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The ratio of plant ^{137}Cs to exchangeable ^{137}Cs in soil is a crucial factor in explaining the variation in ^{137}Cs transferability from soil to plant

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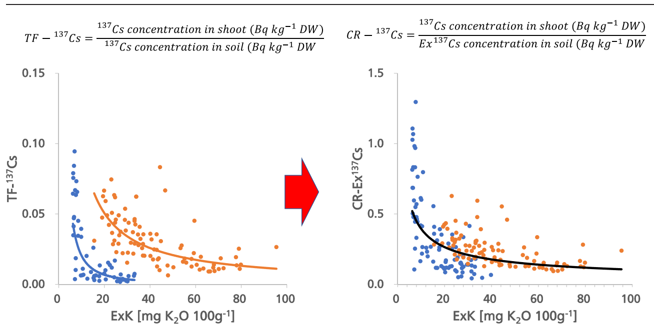
HIGHLIGHTS

- Exchangeable K level is known to be a detrimental factor to mitigate ^{137}Cs uptake by plant.
- Relationship between exchangeable K and the transfer factor was not equal at different soils.
- The amount of exchangeable ^{137}Cs is also an important factor to explain the radioactivity of the plant.
- The concentration ratio of plant ^{137}Cs to soil exchangeable ^{137}Cs showed a significant correlation with exchangeable K.

GRAPHICAL ABSTRACT



Usage of Ex-^{137}Cs improve the prediction of plant ^{137}Cs uptake.

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ABSTRACT

To mitigate radioactive cesium from soil to plant, increasing and maintaining the exchangeable potassium (ExK) level during growth is widely accepted after Tokyo Electric Company's Fukushima Dai-ichi Nuclear Plant accident in Japan. This is because the antagonistic relationship between soil solution K and ^{134}Cs + ^{137}Cs (RCs) concentrations changes the transfer factor (TF: designated as the ratio of radioactivity of plant organ to soil) of RCs. As the relationship between ExK and TF depends on the soil types, crop species, and other environmental factors, the required amount of ExK should be set to a safe side. Eleven years after the accident, as the activity of ^{134}Cs was almost negligible, ^{137}Cs became the main RCs in most of the agricultural fields in Fukushima Prefecture. We propose a new indicator, the concentration ratio of plant ^{137}Cs to soil exchangeable ^{137}Cs (Ex^{137}Cs), instead of TF, which showed a better correlation with ExK even among soils with different properties (or mineralogy).

1. Introduction

Tokyo Electric Power Company's Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident in 2011 caused a severe contamination of radioactive materials to the large area of the Southern Tohoku area and Northern Kanto area of Japan. Eleven years after the accident, the major concern in the agricultural sector was to maintain the countermeasure against ^{137}Cs transfer from soil to plants. Physical removal of contaminated soil by topsoil stripping has been carried out in contaminated areas (Ministry of Agriculture, Forestry and Fisheries (MAFF), 2013); however, complete removal of ^{134}Cs and ^{137}Cs (RCs) is not realistic, and a substantial amount of radioactivity still remains in decontaminated agricultural fields (e.g., Evrard et al., 2019). Regardless of whether decontamination was carried out, Fukushima Prefecture government has recommended that the most reliable countermeasure to mitigate RCs transfer from soil to plant is maintaining an exchangeable potassium (ExK) level higher than 25 mg K_2O 100 g^{-1}

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soil during growth in the case of brown rice production based on the report by Kato et al. (2015), and similar countermeasures were taken in other crops.

On the other hand, a large monitoring survey of paddy fields by MAFF revealed that even at the same ExK level, the TF seems to be different among areas (Yamamura et al., 2018). It is shown that TF becomes high at the same ExK in Hamadori district (coastal area) compared to Nakadori district (basin area between mountainous areas) in Fukushima Prefecture (Yamamura et al., 2018). The reason for the difference in the TF in different areas is not clear, but factors such as soil types, environmental conditions, the quantity and the fallen form of RCs may be important to explain these differences (e.g., Sakai et al., 2021). The effect of soil type on TF has been pointed out after the Chernobyl accident (e.g., International Atomic Energy Agency (IAEA), 2006). The difference among the soil types is as large as 10 times between sandy soil and clay soil (Lembrechts, 1993).

It is known that clay minerals form frayed edge sites (FESs) at increased frayed edges of neighboring layers of mica crystal lattices (Sawhney, 1972; Tamura and Jacob, 1960). FES selectively captured the nonhydrated size of Cs ions (Francis and Brinkley, 1976). It has been reported that the selectivity for Cs sorption to FES is 1000 times and 200 times for K^+ and NH_4^+, respectively (Wauters et al., 1996). Therefore, the dynamics of Cs ions are highly regulated by the amount of FES in the soil (Cremers et al., 1988; Hird et al., 1996). It is considered that RCs interception potential (RIP) can be a good indicator for FES. The importance of RIP was confirmed from a survey on pasture lands after the FDNPP accident. Harada (2014) found that RIP had a significant relationship with pasture radioactivity. However, the actual application of RIP to distinguish the RCs transferability at each soil type-based agricultural field was not predictive in the case of the FDNPP accident. Although RIP is currently able to represent the soil characteristics for RCs selectivity, as agricultural activity change RIP (Ogasawara et al., 2017), it is difficult to apply to the agricultural field. On the other hand, it is difficult to predict precise TF by using only ExK of soil even in the same crop species. Yamamura et al. (2018) showed that TF at the same ExK level was different between the Nakadori and Hamadori regions of Fukushima Prefecture.

It is also known that the relationship between soil ExK and TF is not stable among crop species (e.g., IAEA, 2020). Among the major crop species in this area, soybean and buckwheat seem to have higher TFs at the same ExK of rice based on a large monitoring survey (IAEA, 2020). We used soybean as a representative upland crop in this area.

Furthermore, based on the decontamination process on contaminated agricultural fields after the FDNPP accident, the topsoil was removed, and an equivalent amount of noncontaminated mountainous soil was dressed and cultivated to 15 cm (MAFF, 2013). The field soil characteristics have been changed from the original soil type, which also makes it difficult to decide the field characteristics for RCs behavior of the original soil. Although plant roots take up nutrients (including RCs) from the soil solution, the analysis of the soil solution is time-consuming and sometimes difficult to obtain under upland conditions. Analysis of exchangeable RCs is often performed to assess the availability of RCs in a simplified manner (e.g., Kondo et al., 2015; Yagasaki et al., 2019a, 2019b; Li et al., 2022), and exchangeable RCs can be more easily used for soil diagnosis than RIP measurements in terms of high throughput and low cost. It is also important to consider K availability not only ExK but also non-exchangeable K (NexK), as Kurokawa et al. (2020) showed that in rice cultivation, fields with NexK > 50 mg K_2O 100 g^{-1} soil have a lower risk of transferring RCs to the crop. As distribution of K and Cs fractions such as soil solution, exchangeable, and non-exchangeable fraction for K and Cs change by soil type, there is still a requirement for a more precise estimation of K and Cs availability among different soil types. In this manuscript, an idea to determine the soil with high risk in the meaning of higher transferability of RCs to the plant is presented, where precise management of K application on two different soybean fields for two years is still needed.

