ratio of ^{134}Cs to K in soil solution. Then, linear regression lines were described for each of ryegrass and flooded rice. The difference in the slope and in the intercept of these regression lines between the plant types was tested by the analysis of covariance (ANCOVA) using R version 3.4.0 (The R foundation 2017). The TF was linked with the RIP, soil exchangeable K and crop type by a multiple linear regression using program JMP.

Results

Growth and radiocaesium transfer

All flooded rice plants developed without visible deficiency symptoms. The dry weight (DW) of rice shoots varied significantly between soils (Fig. A2a in Online Resource). The TF of ^{134}Cs for flooded rice varied 110-fold among the soils ($n = 9$, Fig. 1) and the TF for ryegrass ranged 940-fold (Fig. A3 in Online Resource). The TF of the 4.2-year-old ^{137}Cs derived from the Fukushima accident was 3.3-fold smaller than that of freshly-spiked ^{134}Cs for flooded rice (Fig. 1), while it was, on average, 4.5-fold smaller for ryegrass (Fig. A3). The TFs of ^{134}Cs for ryegrass were factors 1.2-14 (geometric mean 4.0) larger than that for flooded rice ($P < 0.05$, paired *t*-test, comparison of paddy soils only, $n = 9$; Figs. 1 and A3).

Solid-liquid RCs distribution

The solid-liquid distribution coefficient (K_D) of ^{134}Cs ranged between 290-6700 L kg⁻¹ among the nine paddy soils which had been incubated under saturated conditions for 28 d after spiking (i.e. measured before the rice growth) (Fig. 2). The K_D of ^{134}Cs of the saturated soils was 1.1-2.5-fold larger than that of the corresponding unsaturated soils (measured before ryegrass growth), except for one soil type (Fig. 2). The difference in the K_D was significant between the flooded and unsaturated soils ($P < 0.05$, paired *t*-test).

Concentrations of K and NH_4^+ are among the factors controlling the distribution (K_D) of ^{134}Cs (Wauters et al. 1996a). The ratio of m_K in the flooded soils to that in the corresponding unsaturated soils was, on average, 0.57 (range 0.39-0.83, Fig. 3), indicating dilution of K in the soil solution with increasing soil water content, as soil water content approximately doubled under flooded compared to the unsaturated conditions. By contrast, the ratio of m_{NH_4} in the flooded soils to the unsaturated soils ranged between 0.59-2.1 (average 1.2, Fig. 3). These ratio values for m_{NH_4} were, on average, larger than those for m_K , indicating enhanced net NH_4^+ accumulation in soil solution due to soil flooding. No significant difference in m_{NH_4} was observed between flooded and unsaturated soils (Fig. 3) according to a pairwise comparison ($P > 0.05$, $n = 9$, paired t -test).

The measured K_D of ^{134}Cs at 28 d after spiking (Fig. 2) exceeded the labile K_{DL} (Eq.[4], valid for 24-h-old RCs) calculated from RIP, m_K and m_{NH_4} (Fig. 3, Tables A1 and A2 in Online Resource), on average, by factors 3.7 for flooded soils and 2.4 for unsaturated soils. This factor for flooded soils was significantly larger ($P < 0.05$, paired t -test) than that for unsaturated soils. The K_D (Fig. 2) was positively and significantly correlated with the labile K_{DL} (Eq.[4]) for flooded soils ($R^2 = 0.79$) and for unsaturated soils ($R^2 = 0.87$, correlations not shown). The systematically larger K_D than K_{DL} can be explained by the fixation of ^{134}Cs (aging effects) in soils which has been incubated for 28 d. The labile K_{DL} accounts for sorption of ^{134}Cs for 24 h of contact time, equivalent to the contact time of the RIP determination.

Plant uptake of RCs

The shoot ^{134}Cs concentration showed little correlation with the ^{134}Cs concentration in soil solution (Fig. 4a). In contrast, the shoot ^{134}Cs concentration was positively and significantly ($P < 0.001$, Fig. 4b) correlated to the concentration ratio of $^{134}\text{Cs}:\text{K}$ in the soil solution both for flooded rice and ryegrass. This significant correlation for ryegrass (Fig. 4b) was well in line with earlier observations (Smolders et al. 1997; Sanchez et al. 1999). The shoot ^{134}Cs concentration of flooded rice can be reasonably well predicted from the ^{134}Cs and K concentrations in the soil solutions. The intercept of the regression line for ryegrass was significantly larger ($P < 0.01$, ANCOVA) than that for flooded rice (Fig. 4b), showing that the shoot ^{134}Cs concentration was 4.1-fold higher for ryegrass than that for flooded rice at the equal ratio of soil solution $^{134}\text{Cs}:\text{K}$. This result suggested that young ryegrass plants take up ^{134}Cs and translocate it to shoots more efficiently than rice at corresponding K levels in soil solutions.

