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Effectiveness of non-exchangeable potassium quantified by mild tetraphenyl-boron extraction in estimating radiocesium transfer to soybean in Fukushima

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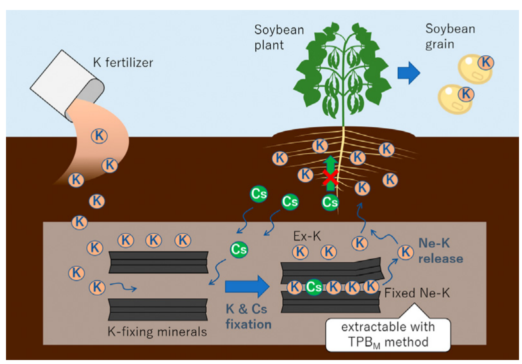
HIGHLIGHTS

- Ne-K content analysis by 3 methods showed varied patterns in Fukushima soils.
- Ne-K by TPB_M method was the most correlated with radiocesium transfer to soybean.
- TPB_M-Ne-K content increased, reflecting K fixation, causing radiocesium fixation.
- Fixed K repressed radiocesium transfer to soybean through fixation and K supply.
- TPB_M extraction method effectively estimated radiocesium transfer risk to soybean.

GRAPHICAL ABSTRACT



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ABSTRACT

Non-exchangeable K released from soil minerals can reduce radiocesium transfer to plants, as well as exchangeable K. We investigated the effect of non-exchangeable K on radiocesium transfer to soybean, and the non-exchangeable K extraction method most suitable for estimating the transfer risk. In Fukushima Prefecture, Japan, 106 soils were collected from 89 soybean fields during 2014–2018 to analyze non-exchangeable K contents using three methods: boiling nitric acid extraction, tetraphenyl-boron extraction, and mild tetraphenyl-boron extraction. The non-exchangeable K contents quantified by the former two methods were dependent on the amount of micas, which are K-bearing minerals. The non-exchangeable K content by mild tetraphenyl-boron extraction depended on the amount of K fertilizer application and K-fixing minerals but not on micas, indicating that it reflects fertilizer K fixed by the minerals. The soil-to-plant transfer factor of radiocesium was most correlated with the non-exchangeable K content by the mild extraction ($r_s = -0.67$). This correlation was also stronger than that between exchangeable K and the transfer factor ($r_s = -0.40$). As non-exchangeable K content increased, the exchangeable radiocesium fraction decreased, indicating that radiocesium was fixed together with K. Additionally, multiple regression analysis indicated that non-exchangeable K by the mild extraction significantly decreased the transfer factor even if the exchangeable radiocesium fraction was kept constant. Thus, the fixed K was considered to repress radiocesium transfer to soybean through both radiocesium fixation and K supply. With the criterion of total extracted K, the sum of exchangeable and non-

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exchangeable K, as 65 mg K_2O 100 g^{-1} by the mild extraction, fields with high and low transfer factors were able to be differentiated more effectively than with a current criterion of exchangeable K as 50 mg K_2O 100 g^{-1}. The results revealed that mild tetraphenyl-boron extraction is effective for estimating radiocesium transfer to soybean.

1. Introduction

Since the accident at the Tokyo Electric Power Company's Fukushima Dai-ichi Nuclear Power Plant (FDNPP[1]) in March 2011, field soils in Fukushima and neighboring prefectures of Japan have been contaminated with radiocesium (RCs[2]; ^{137}Cs, and ^{134}Cs) released from the FDNPP, which can be transferred to agricultural products. Soybean is cultivated in the third largest area after rice and buckwheat in the Fukushima Prefecture. In monitoring inspections after the FDNPP accident in Fukushima Prefecture, a higher percentage of soybean exceeded the standard limit of RCs for general food, that is, 100 Bq kg^{-1} (Ministry of Health, Labor and Welfare, 2012) than cereals (Nihei, 2016; Nihei and Hamamoto, 2019). The percentages in 2012 and 2013 were 2.6% and 1.9% for soybean, 0.007% and 0.003% for rice, and both 0% for wheat. For soybean cultivation, it is important to develop techniques to screen fields with high RCs transfer risk, where countermeasures are needed to produce crops with low contamination.

K fertilizer has been widely applied to reduce RCs contamination in crops. Increasing K^+ concentration in the extracellular solution (soil solution) reduces the influx of Cs^+ into the root cells (Shaw and Bell, 1991; Smolders et al., 1997). It is due to suppressing the expression of high-affinity K transporters that can also carry Cs^+ from the extracellular solution in rice plant (Rai and Kawabata, 2020). Additionally, K fertilization may promote RCs fixation in soil, which may also be a cause to reduce RCs transfer to crops (Kubo et al., 2017). Soil K fertility is generally assessed by quantifying exchangeable K (Ex-K[3]), which is electrostatically adsorbed on the surfaces of minerals and organic matter and extractable with neutral ammonium acetate solution. For soybean cultivation after the accident, the Ministry of Agriculture, Forestry and Fisheries (MAFF[4]) of the Japanese government recommended supplemental K fertilizer application to maintain Ex-K content in soil higher than 25 mg K_2O 100 g^{-1} (Ministry of Agriculture, Forestry and Fisheries, 2015), as well as for rice cultivation (National Agriculture and Food Research Organization, 2012; Kato et al., 2015). However, Ex-K of 25 mg K_2O 100 g^{-1} was sometimes insufficient to reduce RCs transfer to soybean, unlike paddy rice. A survey in 2012 showed that 3.2% of the fields with Ex-K more than 25 mg K_2O 100 g^{-1} produced soybean with exceeding 100 Bq kg^{-1} of RCs concentration in areas where contaminated soybeans were detected in 2011 (Ministry of Agriculture, Forestry and Fisheries, 2015). Accordingly, the recommendation by MAFF included a provision that K fertilizer application should aim to reach Ex-K of 50 mg K_2O 100 g^{-1} for areas where soybeans with high RCs concentrations may be produced. This implies that there are soil factors more influential than Ex-K in impacting RCs transfer from soil to soybean.

A recent study indicated that non-exchangeable K (Ne-K[5]) in soil minerals plays a role in controlling RCs transfer to paddy rice as well as Ex-K (Eguchi et al., 2015, Ogasawara et al., 2019, Kurokawa et al., 2020). Soil K is generally distinguished into four distinct pools, namely solution K, Ex-K, Ne-K, and structural K, in the order of plant availability, with reversible transfer between the pools (Brouder, 2011). Ne-K is stored in the interlayers of micas, which are a group of K-bearing primary minerals, and K-fixing minerals, such as vermiculites (Helmke and Sparks, 1996). It is not easily released by simple exchange with other cations but can be readily diffused into the soil solution and supplied to plants when the K concentration in the soil solution and Ex-K decreased by crop removal and/or leaching. Additionally, the interlayer sites from which Ne-K is released can selectively adsorb and fix RCs, as well as K (Sawhney, 1972; Maes et al., 1999; Ogasawara et al., 2017; Kitayama et al., 2020). It has not been investigated whether Ne-K reduces RCs uptake by crops other than rice. If Ne-K controls RCs transfer to soybean, Ne-K assessment may be a more efficient way to estimate RCs transfer risk, and the amount of K fertilizer application needed to reduce the risk than diagnostics using Ex-K alone.