2. Materials and methods

2.1. Cultivation

Experimental fields were established in a field located in northern Nakadori and northern Hamadori and designated Field A and Field B, respectively. The parent material in the surrounding area is granite and sedimentary rock (Geological Survey of Japan AIST, 2014). In Field A, the field has been used since 2013, and different levels of potassium fertilization have been established for the experiment. In Field B, the field was decontaminated by topsoil stripping (above ca. 5 cm) and then dressed and mixed noncontaminated mountainous soil to decrease the radioactivity of the cultivation soil. The details of the soil physico-chemical properties of the dressed soil are not available. The cultivation was initiated from 2016 as an experimental field. Soybean (*Glycine max* Merr. var. Tachinagaha) was cultivated in 2019 and 2020. Conventional fertilization was carried out except for potassium on the same day as sowing day (20 kg N ha^{-1} as a primary fertilizer by ammonium sulfate, 100 kg P_2O_5 ha^{-1} by superphosphate, and 1000 kg ha^{-1} magnesium lime), and 60 kg N ha^{-1} as additional fertilizer on the day as intertillage ridging day, the middle of July. In the case of K fertilizer, three different levels (Low-K, Middle-K, High-K) were prepared by potassium sulfate. In Field A, the Low-K was fertilized with no potassium and the Middle-K and High-K were fertilized with potassium sulfate to bring ExK level up to 25, 45 mg K_2O 100 g^{-1} soil, and then the conventional amount of K fertilizer (100 kg ha^{-1}) was applied by potassium sulfate. The fertilizer design for Field A was set based on Fukushima Prefecture's recommendation that ExK be increased to 25 mg K_2O 100 g^{-1} soil before conventional fertilizer application. In Field B, 0, 100, and 300 kg K_2O ha^{-1} were applied. The experimental design was a randomized block design with three replications. In each field, three replications were designed for No-K, Middle-K, and High-K plots, respectively. Field A is classified Grey lowland soil, Field B is classified Andosol according to the soil texture classification (The Japanese Society of Pedology, 2017), and the physical properties are shown in Table S1. The plots were designed at 3.75 × 6.0 m in Field A and 4.5 × 5.0 m in Field B. Five rows were created in Field A and seven rows in Field B. Three seeds were sown at 70 cm × 20 cm intervals and each plant was then thinned to two plants.

2.2. Sampling

Plant samples were collected at the vegetative growth stage, flowering stage, pod-forming stage, seed-growing stage and harvesting stage. The details of the sampling dates are shown in Table 1. Three to twenty plants

Table 1
Sampling schedule.

Year	Field	Sampling day	Growth stage					
			Sowing	Vegetative	Flowering	Pod formation	Maturity	Harvest
2019	A (Nakadori)	Days after sowing	0 (27-May)	28	56	91	127	154
	B (Hamadori)	Days after sowing	0 (30-May)	25	53	88	124	151
2020	A (Nakadori)	Days after sowing	0 (23-Jun)	28	43	71	100	125
	B (Hamadori)	Days after sowing	0 (18-May)	44	64	107	136	160

were collected depending on the sampling stage in each replication. After the seed growth stage, detached leaves and petioles were collected by using a net before falling down to the soil. Samples taken from each replication pooled and mixed then used for the subsequent analysis.

Each sample was dried at 80 °C for 48 h and then weighed and ground for subsequent analysis.

Soil was collected in the middle of the sampled plant in a raw. Five samples from each plot were collected and pooled. Soil was mixed well and used for the subsequent analysis. Soil was collected up to 15 cm from the surface.

2.3. Sample analysis

The RCs concentration was determined by using a Germanium semiconductor detector (GC2520-7500SL, GC4020-7500SL, GCW2523-7905-30U-ULB, Million Technology Canberra), and the analysis was carried out until the relative standard deviation was lower than 10 % for plants and 5 % for soils. Efficiency calibration was performed using a mixed nuclide solution (JRIA, MX033U8PP).

The sampled soil was pooled and dried for 1 week at 40 °C and then sieved, and debris larger than 2 mm mesh was removed, then the soil total RCs concentration was determined. For the ExK and exchangeable ^{137}Cs (Ex^{137}Cs), soil was extracted by 1 M ammonium acetate at soil:solution ratio of 1:10, shaken for 1 h and filtered through an ADVANTEC No. 5A quantitative filter paper (Advantec Toyo Kaisha, Ltd., Tokyo, Japan) and 0.45 μm CA Syringe Filter (Osaka Chemical, Osaka, Japan). Then, ExK was determined by using ICP-MS (ELAN DRC-e; PerkinElmer, Waltham, MA, USA). Ex^{137}Cs concentration was also determined by using a Germanium semiconductor detector.

The nonexchangeable fraction of K was determined by the hot HNO_3 method and tetraphenylboron (TPB) method. Hot HNO_3 methods were based on Ogasawara et al. (2019). The potassium extracted by the hot HNO_3 method was designated plant available K (PhK-HNO_3) and was determined by using ICP–MS (to ensure the accuracy of the analysis external calibration standards containing K were measured after every 10 samples). The nonexchangeable fraction of K (NexK-HNO_3) was calculated by subtracting ExK from PhK-HNO_3. The TPB method was based on Carey et al. (2010). The potassium extracted by TPB method was designated plant available K (PhK-TPB) and determined by using atomic absorption spectrometry (AA-6200, SHIMADZU, Kyoto, Japan, to ensure the accuracy of the analysis external calibration standards containing K were measured after every 10 samples). The nonexchangeable fraction of K (NexK-TPB) was calculated by subtracting ExK from PhK-TPB.

The water-soluble fraction of K (WS-K) was followed by the Soil Environment Analysis Method Editorial Committee (1997). Soil was extracted by Ultra-pure water at soil:solution ratio of 1:10, shaken for 1 h and filtered through ADVANTEC No. 6 quantitative filter paper (Advantec Toyo Kaisha, Ltd., Tokyo, Japan). The WS-K was determined by using ICP–MS.