The slope of the regression lines was not significantly different between flooded rice and ryegrass ($P > 0.05$, ANCOVA, unequal slope assumption; Fig. 4b).

Regressions

The TF of ^{134}Cs was negatively and significantly ($P < 0.05$) correlated to both the soil RIP (Fig. A4a in Online Resource) and soil exchangeable K (ExK) (Fig. A4b) for both flooded rice ($n = 9$) and ryegrass ($n = 22$, combined data from paddy and grassland soils). A multiple linear regression linking the TF (Figs. 1 and A3) with the RIP and ExK of the soils (Table 1) yielded a model (Eq.[5]) in which the standard errors of each estimate are shown in the parentheses;

$$\log_{10}(\text{TF}) = 0.380(\pm 0.093) - 0.302(\mp 0.054)\text{RIP} - 1.09(\mp 0.11)\text{ExK} + \begin{cases} 0.14 \text{ if grass} \\ -0.14 \text{ if rice} \end{cases} (\pm 0.055)$$

[5]

in which the soil RIP (mol kg^{-1} , $P < 0.001$), ExK ($\text{cmol}_c \text{ kg}^{-1}$, $P < 0.001$), and plant type ($P < 0.05$) explained 66% of the variation in the TF. The plant type was a significant factor, indicating that the TF was significantly larger for ryegrass than flooded rice at the same RIP and ExK in soils.

Discussion

The results confirmed our hypothesis that RCs transfer to flooded rice can be predicted from soil solution concentrations of RCs and K. The bioavailability of RCs from soil solution to shoots was significantly smaller for flooded rice than for ryegrass at equal K concentrations in soil solutions. The multivariate model (Eq.[5]) also suggested a smaller soil-to-shoot TF of RCs for flooded rice than for ryegrass at equal soil properties. Our findings also showed that the mobility of ^{134}Cs in soils was smaller (i.e. larger K_D) in flooded soils than in unsaturated soils at 28 d after ^{134}Cs spike (Fig. 2), in contrast to our hypothesis that ^{134}Cs can be mobilised by enhanced NH_4^+ in flooded soils (see discussions below).

Differences in RCs mobility between flooded and unsaturated soils due to soil chemical factors

Our study indicated that increasing soil water content lowered m_K while m_{NH_4} was unaffected by soil flooding ($P > 0.05$, Fig. 3). The variable effect of soil flooding on m_{NH_4} between the nine soils (Fig. 3) was corroborated by Wang et al. (2001), reporting contrasting effects of soil flooding on nitrogen production in soils among 20 soils. The variable trend of soil solution NH_4^+ due to flooding (Fig. 3) may be related to differences in mineralisation rate of nitrogen led by soil degradation of organic matter and in the types of expansible clay minerals, where NH_4^+ can be sorbed and/or fixed (e.g. Schneiders and Scherer 1998) between these soils.

On the one hand, soil reduction enhanced m_{NH_4} for some soils by factors up to 2.1 but not for all soils (Fig. 3). This results can be corroborated by the findings of Devkota et al. (2013) and Tete et al. (2015), who reported higher NH_4^+ concentrations in soils under flooded conditions than unsaturated conditions. The enhanced m_{NH_4} under saturated conditions for limited numbers of the soils suggests that nitrification was inhibited (Fageria et al. 2011). On the other hand, the measurable m_{NH_4} for unsaturated soils (Fig. 3) may be caused by NH_4^+ produced by degradation of soil organic matter under aerobic conditions (e.g. Schneiders and Scherer 1998; Said-Pullicino et al. 2014).

Consequently, the parameter ($m_K + 5.3m_{NH_4}$) showing the combined competition effect on RCs exchange (Wauters et al. 1996a), was, on average, only 1.1-fold smaller for flooded soils than for unsaturated soils, suggesting that the combined ion competition effects on RCs were similar between both water managements.