This study investigated the relationship between Ne-K and RCs transfer to soybean with observations in 89 fields in Fukushima Prefecture, focusing on two perspectives. The first is what method of Ne-K quantification is suitable for estimating RCs transfer risk to soybean. Many chemical extractions with different extraction powers and mechanisms have been proposed for the quantification of Ne-K. Previous studies on RCs transfer to paddy rice have applied the boiling nitric acid extraction method (HNO_3 method[6]; Ogasawara et al., 2019, Kurokawa et al., 2020) and tetraphenyl-boron (TPB[7]) extraction methods (Eguchi et al., 2015). Both methods are preferable for convenient assessment of RCs transfer risk because they consume less time and cost compared with other methods (e.g., resin extraction (Barber and Matthews, 1962) and sequential hydrochloric acid extraction (Moritsuka et al., 2003)). This study compared Ne-Ks quantified by the HNO_3 method and two kinds of TPB methods with respect to their characteristics and the correlation with RCs transfer to soybean. The second perspective is to take into account the correlation between Ne-K and RCs fixation in soil. Although Ne-K is generally regarded as an index of K supply other than Ex-K, it is also closely related to soil functions causing RCs fixation (Kurokawa et al., 2019). Previous studies have not analyzed whether the effects of K supply, RCs fixation, or both were responsible for the correlation between Ne-K and RCs transfer to crops. Using multiple regression analysis, this study analyzed the contribution of Ne-K on RCs transfer to soybean statistically separated from the correlation between Ne-K and RCs fixation. Finally, we proposed a method to estimate RCs transfer risk to soybean more effectively than the conventional diagnosis method by Ex-K.

2. Materials and methods

2.1. Sample collection

Soil samples and soybean grains were collected from 89 fields in the Fukushima Prefecture (Fig. 1). The altitude of the fields ranged from 0 to 630 m above sea level (Appendix Table A1). The mean annual temperatures and mean annual precipitations during 2014–2018 ranged between 10.4 °C and 13.8 °C and 1030 mm and 1590 mm, respectively in 15 meteorological stations near the fields (Japan Meteorological Agency, 2021). For the 84 fields managed by farmers (F1–F84 in Appendix Table A1), soil sampling and radiometry of products were conducted from 2014 to 2018 in a monitoring project of MAFF. Soybeans were experimentally cultivated in the other five fields. Two of the five fields were uniformly managed without K fertilizer application in 2018 (E1 and E2 in Appendix Table A1). The other three fields (D1, D2, and KH) were managed under experimental segmentations with different K fertilization strategies, as follows.

The D1 and D2 fields underwent decontamination by scraping surface soil of at least 3 cm depth and soil mixing with uncontaminated earthy materials (Ministry of Agriculture, Forestry and Fisheries, 2013). In these fields, soybean was cultivated in 2017 and 2018 and divided into three plots in three replications with randomized design: one plot with conventional fertilizer application (NPK mixed fertilizer including 20% of K_2O; 6 g K_2O m^{-2} for the D1 field, 10 g K_2O m^{-2} for the D2 field) alone, and the two plots with additional application of potassium

[1] FDNPP: Fukushima Dai-ichi Nuclear Power Plant
[2] RCs: radiocesium
[3] Ex-K: exchangeable K
[4] MAFF: the Ministry of Agriculture, Forestry and Fisheries
[5] Ne-K: non-exchangeable K
[6] HNO_3 method: boiling nitric acid extraction method
[7] TPB: tetraphenyl-boron

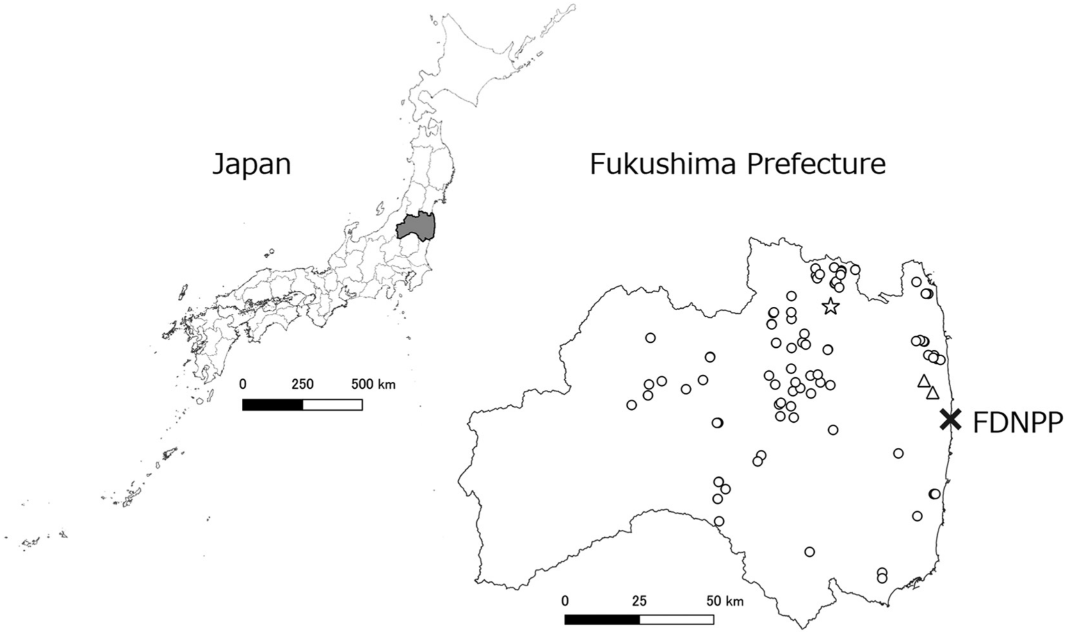

Fig. 1. The locations of study fields Star shows the location of an experimental field with eight plots with different K fertilizer management histories (KH). The two triangles show the locations of the experimental decontamination fields with three plots with different K fertilizer protocols (D1 and D2). The circles show the locations of the remaining 86 study fields.

chloride (KCl), aiming to achieve Ex-K content to the desired values (25 and 50 mg K_2O 100 g^{-1} for the D1 field, and 50 and 75 mg K_2O 100 g^{-1} for the D2 field) (Appendix Fig. A1). The amounts of KCl application were adjusted to the K differences between the desired Ex-K contents and the sum of the Ex-K contents in the last fall plus K input by conventional fertilizer application, assuming a topsoil weight of 150 kg m^{-2}.

The KH field was divided into eight plots of different K fertilizer application histories in three replications with randomized design (Appendix Fig. A1). The field was planted with soybeans in 2018 and prior to that buckwheat was annually cultivated since 2013. One plot received no K fertilizer since 2013. In the other three plots, amounts of K fertilizer (potassium sulfate; K_2SO_4) application were annually adjusted aiming to achieve Ex-K content to the desired values (25, 45, 65 mg K_2O 100 g^{-1}, respectively); the application amounts were adjusted to the difference between the desired Ex-K contents and the ones measured in advance (about 1 month before fertilizer application), assuming a topsoil weight of 150 kg m^{-2}. The remaining four plots were managed in the same way as the above four plots, during 2013–2014, and since 2015, they underwent conventional management where the amounts of K_2SO_4 application were 3 and 10 g K_2O m^{-2} for buckwheat and soybean, respectively. In this study, samples of soybean and soil samples from 2018 were used.

In each field, soybean was sown from early June to mid-July and harvested from early October to early November. For soybean grains and soils of the plowed layer, five subsamples were harvested and mixed to prepare a composite sample for each field or plot replication. For experimental plots, samples from the three replications were also composited to one. In the D1 and D2 fields, six sample sets of soils and crops were collected from the three plots for the two years. In the KH field, eight sample sets were collected from eight plots. In the remaining 86 fields, one sample set was collected per field. Therefore, 106 sample sets were obtained. The samples of soybean grain and soil were air dried, and the soil samples were ground to pass through 2 mm sieves for radiometric and chemical analyses. The moisture content in these air-dried samples was measured and used to correct the analytical values to a weight base of 15% moisture status (soybean grain) or dry matter (soil).