The transfer factor of RCs (TF-^{137}Cs) from soil to plant was determined as follows:

$$\mathrm{TF}-{}^{137}\mathrm{Cs}=\frac{{}^{137}\mathrm{Cs\ concentration\ in\ shoot}\ \left(\mathrm{Bq\ kg^{-1}\ DW}\right)}{\mathrm{total}\ {}^{137}\mathrm{Cs\ concentration\ in\ soil}\ \left(\mathrm{Bq\ kg^{-1}\ DW}\right)}.$$

A quantitative method for soil mineral composition was performed. The internal standard mineral corundum (Al_2O_3) (Baikalox 3.0CR, Baikowski, France) and the soil sample were mixed at a ratio of 4:1 and ground with 7 mL of ethanol in an XRD grinding mill (XRD-Mill McCrone, Verder Scientific, Tokyo, Japan) for 10 min and dried at room temperature. The powder sample was loaded onto a sample plate using the front-loading method and measured using an XRD system (Miniflex 600, Rigaku, Tokyo, Japan) with a scanning angle of 5–65°, a sampling width of 0.01°, a scan speed of 10.0° min^{-1}, a voltage of 40 kV, and a current of 15 mA. The measurement was performed using an XRD system (Miniflex 600, Rigaku, Tokyo, Japan). The quantification method was performed by the full pattern summation method, which is implemented in the "powdR" package (Butler and Hillier, 2020) of the R language and environment for statistical computing (RStudio Version 1.4.1717).

Other soil chemical properties are given in the supplementary data.

3. Results

3.1. Growth of soybean

Successive changes in the dry weight in 2019 and 2020 are shown in Figs. 1 and 2, respectively. In 2019, there was an increase in the dry weight

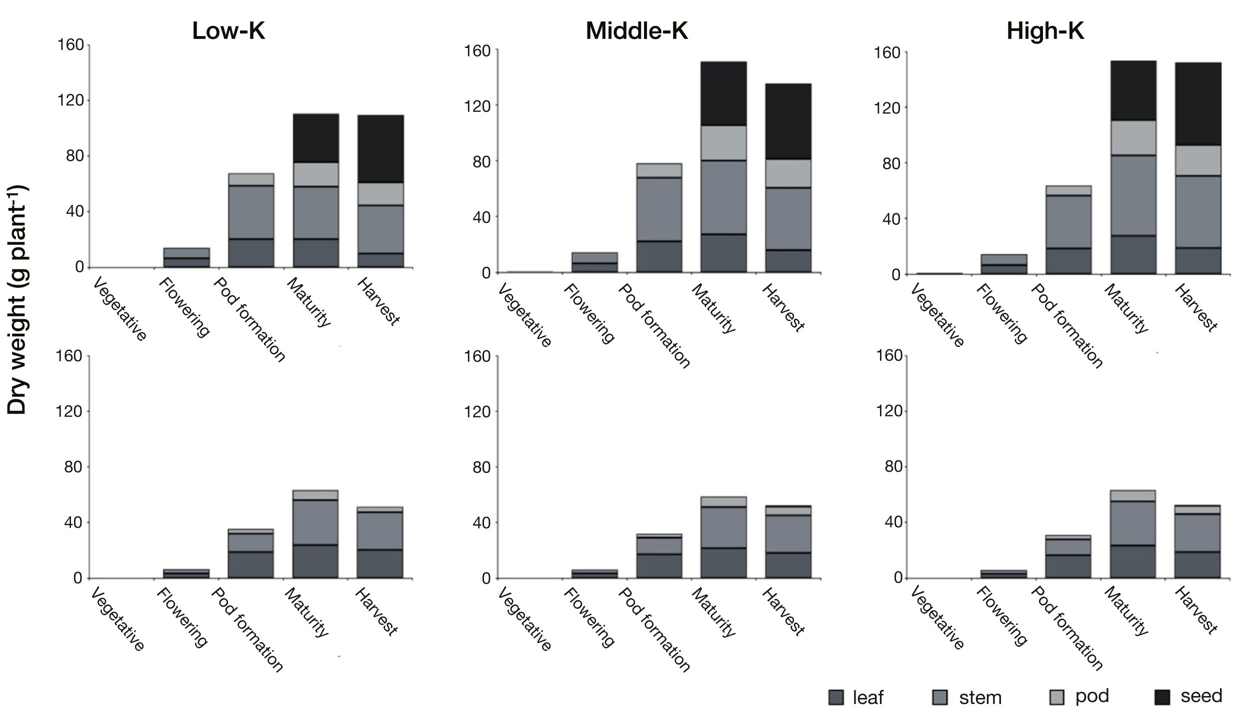

Fig. 1. Successive change in dry weight in fields A (upper) and B (lower) in 2019.

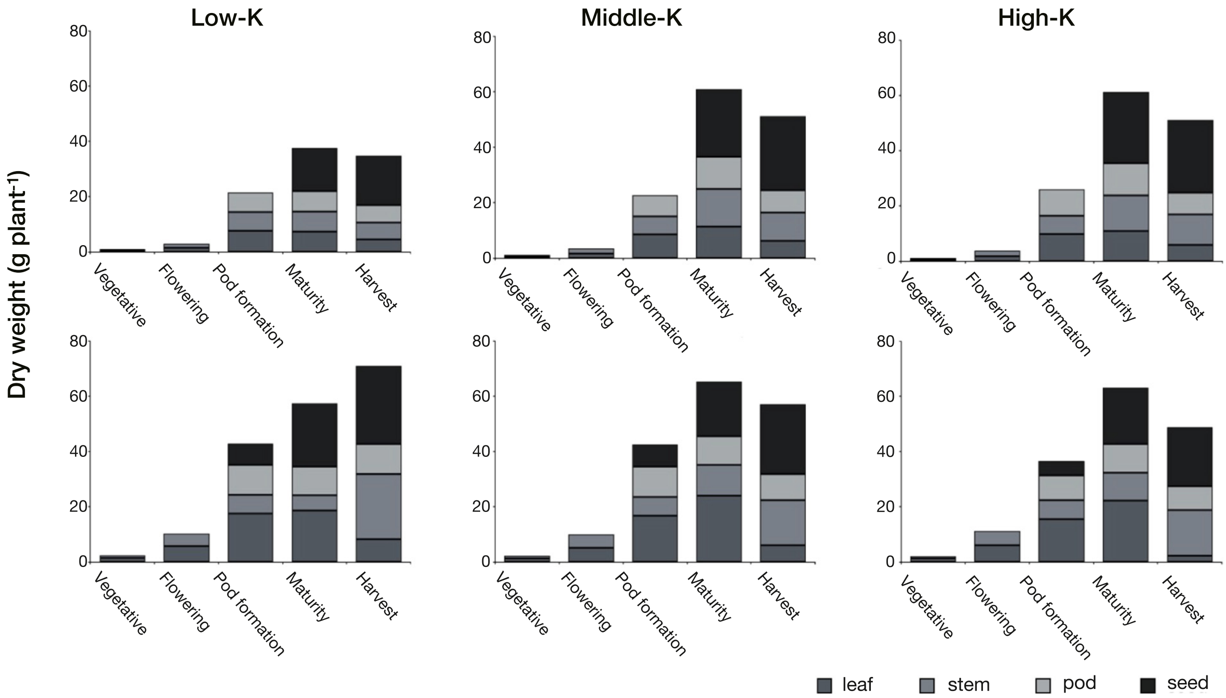

Fig. 2. Successive change in dry weight in fields A (upper) and B (lower) in 2020.

in Field A after the maturation stage, especially in the Middle-K and High-K treatments. In Field B, a severe attack by insects that caused almost all of the harvest was lost (Fig. 1). It was suggested that the farmers did not start cultivation even though the fields, including the experimental field, were decontaminated by the government, which made the experimental field surrounded by nonmanaged areas where weeds invaded; thus, the amount of insects increased. In 2020, the low productivity in the Low-K treatment was confirmed in Field A, but the difference among the K treatments was not seen in Field B.