The ratio of the measured K_D (Fig. 2) to the labile K_{DL} (Eq.[4]) predicted from RIP, m_K and m_{NH_4} was calculated. The ratio was, on average, 1.6-fold larger for flooded soils than unsaturated soils, indicating that the sorption and aging of RCs during 28 d since spiking were significantly ($P < 0.05$) more enhanced in flooded soils than in unsaturated soils. Experiments by Zhang and Scherer (2002) and Matsuoka and Moritsuka (2015) indicated that soil flooding treatments may enhance the fixation of NH_4^+ . This NH_4^+ fixation has been attributed to reductive dissolution of Fe(III) minerals, which are coating clay minerals, by anoxic microbial activity (Zhang and Scherer 2002) and to negative charge induced by the reduction of octahedral Fe(III) to Fe(II) in soil clay minerals (Nieder et al. 2011). It may be possible that the Fe reduction opens up some frayed edge sites (i.e. increases RIP). Along the same lines, it might be speculated that the aging of RCs in the interlayer cation exchange sites may be

enhanced under reduced conditions due to reduction of Fe in soil clays leading to opening of frayed edge sites (i.e. by the processes similar to those for NH_4^+), since NH_4^+ and RCs^+ can compete for the same interlayer sites in clay minerals (Sawhney 1972).

To summarise, contrary to our hypothesis, our findings indicated that NH_4^+ was unaffected by soil flooding ($P > 0.05$, Fig. 3), and hence, increased RCs mobility in flooded soil was not demonstrated. In contrast, the soil solution RCs concentration was rather enhanced by flooding (Fig. 2), suggesting suppressed RCs mobility in flooded soils by enhanced RCs fixation.

RCs bioavailability to flooded rice and differences between flooded and unsaturated soils

The strong and significant correlations (Fig. 4b) confirmed that the ^{134}Cs availability to flooded rice shoots from soil solution was sharply stimulated at low m_K , as it had been established for ryegrass and for soils from temperate European regions (Smolders et al. 1997; Waegeneers et al. 2001). This findings of the effects of soil solution K on RCs uptake by flooded rice may be useful to predict RCs transfer to rice. The results indicated that K^+ plays a competitive role on RCs^+ uptake by plants which was similar, but not identical for both plants. Ryegrass plants took up more RCs and more strongly discriminated RCs over K than flooded rice, as suggested from the significantly different intercept in the regression lines, corresponding to a factor 4.1 difference in the shoot ^{134}Cs concentration (Fig. 4b). This difference may be related to a genuine difference in RCs uptake mechanisms between these two plants. No study was found in which seedling of both plants were grown hydroponically side-by-side in RCs spiked solutions to test this hypothesis. Compiled data of plant availability of RCs for different plants suggested that the difference in RCs uptake between different plant species at the same external K concentration is smaller than the difference in the RCs uptake for the same plant under contrasting K concentrations in external solution (Zhu and Smolders 2000).

Alternatively, we attribute this difference in RCs uptake to the differences in the $^{134}\text{Cs}:\text{K}$ ratio in the rhizosphere solutions between flooded and unsaturated soils. To underpin this hypothesis, the plant uptake rates of ^{134}Cs and K were combined with a transport model to estimate time- and root surface area averaged concentrations.

The transport model (Baldwin et al. 1973) predicted that the ^{134}Cs and K concentrations in rhizosphere soil solutions of flooded soils were depleted by 7.2% (^{134}Cs) and by 27% (K)

compared to the bulk soil solution concentrations (Table 2). The corresponding predicted concentrations in the rhizosphere for ryegrass (unsaturated) were negative values (Table 2). This result for ryegrass means that the observed uptake of ^{134}Cs and K per unit root surface area is larger than what can be obtained in a zero-sink scenario at the root surface. This anomaly is the result from incorrect parameter estimates, e.g. underestimated root surface area or overestimated effective diffusion coefficients. The contrasting predictions for flooded rice and ryegrass suggested, however, that the depletion of mainly K and, to a lesser extent, ^{134}Cs in ryegrass rhizosphere was considerable.