2.2. Radiometric and chemical analyses

The samples of soybean grain and soil were uniformly packed into 2.0 L and 100 mL plastic containers, respectively. Concentrations of ^{137}Cs were determined using Ge gamma-ray detectors (GC2520-7500SL, GC4020-7500SL, or GCW2523-7905-30U-ULB, Canberra, Meriden, USA). The observed ^{137}Cs concentrations were decay-corrected to the harvest dates of the samples. The ^{137}Cs concentrations in soybean grains at 15% moisture content ranged from 0.4 to 170 Bq kg^{-1}. The ^{137}Cs concentrations in soils as dry matter ranged from 53 to 5950 Bq kg^{-1}. The soil-to-plant transfer factor (TF[8]) was determined as follows:

$$\mathrm{TF} = \frac{^{137}\mathrm{Cs\ concentration\ of\ soybean\ grain}\ (\mathrm{Bq\ kg^{-1}, 15\%\ moisture\ content})}{^{137}\mathrm{Cs\ concentration\ of\ soil}\ (\mathrm{Bq\ kg^{-1}, dry\ matter})} \tag{1}$$

The exchangeable fraction of ^{137}Cs (Exf-^{137}Cs[9]) was also analyzed as an index of RCs fixation in soil. Exchangeable ^{137}Cs was extracted by shaking with 1 mol L^{-1} ammonium acetate (CH_3COONH_4) solution for 1 h at a solution/soil ratio of 10 mL g^{-1}. The ^{137}Cs concentration in the extract was quantified using Ge gamma-ray detectors in a manner similar to that of the grain samples. Exf-^{137}Cs were determined using the following equation:

$$\mathrm{Exf}-^{137}\mathrm{Cs}\ (\%) = \frac{\mathrm{Exchangeable}\ ^{137}\mathrm{Cs\ concentration}\ (\mathrm{Bq\ kg^{-1}})}{^{137}\mathrm{Cs\ concentration\ of\ soil}\ \left(\mathrm{Bq\ kg^{-1}}\right)} \times 100 \tag{2}$$

Ex-K was extracted in the same way as exchangeable ^{137}Cs and measured using flame spectrophotometry and atomic absorption spectrophotometry. For the 84 fields surveyed in the monitoring project, unpublished datasets containing TF, Exf-^{137}Cs, and Ex-K were provided by MAFF.

Ne-K content was quantified by three methods: HNO_3 method (Helmke and Sparks, 1996), TPB extraction method slightly modified from Carey et al. (2011) (TPB_C method[10]), and the newly developed mild TPB extraction method (TPB_M method[11]). The HNO_3 method extracts Ne-K by

[8] TF: soil-to-plant transfer factor
[9] Exf-^{137}Cs: exchangeable fraction of ^{137}Cs
[10] TPB_C method: TPB extraction method slightly modified from Carey et al. (2011)
[11] TPB_M method: mild TPB extraction method

decomposing the lattice structure of minerals, and it is the most commonly used method for Ne-K extraction. The TPB methods promote the diffusive release of Ne-K from the interlayers of minerals by reducing the K concentration in the contact solution through the formation of potassium TPB (KTPB) precipitates. Therefore, TPB extraction methods can mimic the action of plant roots by the depletion of K in soil solution (Cox et al., 1999).

In the HNO_3 method, K was extracted by heating a 2.5 g soil sample with 25 mL of 1 mol L^{-1} nitric acid (HNO_3) and filtering through filter paper. The K concentration in the extract was determined using atomic absorption spectroscopy (AA-6200, Shimadzu, Kyoto, Japan).

In the TPB_C method, a 1 g soil sample was extracted with 3 mL of combined reagent of 0.1 mol L^{-1} sodium TPB (NaTPB)/1.7 mol L^{-1} sodium chloride (NaCl)/0.01 mol L^{-1} disodium ethylenediaminetetraacetic acid (Na_2EDTA) for 4 h at 20 °C. The reaction was halted using 10 mL of 0.5 mol L^{-1} ammonium chloride (NH_4Cl). The suspension with addition of 1.5 mL of 0.5 mol L^{-1} cupric chloride ($CuCl_2$) was boiled gently for 30 min to dissolve KTPB precipitate and cooled for 5 min. After adding 1 mL of 3 mol L^{-1} hydrochloric acid (HCl) to the suspension, the volume was brought to 50 mL with deionized water. Immediately, it was filtered through a 0.4 μm membrane filter (PP025045, Osaka Chemical ind. Co. Ltd., Osaka, Japan). The K content in the extract was determined by atomic absorption spectroscopy (ZA3000, Hitachi High-Tech Science, Tokyo, Japan).

In this study, milder TPB extraction was also performed to selectively extract the highly diffusible portion of Ne-K, which may be closely related to plant-available K although it cannot be released by a simple ion-exchange reaction (Cox et al., 1999; Li et al., 2018; Wang et al., 2010). The extraction power of TPB methods can be controlled by the Na^+ concentration (Wang et al., 2010) and extraction time (Scott et al., 1960; Wang et al., 2010) and Ne-K released by the weaker extraction reaction is readily available for plants (Cox et al., 1999: Li et al., 2016). In the TPB_M method, NaCl was not included in the extractant, and the extraction time was shortened to 5 min. The other treatments were the same as those used in the TPB_C method.

Because each method extracts both Ex-K and Ne-K, the Ne-K content was calculated by subtracting the Ex-K content from the total extracted K (TEr-K[12]) content as follows:

$$\mathrm{Ne-K}\ \left(\mathrm{mg\ K_2O\ 100\ g^{-1}}\right) = \begin{cases} \mathrm{TEr-K{-}Ex-K}\ (\mathrm{TEr-K} > \mathrm{Ex-K}) \\ 0\ (\mathrm{TEr-K} \leq \mathrm{Ex-K}) \end{cases} \quad (3)$$

2.3. Statistical analyses

R v.3.1.1 software (R Core Team, 2019) was used for statistical analyses. Spearman's rank correlation (r_s[13]) was analyzed between TF and soil parameters. To analyze the effect of K fertilization on soil K parameters, linear regression analysis was performed between the average amount of K fertilizer application during 2013–2018 and the Ex-K and Ne-K contents for the KH field.

Multiple linear regression analysis was performed to analyze the relative contributions of soil parameters on RCs transfer to soybean. The Shapiro-Wilk test indicated that TF and all soil parameters were not normally distributed, and these values were log-transformed to reduce positive skewness before multiple regression. For the data of Ne-K, which contained zero values, 1 was added to the value of all samples before log-transformation. The variance inflation factors were less than 1.6 in all cases, indicating the absence of severe multicollinearity.

Using the geographical information system software QGIS 3.4, the study fields were divided into two groups: the granitic area group, located on or within 1 km of the granite and granodiorite areas in the geological map (Geological Survey of Japan, AIST, 2017), and the non-granitic area group. The geometric mean values of each Ne-K content +1 were compared between the two groups using the student's unpaired *t*-test. For the *t*-test, single data were used per field by applying the average of each Ne-K for the D1, D2, and KH fields.

The dataset of 84 fields provided by MAFF included X-ray diffraction analysis data of clay size (< 2 μm) soil fraction, in which the intensity of peak (or peak shift by chemical or thermal treatments) reflecting each 2:1 type mineral was classified into four levels (see Table 1). To investigate the relationship between clay types and the Ne-Ks, the soil samples were grouped by the peak intensity of each mineral, and the geometric mean values were compared using Tukey's honestly significant difference test ($\alpha = 0.05$).