3.2. ^{137}Cs concentration of plants and soil

In Field A, the ^{137}Cs concentration of shoots in the Low-K plot was significantly higher ($p < 0.05$) than those in the Middle-K and High-K plots (Table S2), and it increased with growth only in the Low-K plots (Fig. 3). In Field B, the difference in ExK content among the K plots was small and decreased gradually with growth (Fig. 5). In the case of ^{137}Cs, the concentration was high in the Low-K and subsequently Middle-K and High-K plots. The concentration gradually decreased, especially in the Low-K plots, but the change was very small in High-K 2019 and in Middle-K and High-K 2020.

The soil ^{137}Cs concentration was constant in both fields regardless of the K level or sampling time (Fig. 4). It was higher in Field B than in Field A, and as the standard deviation values were higher in Field B, the RCs concentration is considered to be more homogenous in Field A.

3.3. Different K forms in the soil

ExK was high in the High-K plots, followed by Middle-K, and Low-K was the lowest in both fields and higher in Field B than in Field A, regardless of

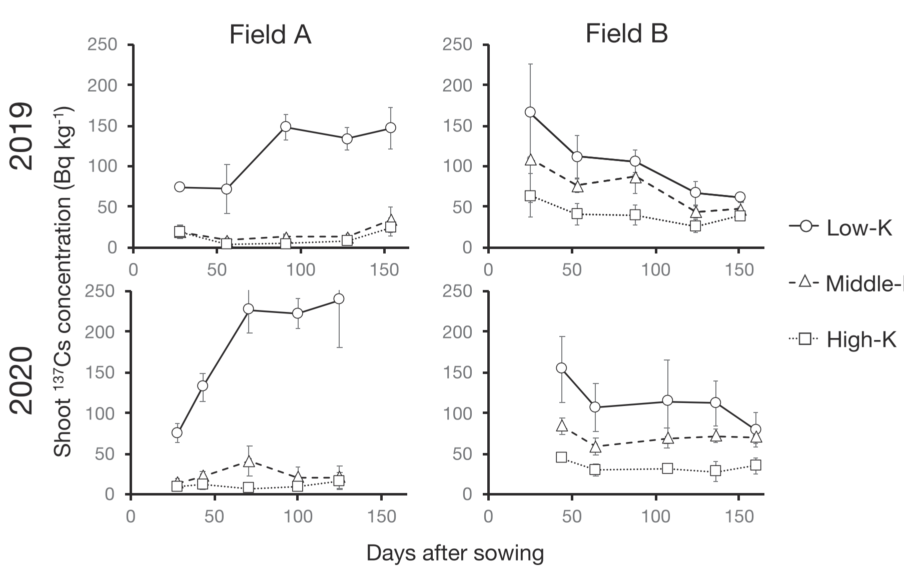

Fig. 3. Successive change in shoot RCs.

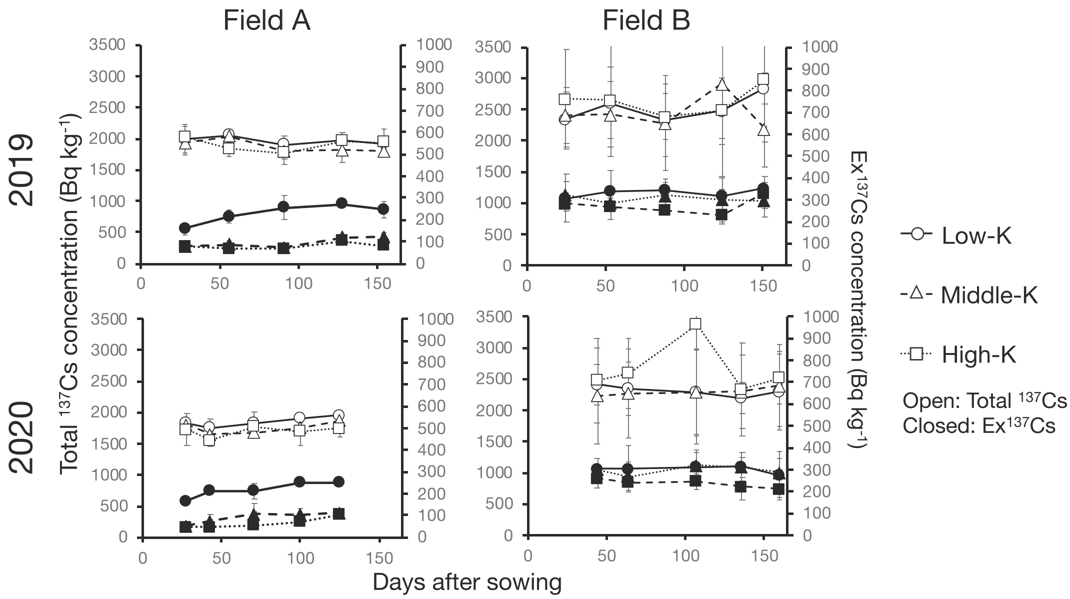

Fig. 4. Successive changes in the total ^{137}Cs concentration and exchangeable ^{137}Cs concentration (Ex^{137}Cs) in the soil.

the year (Fig. 5). The concentration was rather stable in Field A and gradually decreased in Field. B. On the other hand, WS-K was very low in both fields regardless of the year or the K level. The seasonal change was very small (Fig. 6). Nonexchangeable K (NexK) was determined by two methods. In Field A, NexK-HNO_3 was not different among K treatments, and the value was stable in 2019 and decreased at harvest in 2020 (Fig. 7). In Field B, the level was higher than that in Field A, the difference among the treatments was not clear, and the value was slightly decreased at harvest in 2019. In Field A, the NexK-TPB of the High-K plots was slightly higher than that of the other K plots, and the value increased gradually with growth (Fig. 7). The trend with growth was also shown in Field B, but a difference among the treatments was not observed.

3.4. Transfer factor of RCs

In Field A, the TF-^{137}Cs of shoots was higher in the Low-K plot than in the other K plots throughout growth (Fig. 8). The difference between Middle-K and High-K was obscure. It increased from the vegetative growth stage to the pod-forming stage and then peaked in the Low-K plot. The difference among the growth stages was not clear in the other plots. In Field B, TF-^{137}Cs was in the order of the Low-K, Middle-K, and High-K plots. It decreased gradually in both years, and the tendency was clearer in the Low-K plot. The detail of these data is shown in Table S2.