We further estimated the rhizosphere concentrations using the model (Baldwin et al. 1973) with the soil parameter of the unsaturated soils and the root uptake parameters of rice plants, and vice versa, i.e. with the soil parameter of the flooded soils and the root uptake parameters of ryegrass plants. The estimated rhizosphere K concentration for rice under unsaturated conditions was smaller than in flooded conditions, but still a positive value (Table A3 in Online Resource). The larger gradients of the K concentrations in the rhizosphere of the unsaturated soils than in flooded soils for the same plant parameter is the mere consequence of the differences in the effective diffusion coefficients that increase proportionally to the square of the volumetric soil water content. The modelled rhizosphere K concentration under flooded conditions using the ryegrass parameters yielded a negative value as in the unsaturated soil for the grass (Table A3). This result suggests that the effects of plant properties, i.e. larger root uptake of K by ryegrass plants than by rice, on rhizosphere K concentrations dominate over the effects of soil water regime. This suggests that the overall larger ^{134}Cs bioavailability to ryegrass in unsaturated soils is related to the larger K uptake rate of grass, yielding more K depletion in its rhizosphere with local lower K^+ facilitating the RCs^+ uptake by the roots. Indeed,, total root K uptake rates are is larger for ryegrass than for rice. This difference is indicated by the more significant decrease in bulk K concentrations in ryegrass than in flooded rice during growth (Table A1) and by the significantly larger K content in ryegrass shoot than

that in flooded rice (Fig. A2). The bulk soil solution concentrations of K after plant growth were factors 7.4-46 lower than the initial concentrations for ryegrass-grown soils, whereas the corresponding factors for rice-grown soils were 1.4-9.0. Although increasing the plant density of rice might enhance K depletion in soil solution by plant uptake, this K depletion is assumed to be only small as K uptake per area would not largely change since increasing plant biomass may dilute K in plants (e.g. Dobermann et al. 1996).

The larger K depletion in the unsaturated soils by the faster growing ryegrass than that in the flooded soils by rice might obscure any physiological differences in RCs and K uptake between these plants. The comparison of genuine differences in RCs bioavailability between plants requires rescaling the $^{134}\text{Cs}:\text{K}$ concentration ratio of bulk soil solutions to that in the rhizosphere, a transformation which is not feasible due to the uncertainties in the model.

To conclude, soil-to-plant RCs transfer to flooded rice is determined by the mechanisms similar to those established for unsaturated soils, confirming the earlier knowledge of the role of K on RCs. The role of NH_4^+ on RCs bioavailability was not very large, contrary to our hypothesis, at least not in these potted soils. Our findings also explain why RCs transfer to flooded rice can be predicted in a semi-mechanistic way from the combination of soil properties such as soil clay content and exchangeable K which both affect the soil solution RCs:K ratio. This finding suggests that field-based TFs can be predicted from these properties, as was done before for unsaturated soils and upland plants (Absalom et al. 2001). This prediction for field data might require a simple recalibration of existing models since the RIP and m_K are both somewhat smaller in the soils from the Fukushima affected area than in the soils from temperate Europe, for which the model (Absalom et al. 2001) had been calibrated (Uematsu et al. 2015, 2016).

Acknowledgements S. U. acknowledges the PhD fellowship provided by the Flemish Government, Belgium. We cordially thank the following researchers for their great support on our soil sampling campaign for this research: Drs. N. Yamaguchi, I. Taniyama, and T. Ohkura (National Inst. for Agro-Environ. Sci.), T. Saito, K. Yoshioka, I. Arakawa, and T. Saitou

(Fukushima Agr. Tech. Cent.), T. Ota (Tohoku Agr. Res. Cent.), D. Yamada and H. Harada (National Inst. Livestock and Grassland Sci.), T. Nishimura and M. Mizoguchi (The Univ. of Tokyo), K. Noborio (Meiji Univ.), K. Nanba (Fukushima Univ.), K. Okada (Iwate Univ.), M. Hojito, N. Ito and M. Natsubori (Kitasato Univ.), the group Resurrection of Fukushima and local farmers in Fukushima. Technical support from Dr. M. Bruggeman (SCK•CEN) for ^{134}Cs and ^{137}Cs analysis in the plant samples was appreciated. We thank the anonymous reviewers for their constructive comments and suggestions.