3. Results and discussions

3.1. Characteristics of Ne-Ks extracted by three methods

In the KH field, the Ne-K contents determined by the different extraction methods were compared with respect to the relationship with the average K fertilizer application amount for the last 6 years (Fig. 2). In soil without K fertilizer application, HNO_3-Ne-K showed the highest value, followed by TPB_C-Ne-K and TPB_M-Ne-K. The Ne-Ks extracted from the unfertilized soil were mostly considered to be "native" Ne-K, which is naturally contained in the unexpanded interlayer of micas. The HNO_3-Ne-K content did not significantly change with K fertilizer management, and this result was in accordance with the observations in paddy fields in Fukushima Prefecture (Kurokawa et al., 2020). On the other hand, the Ne-K content by TPB methods significantly increased with the amount of K fertilizer application as well as the Ex-K content. These increases are attributed to the K-fixing minerals fixing K applied. The expanded interlayers of vermiculites and weathered (vermiculized) micas can fix K by collapsing when much K has been adsorbed there (Douglas, 1989; Fanning et al., 1989). In addition, the expanded interlayer of smectites can fix K by collapsing after repeated wetting and drying (Šucha and Širáňová, 1991). Applying linear regression, the slope can be interpreted to reflect the K "recently fixed" by these K-fixing minerals, and it was higher in the order of the $TPB_C > TPB_M > HNO_3$ methods. The ratio of slope/intercept, which indicates the extraction ratio of recently fixed K with native Ne-K, was higher in the order of the $TPB_M > TPB_C > HNO_3$ methods, indicating that the variation in TPB_M-Ne-K was relatively heavily dependent on the degree of K fixation/release. These observations in the KH field suggest that HNO_3-Ne-K reflects native Ne-K but not recently fixed K, while the Ne-Ks by the TPB methods, particularly TPB_M-Ne-K, depend on the recently fixed K.

Among all 106 soils, the Ne-K contents of the three methods were significantly and positively correlated with each other, and the correlation was relatively weak between HNO_3-Ne-K and TPB_M-Ne-K (Fig. 3). The HNO_3-Ne-K content was, on average, 80% of the TPB_C-Ne-K content. In 36 soils, the TPB_C-Ne-K content was more than twice as high as the HNO_3-Ne-K content, while the opposite was true in five soils. The TPB_M method extracted 32% of TPB_C-Ne-K on average. The method extracted more than 50% of TPB_C-Ne-K in 15 soils, and less than 10% of that in 16 soils. The discrepancy in the variations between the three Ne-K values is possibly due to the fact that native Ne-K and recently fixed K might be extracted with different proportions between the three methods.

The HNO_3-Ne-K and TPB_C-Ne-K contents showed no significant correlation with the Ex-K content among all soil samples ($r_s = -0.069, 0.007$; Appendix Fig. A2). Independence of HNO_3-Ne-K on Ex-K has been reported (Kitagawa et al., 2018; Kurokawa et al., 2020). The TPB_M-Ne-K content was weakly but significantly positively correlated with the Ex-K content among all soils ($r_s = 0.381, p < 0.001$). Ex-K is the K pool which largely varies depending on the fertilizer application rate, as observed in the KH field (Fig. 2). In the paddy fields in Fukushima Prefecture, the average increase in Ex-K content during 2011–2014 was significantly correlated with the average amount of additional K fertilizer applied as a countermeasure for RCs contamination ($R^2 = 0.71, p < 0.001$) and effective cation exchange capacity ($R^2 = 0.54, p < 0.05$) of the14 district (Nakayama et al., 2019). The correlation of only TPB_M-Ne-K with Ex-K was possibly due to

[12] TErK: total extracted K
[13] r_s: Spearman's rank correlation coefficient

Table 1
Geometric mean (geometric standard deviation) of the Ne-K content for soils classified by the peak intensity of 2:1 type minerals in clay-size fraction.

2:1 type minerals	Peak intensity classes	n	Ne-K content +1 (mg K_2O 100 g^{-1}) HNO$_3$ method			TPB$_C$ method			TPB$_M$ method		
Micas	±	2	7	(1.0)	a	16	(1.2)	a	16	(1.0)	ns
	+	78	42	(2.9)	b	83	(2.1)	b	23	(1.9)	
	+ +	4	295	(1.3)	c	197	(1.5)	c	19	(8.0)	
Vermiculites	−	30	18	(3.0)	a	49	(2.0)	a	17	(1.6)	a
	±	11	42	(1.5)	b	87	(1.7)	b	20	(1.7)	ab
	+	43	85	(2.4)	b	119	(1.9)	b	29	(2.3)	b
Hydroxyl-interlayer vermiculites	−	14	19	(4.7)	a	56	(2.5)	a	16	(2.4)	a
	±	14	26	(2.7)	a	60	(2.0)	ab	18	(1.8)	ab
	+	56	63	(2.5)	b	99	(2.0)	b	26	(2.0)	b
Chlorites	−	11	137	(2.6)	b	157	(1.8)	b	50	(1.9)	b
	±	12	40	(3.9)	a	76	(2.3)	a	19	(2.8)	a
	+	61	37	(2.9)	a	75	(2.1)	a	21	(1.8)	a
Smectites	−	21	61	(3.8)	ns	83	(2.4)	ns	17	(2.3)	a
	±	16	36	(2.7)		70	(2.1)		21	(1.8)	ab
	+[#]	47	41	(3.1)		88	(2.1)		27	(2.0)	b

Because the data of the Ne-K content included 0, 1 was added when calculating the geometric mean. Peak intensity classes: −, no peak; ±, faint peak; +, clear peak; + +, prominent peak. [#]one sample with prominent peak of smectite (+ +) was included in this group.
Different letters indicate significant differences between soils classified by the peak intensity of each mineral according to Tukey HSD test ($\alpha = 0.05$), while "ns" means no significant difference.

that differences in K fertilizer management among the fields influenced the both Ex-K and TPB$_M$-Ne-K but not much the other Ne-K.

With respect to geological conditions, the geometric mean of Ne-K +1 was compared between the granitic and non-granitic area groups. Granitic soils in Fukushima Prefecture are rich in biotite (trioctahedral mica) (Mukai et al., 2014; Ogasawara et al., 2019), which is a native source of Ne-K. HNO$_3$-Ne-K was significantly higher in the granitic area (111 mg K_2O 100 g^{-1} as [Ne-K +1]) than in the non-granitic area groups (31 mg K_2O 100 g^{-1}; Appendix Table A2), consistent with a previous study on paddy fields in Fukushima Prefecture (Kurokawa et al., 2020). In TPB$_C$-Ne-K, the difference between the granitic and non-granitic area groups was relatively small (104 and 75 mg K_2O 100 g^{-1}, respectively) because the non-granitic soils did not show low values as was the case for HNO$_3$-Ne-K. In TPB$_M$-Ne-K, the mean values were almost the same between the granitic and non-granitic area groups (23 and 22 mg K_2O 100 g^{-1}, respectively). The results indicate that having granitic rocks as soil parent materials, and maybe the mica contents, largely influence HNO$_3$-Ne-K, while these factors are not determinants of TPB$_M$-Ne-K.