4. Discussion

It is known that most dissolved RCs are selectively retained by FES and then fixed into mica interlayers (Nakao et al., 2008), and as the selectivity to Cs ions is very high (Cremers et al., 1988; Delvaux et al., 2000), most of the dispersed Cs in ion form from the accident is considered to be fixed to this site in a sparsely available or unavailable form (Yin et al., 2017), while it is also known that a part of the RCs can be a weakly adsorbed exchangeable fraction in the readily available form (Sanchez et al., 2002).

Cs uptake by plants is regulated by the transmembrane ion transporting system of plants, including potassium transporters (Noda and Furukawa, 2018). The ability to take up Cs from solution to plant organs is thus

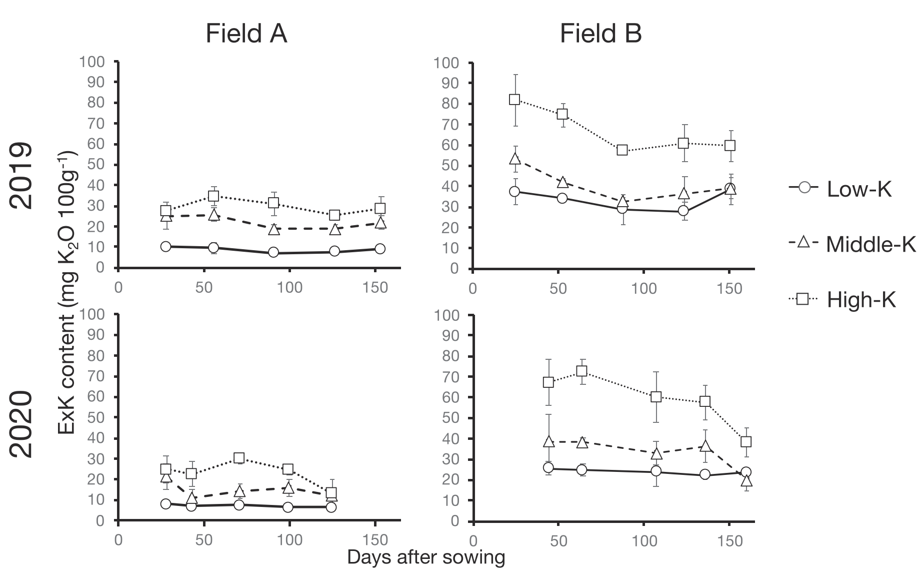

Fig. 5. Successive change in exchangeable K (ExK).

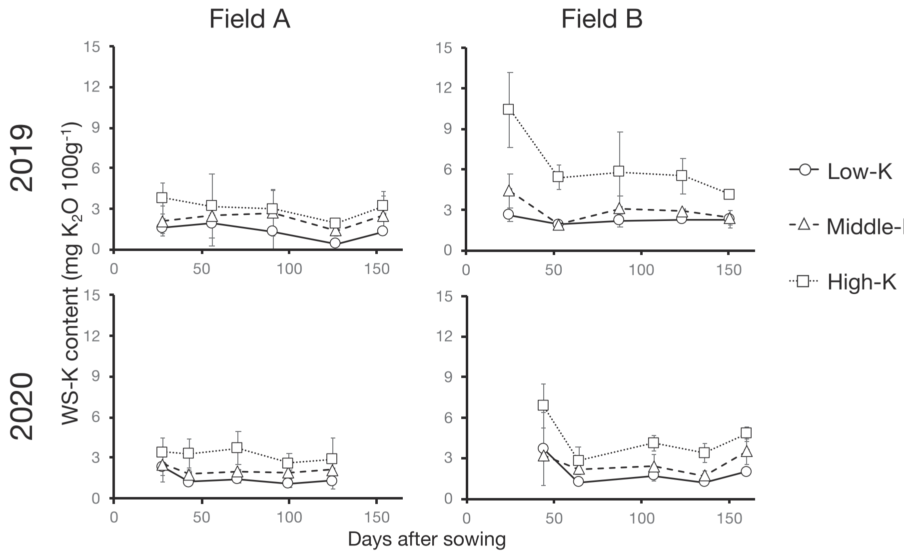

Fig. 6. Successive change in water-soluble K (WS-K).

dependent on the physiological characteristics of the plant and defined as the concentration factor (CF) (Absalom et al., 1999). The CF decreases with the increase in K^+ in the solution (Show and Bell, 1991; Smolder et al., 1996; Zhu and Smolder, 2000). The decrease in the CF by an increase in K^+ directly decreases TF, which is repeatedly confirmed by the development of a countermeasure of the application of a large amount of K fertilizer after agricultural field RCs contamination (e.g., IAEA, 2009, 2020; Yamamura et al., 2018).

The relationship between ExK and TF is shown in Fig. 9. There was a clear different tendency between Field A and Field B, regardless of year or sampling stage; that is, TF was higher in Field B at the same ExK level. This suggests that there are other factors besides ExK that define TF, and one of the main factors is the clay mineral composition (i.e., vermiculite and/or smectite). Soil with a relatively large amount of vermiculite has the ability to strongly retain RCs, which decreases the transferability of RCs from soil to plants even at the same ExK level (Kato et al., 2015). On the other hand, when the Andosol with rich amorphous minerals is dominant, the retention to the soil is low because RIP of Andosol is mostly dominated by short range ordered minerals (e.g., allophane and imogolite) (Vandebroek et al., 2012). These soil characteristics have been considered based on the difference by the soil mineralogy (i.e., clay content, clay types, etc., and used for predicting the model of RCs transfer from soil to plants (Absalom et al., 1999, 2001). The mineralogy of each field soil is shown in Table 2. It is shown that the presence of illite was much higher in Field A than Field B. Since FES is the boundary between vermiculite and mica structures, RIP is highly dependent on the content of mica and the abundance of FES in mica (Nakao et al., 2015). Because the mica content was high in Field A, the RIP may also be higher, which would be expected to result in a lower TF-^{137}Cs. In Field B, the presence of halloysite is much higher than in Field A. Halloysite has been reported to be highly selective for K^+ (Joussein, 2016; Takahashi et al., 2018). Therefore, one of the possibilities to contribute to the higher NexK in Field B may be the amount of halloysite. However, in Field B, the amount of NexK (both NexK-HNO_3 and NexK-TPB) did not result in a great reduction in TF-^{137}Cs. As Ogasawara et al. (2020) mentioned, differences in mineral composition give rise to K release and Cs fixation, and our results also confirmed this. Further research on how the different minerals contribute to K and Cs availability should be investigated in the future.