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Figure captions

Fig. 1 The soil-to-plant transfer factor (TF) of 28-day-old ^{134}Cs and 4.2-year-old ^{137}Cs (accident-derived, detected in soil I only) determined for flooded rice grown in the paddy soils. The mean (symbols) and the standard deviation (error bars) ($n = 3$) are presented. The error bars may be invisible when the error bar is smaller than the symbol

Fig. 2 The solid-liquid distribution coefficient (K_D) of ^{134}Cs for corresponding paddy soils incubated for 28 d after spiking either under flooded conditions (before sowing rice) or under unsaturated conditions (before sowing ryegrass). Paired t -test was performed for log-transformed K_D and revealed an overall significant effect of water regime on $\log(K_D)$; *: $P < 0.05$

Fig. 3 The soil solution concentrations of K (m_K) and NH_4^+ (m_{NH_4}) in soils incubated for 28 d either under flooded or under unsaturated conditions for the corresponding soils. The dotted line indicates a 1:1 line

Fig. 4 (a) A scatter plot of the shoot ^{134}Cs concentration and the soil solution ^{134}Cs concentration (measured before sowing) for flooded rice and ryegrass. (b) Positive correlations between the shoot ^{134}Cs concentration and the soil solution concentration ratio of $^{134}\text{Cs}:\text{K}$ (before sowing) for both plants. The correlation determined for ryegrass grown in potted soil trials (Smolders et al. 1997; $n = 30$) is also presented. The linear regression lines are shown in (b), as the solid line for flooded rice and broken line for ryegrass from the presented study, and the dotted line for Smolders et al. (1997) ryegrass data. In (b), linear regression was made between $\log(y)$ and $\log(x)$ for each data group where y is the shoot ^{134}Cs concentration and x is the soil solution $^{134}\text{Cs}:\text{K}$ concentration ratio. The significance of the correlation was indicated after the R^2 values in figures (a) and (b); ***: $P < 0.001$, ns: $P > 0.05$. The differences in the slope and in the intercept between these data groups were tested by the analysis of covariance (ANCOVA), and indicated as; **: $P < 0.01$, *: $P < 0.05$, ns: $P > 0.05$

Table captions

Table 1 Selected soil parameters^a of the soils used for the pot trials.

Table 2 Composition of bulk soil solutions at the harvest of the plants and composition estimated in the rhizosphere using a steady-state assumption of a soil-to-plant transport model^a.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

1 **Table 1** Selected soil parameters^a of the soils used for the pot trials.

Field land use	Pot trial	<i>n</i> ^b	RIP ^c (mmol kg ⁻¹)	ExK (cmol _c kg ⁻¹)	Clay content ^c (%)
Paddy rice field	Rice (flooded) & ryegrass (unsaturated)	9	997 (283-1883)	0.46 (0.17-0.83)	17.0 (7.5-25.9)
Grassland	Ryegrass (unsaturated)	13	153 (288-4773) 3	0.99 (0.21-1.93)	17.4 (6.8-30.7)

2

3 ^a RIP: radiocaesium interception potential of soils, ExK: soil exchangeable K content. The average and the range (in the parenthesis) are presented.

4 The ExK was measured after spiking the soils with ¹³⁴Cs.

5 ^b The number of the soil sampling sites.

6 ^c The RIP and clay content for five out of the nine paddy soils and seven out of the 13 grassland soils were published (Uematsu et al. 2015). The

7 RIP of the 22 soils was listed in Table A2 (Online Resource).

Table 2 Composition of bulk soil solutions at the harvest of the plants and composition estimated in the rhizosphere using a steady-state assumption of a soil-to-plant transport model^a.

Soil water management- Plant type	¹³⁴ Cs (Bq L ⁻¹)		K (μmol L ⁻¹)	
	Bulk	Rhizosphere ^b	Bulk	Rhizosphere ^b
Flooded soils-Rice	627 (41-1962)	601 (34-1960)	85 (10-197)	72 (3-179)
Unsaturated soils- Ryegrass	195 (10-789)	< 0 ^c	37 (5-95)	< 0 ^c

^a The model calculation is detailed in Method A2. The general average and range (in the parenthesis) of the concentration of different soils ($n = 9$; paddy soils only) are presented.

^b Estimated soil solution concentrations at the root surface.

^c Estimation yielded negative values: the uptake flux is larger than that estimated from a zero sink scenario, see discussion.