To investigate the relationship with several kinds of 2:1 type clay minerals, the geometric mean of Ne-K +1 was compared between soils grouped by these peak intensities (Table 1). Every Ne-K was lower along with the peak intensity of chlorites, which interlayers do not contain native Ne-K or fix K because they are completely filled with a hydroxide sheet. Every Ne-K increased with the increase in the peak intensities of vermiculites and hydroxyl-interlayer vermiculites, both of which are weathering products of micas and can fix K in their interlayers. The Ne-K contents of the HNO$_3$ and TPB$_C$ methods were considerably higher along with the peak intensity of mica. In the HNO$_3$-Ne-K content, all four soils showing prominent (+ +) mica peaks were ranked within the top five, indicating that micas were the most influential minerals in their content. The TPB$_M$-Ne-K content, on the other hand, did not differ significantly

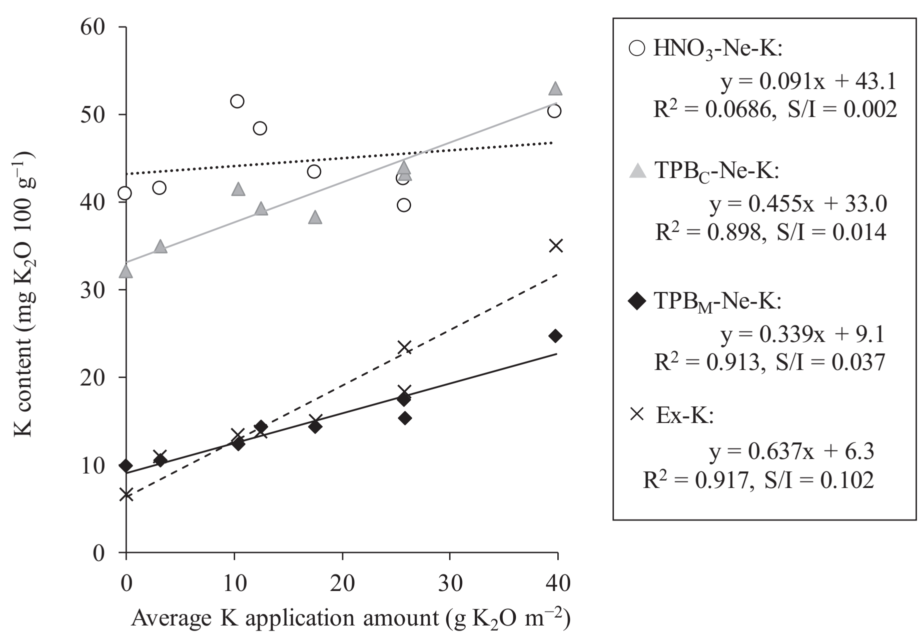

Fig. 2. Relationships of the average K application amount for the last 6 years with the Ex-K and Ne-K contents by three extraction method in an experimental field (KH) The sloping lines indicate linear regression results. S/I: ratio of slope to intercept of regression.

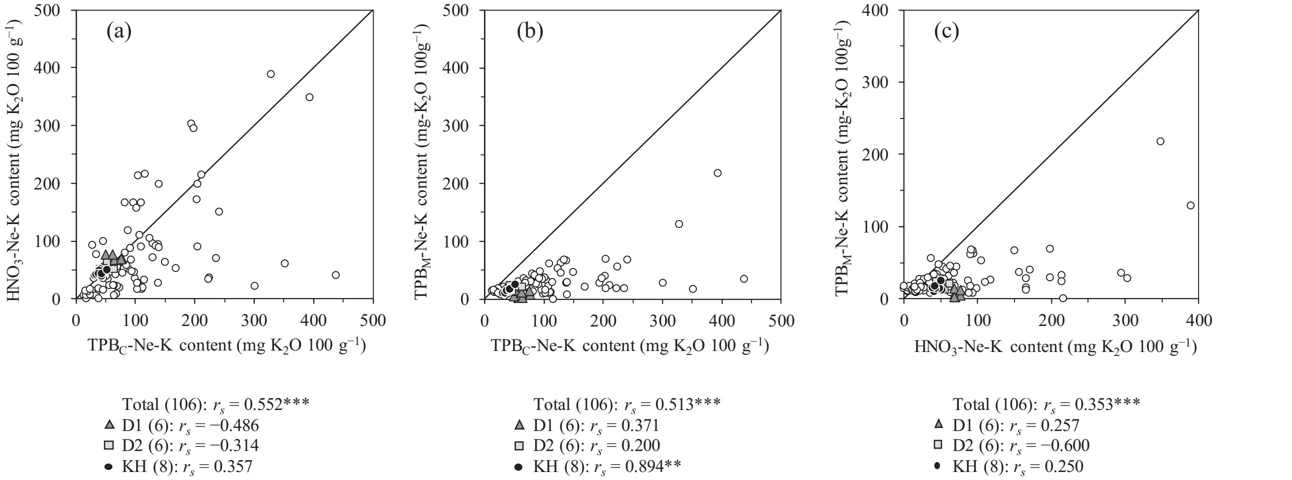

Fig. 3. Relationship between the Ne-K contents by three extraction methods The diagonal lines indicate a 1:1 relationship. r_s indicates Spearman's rank correlation coefficient (***, $p < 0.001$; **, $p < 0.01$).

depending on the mica peak. Unlike the other Ne-Ks, the content was significantly higher along with the peak intensity of smectites, which interlayers can fix K.

These trends observed among the various soils were consistent with the discussion in the KH field that HNO_3-Ne-K and TPB_M-Ne-K were most reflective of native Ne-K and recently fixed K, respectively. The variation in TPB_M-Ne-K was considered to reflect K fixation by or its release from K-fixing minerals, such as vermiculites and smectites, more than the other Ne-Ks. Accordingly, K fertilizer management and soil mineralogy to determine K fixation capacity (Portela et al., 2019) may directly influence Ne-K. The recently fixed K may be held at lower energies in the interlayer of K-fixing minerals than native Ne-K contained in micas (Cox et al., 1996), therefore mild extraction can selectively release it. Compared with the TPB_M method, the TPB_C method can extract more native Ne-K from micas along with a recently fixed K, as observed in the KH field. Consequently, TPB_C-Ne-K was slightly higher in the granitic area, reflecting the geological background of the micas. HNO_3-Ne-K reflected the geological background and amount of micas more than TPB_C-Ne-K. On the other hand, curiously, HNO_3-Ne-K did not increase when K fixation occurred. The fact that the HNO_3 method underestimated the Ne-K increase by K fixation has been reported; this was attributed to the fact that the K concentration in the extract solution increased during the extraction step in the HNO_3 method, unlike the TPB methods which remove the released K as KTPB precipitate (Cox et al., 1996). As the K concentration gradient between the solution and solid phases is the driving force for Ne-K diffusion (Brouder, 2011), an increase in K concentration in the extract solution may retard further release of Ne-K, resulting in a relatively constant amount of Ne-K extracted from individual soil regardless of the degree of K fixation. In conclusion, the Ne-Ks by the three methods can be characterized by the difference in the contribution of two factors: the K fixation degree and the amount of micas.

3.2. Correlation of K parameters with TF of soybean

Ex-K was significantly negatively correlated with TF among all soils, and the r_s value was -0.40 (Fig. 4). A similar negative correlation was also observed among the plots in each field, although r_s values were slightly higher than the significance level in the D1 and D2 fields ($p = 0.07$). However, TF at the same Ex-K level was largely different among fields; for example, when Ex-K was ca. 35 mg K_2O 100 g^{-1}, TF was 0.0011 for the KH field and 0.019–0.023 for the D1 and D2 fields.