Soil K availability can be distinguished not only by ExK; WS-K is considered to represent the ready-to-use form of K (Sharpley and Kamprath, 2008), and the relationship between WS-K and TF is shown in Fig. 10. The difference between the two soils was diminished. It is suggested that

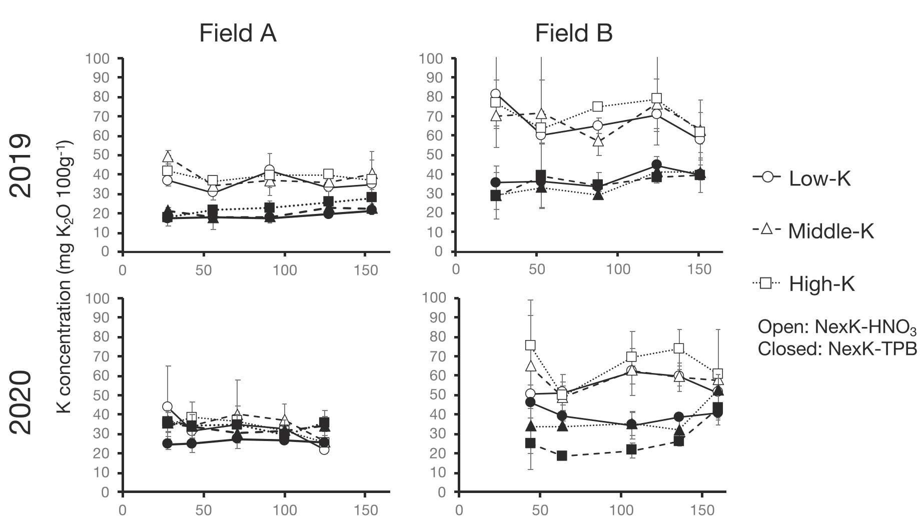

Fig. 7. Successive change in nonexchangle K by HNO_3 (NexK-HNO_3) and TPB (NexK-TPB).

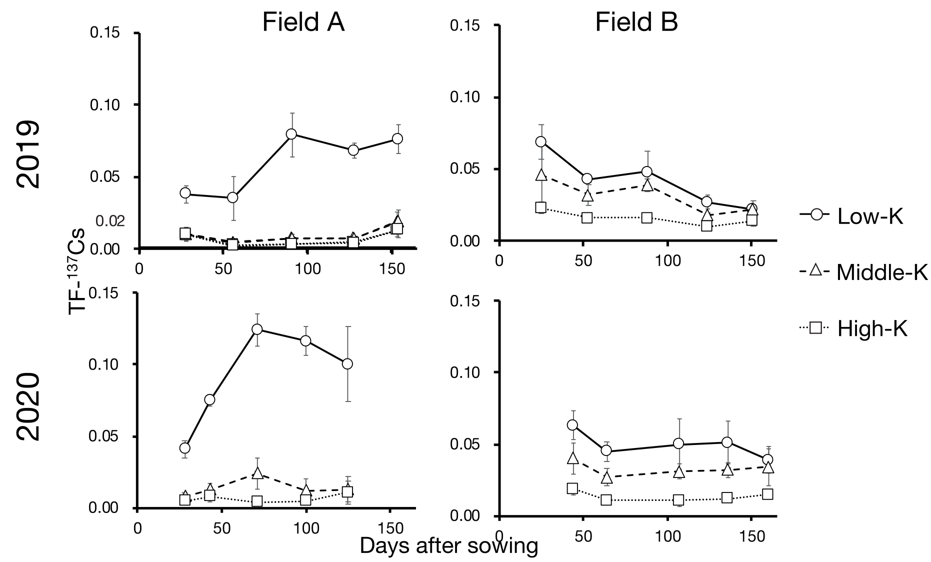

Fig. 8. Successive change in the transfer factor of ^{137}Cs (TF-^{137}Cs).

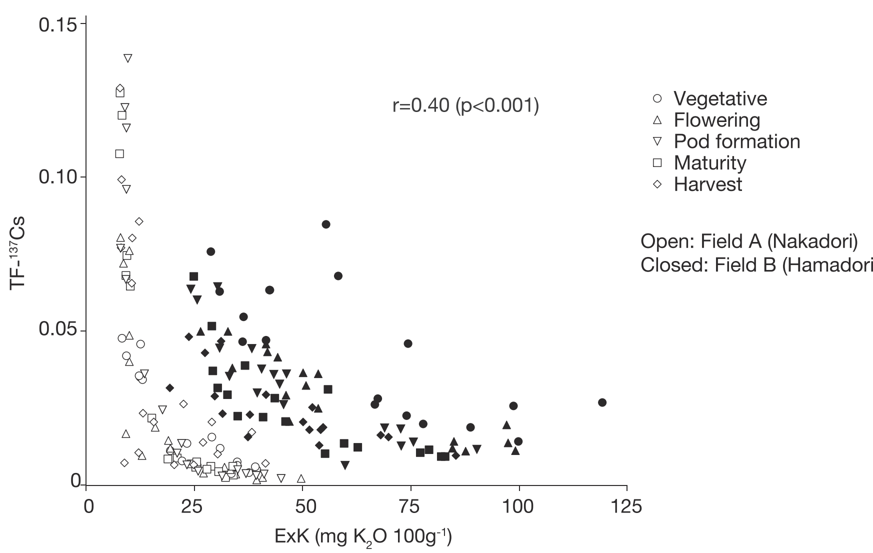

Fig. 9. Relationship between exchangeable K (ExK) and the transfer factor of ^{137}Cs (TF-^{137}Cs).
Spearman's rank correlation coefficient was indicated as r. Data are combined for two years of experiments.

WS-K more likely represents the plant available form of K in the soil, and the evaluation of Ex-K based on the difference by the soil types seems to be diminished. From paddy field research, Kurokawa et al. (2020) found that NexK contributed to the availability of K transfer from paddy soil to brown rice. Eguchi et al. (2021) searched for the relationships between plant ^{137}Cs uptake and soil K (WS-K, ExK and NexK-TPB (PhK-TPB)). We also noted the importance of WS-K as an indicator of the actual K pool for immediate plant uptake. Then, we analyzed NexK by two methods, the hot HNO_3 and TPB methods. The former is considered to be more destructive, and the value of K was relatively higher than those of NexK-TPB (Fig. 7). The other important point is that the range of the data is larger in Nex-HNO_3 than in NexK-TPB. However, the relationship between each NexK and TF was not clear (Fig. 11). It is considered that the sum of ExK and NexK represents Phytoavailable K, and the relationship between TF is plotted in Fig. 12. The relationship was negatively correlated, but the difference between Field A and Field B was still observed.

Thus, at least in the field of soybean, it is considered that phytoavailability of K is not the single determinant factor to explain the difference in RCs transferability of the soils. As Cs uptake is regulated by the concentrations of Cs and K at surface of the plant root, Cs

Table 2
The mineralogy of soil in both fields (wt%).