The TEr-Ks (sum of Ex-K and Ne-K) by the three methods were significantly negatively correlated with TF among all soils. The r_s values were lower than those in the case of Ex-K (Fig. 5 a–c). In particular, TPB_M-TEr-K had the lowest r_s values among all soils and within each field.

Significant negative correlations with TF were also recognized in the Ne-Ks by the three methods among all soils (Fig. 5 d–f). For the HNO_3 and TPB_C methods, the correlations of the Ne-Ks were slightly weaker than those of Ex-K and TEr-Ks. On the other hand, for the TPB_M method, the correlation of Ne-K was stronger than that of TEr-K. The Ne-K values obtained by the TPB methods were also significantly correlated with the variation in TF within the KH field. For the other cases, there was no significant correlation between Ne-Ks and TF within individual fields.

In pot experiments with paddy rice, the TEr-Ks by the HNO_3 method (Ogasawara et al., 2019) or the TPB_C method (Eguchi et al., 2015) showed stronger correlation with TF than Ex-K. A survey of paddy fields in Fukushima Prefecture reported that HNO_3-Ne-K was negatively correlated with the TF of brown rice, independent of Ex-K (Kurokawa et al., 2020). Similar results were observed in this survey of soybean cultivated fields. However, these TEr-Ks and Ne-Ks showed weaker correlations with TF than the reported values for paddy rice (r_s or r_p, Pearson's correlation coefficient, ≤ -0.60). This study indicates that the TPB_M method is more effective for estimating the RCs transfer risk of soybean than the HNO_3 and TPB_C methods. Additionally, the stronger correlation with TF of TPB_M-Ne-K than Ex-K implies that TPB_M-Ne-K contributes to the control of RCs transfer to soybean more than Ex-K. The larger contribution, however, cannot be explained only from the K supply, since phytoavailability is naturally higher in Ex-K than in Ne-K. Accordingly, we analyzed the relationship between Ne-Ks and RCs fixation as another aspect of the Ne-K effect.

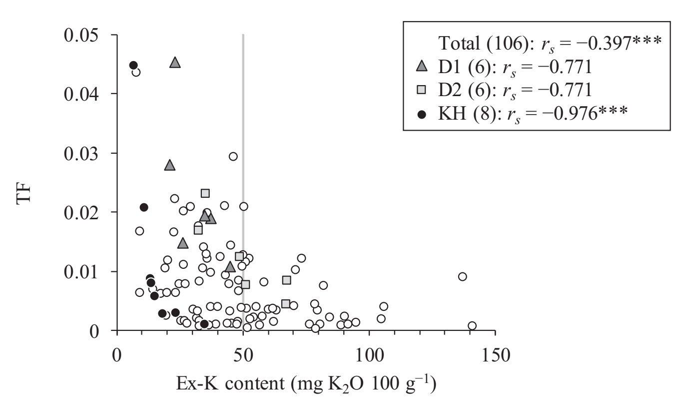

Fig. 4. Relationship between the Ex-K content and TF of soybean r_s indicates Spearman's rank correlation coefficient (***, $p < 0.001$). The gray line shows the recommended value of Ex-K, 50 mg K_2O 100 g^{-1} for areas where soybeans with high RCs concentrations may be produced.

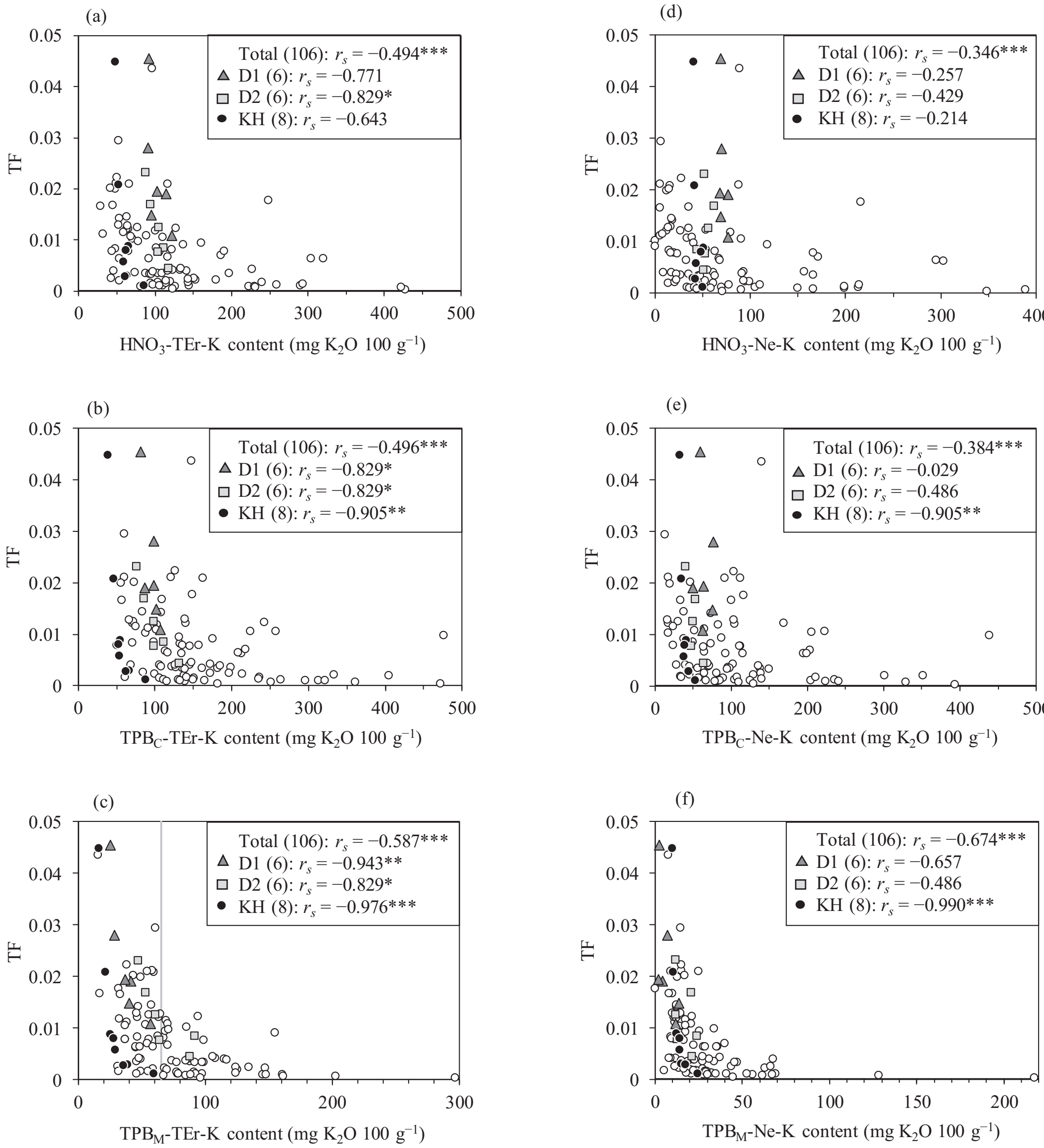

Fig. 5. Relationships of the TEr-K (a, b, c) and Ne-K (d, e, f) contents by three extraction methods with TF of soybean r_s indicates Spearman's rank correlation coefficient (***, $p < 0.001$; **, $p < 0.01$; *, $p < 0.05$). The gray line in (c) shows TPB_M-TEr-K of 65 mg K_2O 100 g^{-1}, as the criterion value suggested in this study for soybean cultivation in areas with high soil RCs concentrations.