	Mica(Tri)	Illite	Muscovite	Total mica	Quartz	K-feldspar	Plagioclase	Dolomite	Vermiculite	Halloysite	Kaolinite
Field A	0.00	2.61	0.04	2.65	14.12	9.97	11.91	0.22	0.00	9.93	0.37
Field B	0.00	0.95	0.00	0.95	12.69	10.56	7.87	0.30	0.00	14.31	4.43

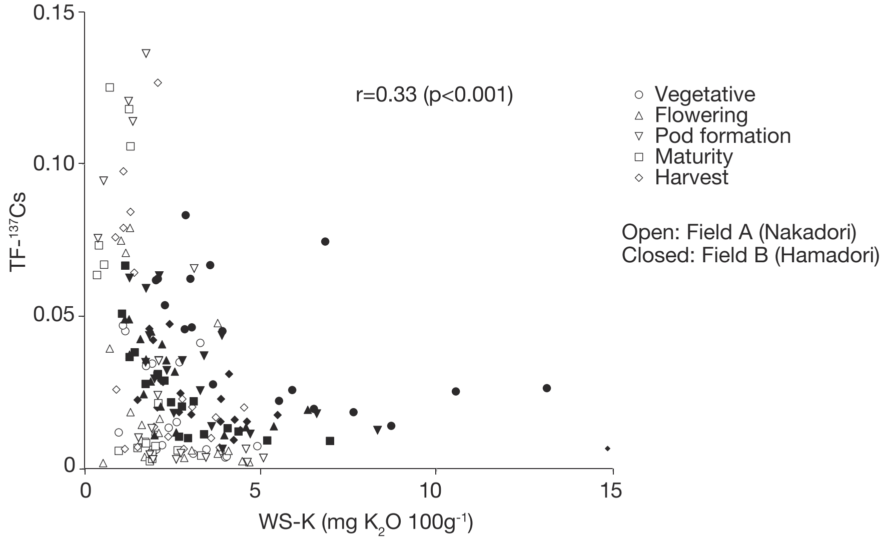

Fig. 10. The relationship between the water-soluble fraction of K (WK) and transfer factor of ^{137}Cs (TF-^{137}Cs). Spearman's rank correlation coefficient was indicated as r. Data are combined for two years of experiments.

behavior becomes important. Several reports have pointed out the importance of exchangeable RCs to explain the transferability of RCs from soil to plants (Kondo et al., 2015; Yagasaki et al., 2019a, 2019b). We analyzed Ex^{137}Cs to evaluate phytoavailable ^{137}Cs, and the relationship between the ratio of plant ^{137}Cs to Ex^{137}Cs (CR-Ex^{137}Cs) and ExK is plotted in Fig. 13.

CR-Ex^{137}Cs is designated by the following equation:

$$CR-Ex^{137}Cs = \frac{^{137}Cs\ \text{concentration in shoot}\ \left(Bq\ kg^{-1}\ DW\right)}{Ex\ ^{137}Cs\ \text{concentration in soil}\ \left(Bq\ kg^{-1}\ DW\right)}$$

It is shown that the difference between different soils is decreased, and it can be explained in a single relationship. However, the contribution of stable Cs and other interfering ions, such as Na^+ and NH_4^+, should be considered to improve the prediction. From these results, it is considered that CR-Ex^{137}Cs rather than TF-^{137}Cs more clearly reflects the competition between K and ^{137}Cs absorption by plants.

The uptake of ^{137}Cs by plants from soil has been described by the sorption-desorption reaction of soil and the physiological mechanism to uptake ^{137}Cs from the soil solution (CF: concentration factor) (Smolder et al., 1997). In the present study, WS-K represents soil solution K, and WS-K is also a good indicator to predict TF among different soils (Fig. 10). However, a simpler and more stable analytical method is required to evaluate and determine high-risk soil (in the meaning of higher transferability of ^{137}Cs from soil to plant).

Based on Fig. 13, plant ^{137}Cs to Ex^{137}Cs was negatively regulated by phytoavailable K (regardless of ExK or ExK plus NexK, data not shown), and the difference between the soils became small. This indicates that the difference in the soil is regulated by the ratio of Ex^{137}Cs to total ^{137}Cs, which is an important factor.

To reveal the mechanism of the transfer of ^{137}Cs from soil to plants, Yoshikawa et al. (2020) pointed out the importance of the existence ratio between ^{137}Cs, K^+ and NH_4^+ in the soil solution based on observations for two years of cultivation after the FDNPP accident. Furthermore, the availability of mineral nutrition in the soil is critical for plant uptake, and thus, it has been established to evaluate the available fraction of mineral nutrition. A similar approach for the availability of Cs has been investigated. Kondo et al. (2015) demonstrated the importance of the ratio of exchangeable Cs to exchangeable K regardless of stable or radioactive Cs to explain the Cs content of brown rice. A similar tendency was also reported by Hirayama et al. (2022) in soybean seeds. Yagasaki et al. (2019a, 2019b) found that the exchangeable RCs concentration distribution in the contaminated area is an important factor in determining the RCs concentration of brown rice as well as exchangeable K from a survey of approximately 300 field data. We also confirmed the importance of Ex^{137}Cs in regulating

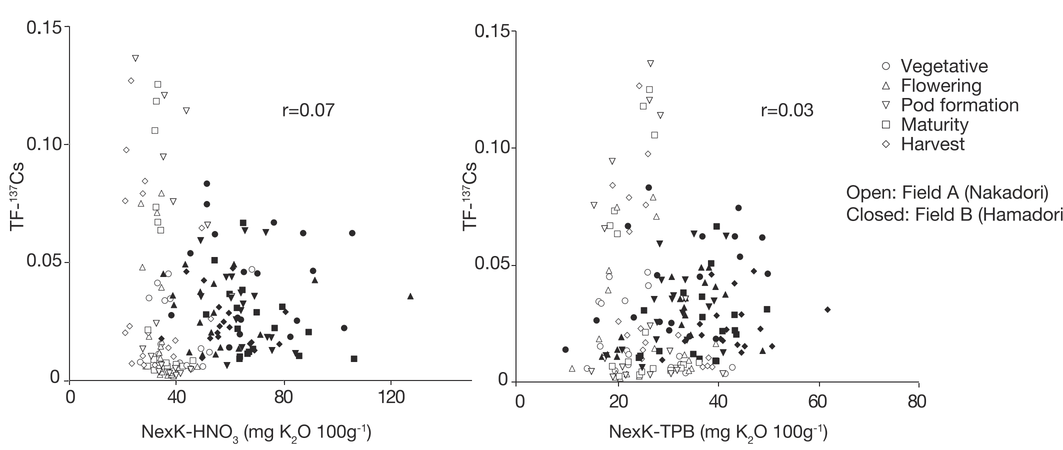

Fig. 11. The relationship between nonexchangeable K (NexK) and the transfer factor of ^{137}Cs (TF-^{137}Cs). Spearman's rank correlation coefficient was indicated as r. Data are combined for two years of experiments.