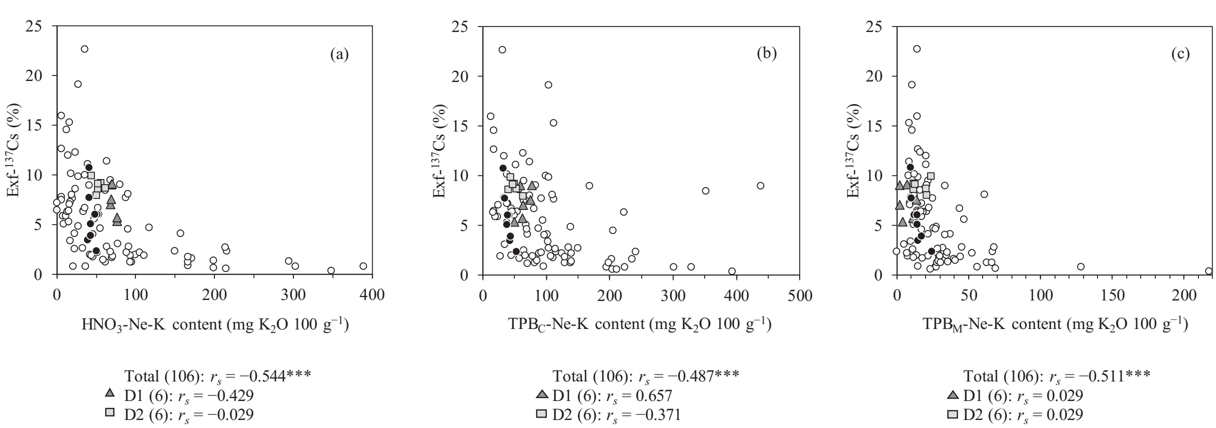

Fig. 6. The relationships between the Ne-K contents by three extraction methods and Exf- ^{137}Cs r_s indicates Spearman's rank correlation coefficient (***, $p < 0.001$; **, $p < 0.01$).

3.3. Correlation of K parameter with RCs fixation

Exf-^{137}Cs was significantly negatively correlated with Ne-Ks by all three methods among all soils (Fig. 6). The r_s values were approximately -0.5, regardless of which method was used. In the KH field, Exf-^{137}Cs largely varied from 2.3 to 10.8%, and the variation was significantly negatively correlated with the variations in the Ne-Ks by the TPB methods but not in HNO_3-Ne-K. The variation in Exf-^{137}Cs was not correlated with Ne-Ks in the D1 field, and Exf-^{137}Cs was almost constant in the D2 field.

The negative correlation between HNO_3-Ne-K and Exf-^{137}Cs, which is in accordance with Kurokawa et al. (2019), may reflect the abundance of weathered micas. The content of micas is highly correlated with HNO_3-Ne-K (Kurokawa et al., 2020), and it can also be regarded as an intrinsic soil factor that controls RCs mobility (Nakao et al., 2015) along with their degree of weathering and types (Hwang et al., 2021; Kitayama et al., 2020; Ogasawara et al., 2017). Weathered micas can selectively adsorb RCs at frayed edge sites (FES[14]; the spaces between the expanded and unexpanded interlayer) to fix it by collapsing the interlayer (Sawhney, 1972). In the geological situation of contamination area by the FDNPP accident, it has been reported that the presence of weathered biotite is a key factor to decrease RCs mobility in soil (Mukai et al., 2014, 2016). The correlation of TPB_C-Ne-K may also partly reflect the content of weathered micas. On the other hand, a similar cause is unlikely for the correlation of TPB_M-Ne-K because it was not significantly dependent on granite (Appendix Table A2) and peak intensity of mica (Table 1).

Exf-^{137}Cs is not determined only by soil mineralogical composition, and it can fluctuate in a field, such as KH. The correlation of Exf-^{137}Cs with the Ne-Ks by the TPB methods within the KH field indicates that ^{137}Cs was fixed or released together with the recently fixed K. Two possible mechanisms were considered for this. First, expanded interlayers of vermiculites and weathered micas are collapsed through K fixation and then trap labile RCs adsorbed there (Absalom et al., 1995). Second, Cs-selective sites can increase when K application collapses these interlayers (Murota et al., 2020). With drying-wetting replications, K application to smectites can also collapse a part of their expanded interlayer and generate Cs-selective sites like FES, where RCs can be temporally fixed (Degryse et al., 2004; Maes et al., 1985). Both processes may be reversible when K is released from the collapsed interlayer. In fact, exchangeable ^{137}Cs concentrations have been reported to increase with decreasing Ex-K content or K concentration in the soil solution during soybean (Matsunami et al., 2021) and rice cultivation (Eguchi et al., 2021). For TPB_M-Ne-K, which is most dependent on K fixation (see Section 3.1), the correlation with Exf-^{137}Cs may reflect RCs fixation by such dynamic mechanisms.

Ex-K was reported to be negatively correlated with ^{137}Cs exchangeability within a field (Kubo et al., 2017), and in this study, significant correlations were also observed between Exf-^{137}Cs and Ex-K in the D1 field ($r_s = -0.94$, $p < 0.01$; Appendix Fig. A3), and KH field ($r_s = -1.00$, $p < 0.001$). However, there was no correlation among all soils ($r_s = 0.05$), indicating that the variation of Ex-K itself is not a direct factor in controlling RCs exchangeability. The factor is probably K fixation/release that occurred in parallel with the Ex-K variation (Fig. 2).

The exchangeable RCs concentration was reported to correlate RCs concentration in soybean grains (Hoshino and Komatsuzaki, 2018). In this study, Exf-^{137}Cs was significantly positively correlated with the TF of soybean among all soils ($r_s = 0.648$, $p < 0.001$; Appendix Fig. A4), and within the KH field ($r_s = 0.976$, $p < 0.001$). Accordingly, the effect of Ne-Ks to repress RCs transfer to soybean was attributed to RCs fixation degree, at least in part.

3.4. Regression estimation of the effects of soil parameters on TF

A number of linear regression models for log-transformed TFs were calculated using one to three log-transformed soil parameters, Ex-K, Exf-^{137}Cs, Ne-K, and TEr-K (Table 2). This analysis has two purposes: 1) to show how much of the $\log_{10}$(TF) variation can be explained by using these single or multiple parameters, and 2) to analyze the inhibitory effect of Ne-K on RCs transfer to soybean through K supply separately from that through RCs fixation.

In all cases where Ne-K or TEr-K was included as an explanatory variable, the adjusted coefficient of determination (adj. R^2[15]) of the TF models was higher when using the values obtained by the TPB_M method than the other two methods. Therefore, the values of the TPB_M method were adopted as parameters of Ne-K and TEr-K in Table 2, and the models using the values of HNO_3 and TPB_C methods are separately summarized in Appendix Table A3.

Among the 1-variable models (I-a–d), the adj. R^2 was higher in the order of models using TPB_M-Ne-K > Exf-^{137}Cs > TPB_M-TEr-K > Ex-K (Table 2), which was similar to the order of the absolute value of r_s between each parameter and TF. TPB_M-Ne-K was singly able explained 44% of the variation in $\log_{10}$(TF) (model I-d). Takeda et al. (2014) performed multiple regression analysis for $\log_{10}$(TF) of soybean cultivated in 2011 using logarithmic values of Ex-K and RCs interception potential and found an adj. R^2 value of 0.42. Similar adj. R^2 values were obtained using only TPB_M-Ne-K in this study, although the cultivation environment was possibly different from 2011, the first year after the FDNPP accident.