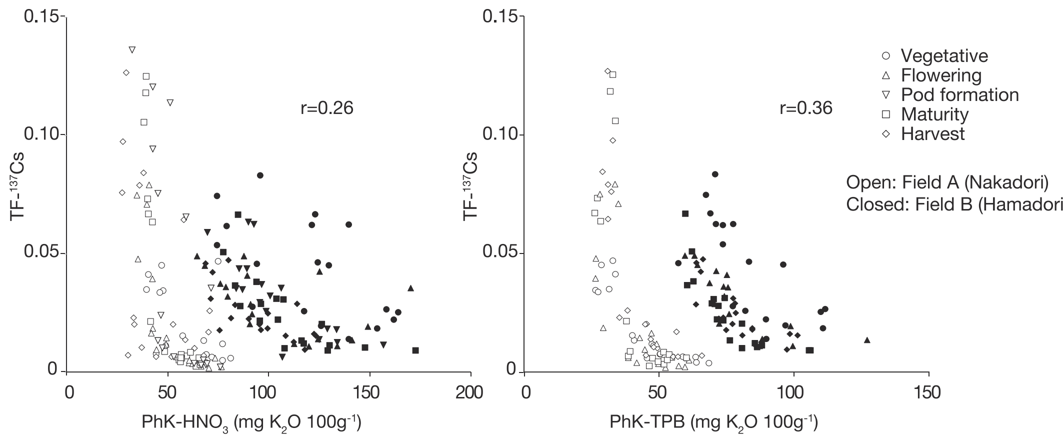

Fig. 12. The relationship between phytoavailable K (PhK) and the transfer factor of ^{137}Cs (TF-^{137}Cs). Spearman's rank correlation coefficient was indicated as r. Data are combined for two years of experiments.

^{137}Cs transfer from soil to plants, and the introduction of the ratio of plant ^{137}Cs to Ex^{137}Cs could be a substitute value instead of TF to determine the effect of potassium on ^{137}Cs transferability among different soils.

This research clearly demonstrated the importance of investigating the mechanism regulating Ex^{137}Cs to total ^{137}Cs which has been raised. It is considered that RCs strongly retained by FES and mica interlayers, which is hardly available to plants, and some of the RCs can also be captured by the available site (e.g., RES fraction (Sanchez et al., 2002) and/or other sites that may loosely bind RCs). If the behavior of RCs in the soil minerals changes more dynamically by the plant species, aging, surrounding nutrient conditions, etc., further research is required to discuss how the RCs exist and change their availability to plants, especially in different environments. Furthermore, we have confirmed the importance of Ex^{137}Cs and also observed the decrease of Ex^{137}Cs level by the application of K (Fig. 4), which indicates that the K has the ability to change the Cs fixation capacity of minerals. As the selectivity for Cs at the FES site is very high (1000:1 in the case of K, Wauters et al., 1996), it is considered that the RCs availability is readily decreased with fixation. On the other hand, even after 10 years, there is a clear difference in the ratio of exchangeable RCs to total RCs of soil, as shown in this research, which indicates that a substantial amount of RCs was not fixed to the FES site. Although the soil RCs concentration can be predicted by using the air dose rate, we do not have any conventional method to predict the ratio of ExRCs to RCs in the soil. It is necessary to monitor the ExRCs level to have a more precise application level of K to the soil.

5. Conclusion

To mitigate RCs transfer from the soil to soybean plants, the ExK level is a critical factor in regulating TF. We compared two soils where the relationship between ExK and TF was different. The WS-K level is a good indicator to predict TF between different soils, while there is difficulty in the practical analysis of soil water under upland conditions. We introduced the relationship between ExK and the ratio of plant RCs concentration to soil ExRCs concentration and found that the difference between soils was greatly decreased. This indicates the importance of RCs availability as expressed by ExRCs. The importance of monitoring the ExRCs level was discussed. Although this study only proposes this indicator, we would like to consider developing a simplified method that could eventually replace this indicator.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.159208.

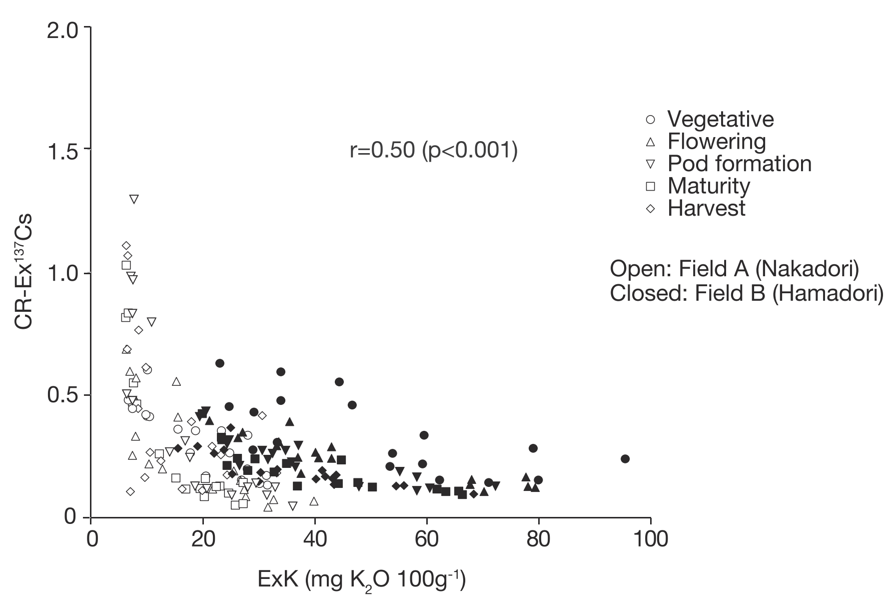

Fig. 13. The relationship between exchangeable K (ExK) and the concentration ratio of plant ^{137}Cs to exchangeable ^{137}Cs (CR-Ex^{137}Cs). Spearman's rank correlation coefficient was indicated as r. Data are combined for two years of experiments.

CRediT authorship contribution statement

Masataka Suzuki: Data analysis of plant, Writing-original draft preparation. Tetsuya Eguchi: Methodology, Reviewing. Kazuki Azuma: Data analysis of soil. Atsushi Nakao: Data curation of soil analysis. Katashi Kubo: Cultivation of plant. Shigeto Fujimura: Management of the fields. Muhamad Syaifudin: Data analysis of soil. Hayato Maruyama: Sampling. Toshihiro Watanabe: Methodology, Data curation of plant analysis. Takuro Shinano: Conceptualization, Writing-reviewing and editing, Supervision, Funding.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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