In the 2-variable model with a combination of TPB_M-Ne-K and Ex-K (model II-a), the standardized partial regression coefficient (SPRC[16]) was higher for TPB_M-Ne-K than for Ex-K. SPRC is the number of standard deviations that the objective variable would change for every one standard deviation change in an explanatory variable with the influence of all the remaining variables held constant. Therefore, SPRC represents the relative contribution of the explanatory variables. The higher SPRC indicates that TPB_M-Ne-K contributes more than Ex-K to repress RCs transfer to soybean. The larger contribution of Ne-K was partly due to RCs fixation, as discussed in the previous section. When the direct parameter of RCs fixation (Exf-^{137}Cs) was added to the regression model, TPB_M-Ne-K was less valid than Ex-K as the second parameter; the adj. R^2 was higher in the combination model between Exf-^{137}Cs and Ex-K (model II-c) than the models using TPB_M-Ne-K (model II-b).

The combination of Exf-^{137}Cs and Ex-K explained 62% of the variation in $\log_{10}$(TF) (model II-c). After the FDNPP accident, 2-variable models using Ex-K and exchangeable ^{137}Cs concentrations have been reported to be applicable for predicting logarithmic RCs concentrations in the edible parts of paddy rice (Yagasaki et al., 2019) and tomatoes (Endo et al., 2013). Exf-^{137}Cs, when combined with an index of K availability, may be widely applicable for predicting the TF of multiple crops.

To measure K availability, the TPB_M method appears to be more suitable than CH_3COONH_4 extraction. The adj. R^2 of the model combined with Exf-^{137}Cs was increased by replacing Ex-K (model II-c) with TPB_M-TEr-K (model II-d), or adding TPB_M-Ne-K as the 3rd variable (model III). Model III explained 67% of the variation in $\log_{10}$(TF). The fact that the SPRC of TPB_M-Ne-K in the model was significant means that TPB_M-Ne-K has a suppressive effect on TF even when Ex-K and Exf-^{137}Cs are kept constant, indicating that the K supply from Ne-K itself has an effect. The results of multiple regression analyses indicate that TPB_M-Ne-K contributes to the inhibition of RCs transfer to soybean through both K supply and RCs fixation.

In the cases where Ne-Ks of the HNO_3 and TPB_C methods were used in the multiple regression model, SPRCs of the Ne-Ks became insignificant when Exf-^{137}Cs was given as an explanatory variable (models H-II-b, H-III, C-II-b, C-III in Appendix Table A3). This indicates that the correlation of these Ne-Ks with TF (Fig. 5 d, e) was almost entirely due to RCs fixation. For paddy rice, the 2-variable model combining HNO_3-Ne-K and Ex-K was reported to explain 61% of the variation in $\log_{10}$(TF) (Kurokawa et al., 2020). However, only 36% of the variation was explained by this combination for soybean (model H-II-a in Appendix Table A3). HNO_3-Ne-K is not applicable for predicting TF in soybean, unlike paddy rice. The optimal

[14] FES: frayed-edge sites

[15] adj. R^2: adjusted determination coefficient

[16] SPRC: standardized partial regression coefficient

Table 2
Results of regression analysis.

Regression model		adj. R^2	Standardized partial regression coefficient			
No.	Formula		$\log_{10}$(Exf-^{137}Cs)	$\log_{10}$(Ex-K)	$\log_{10}$(Ne-K +1)	$\log_{10}$(TEr-K)
I-a	$\log_{10}(TF) = -0.80 \log_{10}(Ex\text{-}K) - 1.01$	0.178				
I-b	$\log_{10}(TF) = -1.31 \log_{10}(TEr\text{-}K) - 0.07$	0.399				
I-c	$\log_{10}(TF) = 0.83 \log_{10}(Exf\text{-}^{137}Cs) - 2.79$	0.421				
I-d	$\log_{10}(TF) = -1.01 \log_{10}(Ne\text{-}K + 1) - 0.96$	0.439				
II-a	$\log_{10}(TF) = -0.42 \log_{10}(Ex\text{-}K) - 0.89 \log_{10}(Ne\text{-}K + 1) - 0.45$	0.480		−0.227	−0.588	
II-b	$\log_{10}(TF) = 0.54 \log_{10}(Exf\text{-}^{137}Cs) - 0.69 \log_{10}(Ne\text{-}K + 1) - 1.71$	0.577	0.429		−0.457	
II-c	$\log_{10}(TF) = 0.84 \log_{10}(Exf\text{-}^{137}Cs) - 0.82 \log_{10}(Ex\text{-}K) - 1.49$	0.616	0.661	−0.444		
II-d	$\log_{10}(TF) = 0.66 \log_{10}(Exf\text{-}^{137}Cs) - 1.02 \log_{10}(TEr\text{-}K) - 0.85$	0.650	0.521			−0.498
III	$\log_{10}(TF) = 0.65 \log_{10}(Exf\text{-}^{137}Cs) - 0.62 \log_{10}(Ex\text{-}K) - 0.45 \log_{10}(Ne\text{-}K + 1) - 1.10$	0.670	0.513	−0.338	−0.299	

TEr-K and Ne-K used as explanatory variables are both values obtained by the TPB_M method. All listed standardized partial regression coefficients were statistically significant ($p < 0.01$).

method to quantify Ne-K that controls the transfer of RCs to plants may be different between crop types and/or between paddy and upland fields.

3.5. Estimation method of RCs transfer risk in soybean cultivation in Fukushima

For cultivation of soybean in areas contaminated by the FDNPP accident, it is recommended to maintain Ex-K above 25 mg K_2O 100 g^{-1} (Ministry of Agriculture, Forestry and Fisheries, 2015). In a survey of paddy rice cultivation, TF was considerably low (< 0.005) in fields with Ex-K content of 25 mg K_2O 100 g^{-1} or higher (Kurokawa et al., 2020). The TF of soybean, however, was not always low even when the Ex-K content was above 25 mg K_2O 100 g^{-1}, and some fields with higher Ex-K content showed relatively high TF in this study (0.02–0.03; Fig. 4).

The MAFF and Fukushima Prefecture government recommended that Ex-K should be 50 mg K_2O 100 g^{-1} or higher in areas where soybeans with high RCs concentrations had been harvested in the past or where soil RCs concentrations are high (Ministry of Agriculture, Forestry and Fisheries, 2015), and where soybeans are cultivated for the first time after the FDNPP accident (Agriculture, Forestry and Fisheries Department of Fukushima Prefecture, 2019). Among the fields with an Ex-K content of 50 mg K_2O 100 g^{-1} or more, the maximum TF was 0.020 (Fig. 4), indicating that the RCs concentration in soybean grains is unlikely to exceed the standard limit of food (100 Bq kg^{-1}, Ministry of Health, Labor and Welfare, 2012) when the soil RCs concentration is below 5000 Bq kg^{-1}. According to the guidelines for farmland decontamination, in principle, soil RCs concentrations are reduced by scraping surface soil away for fields where soil has never been plowed since the FDNPP accident and the average soil RCs concentration up to 15 cm depth exceeds 5000 Bq kg^{-1} (Ministry of Agriculture, Forestry and Fisheries, 2013). Therefore, if Ex-K can be increased to 50 mg K_2O 100 g^{-1}, the RCs concentration in soybean grain will be lower than 100 Bq kg^{-1} in most fields. However, the enhancement is often unnecessary even if the soil RCs concentration is high, and in fact, TF was considerably low (< 0.005) in 22 fields with Ex-K content below 50 mg K_2O 100 g^{-1}.

If TPB_M-TEr-K is used instead of Ex-K as a diagnostic criterion for soil K status, it should be possible to estimate the RCs transfer risk more effectively. In the soils with TPB_M-TEr-K above 65 mg K_2O 100 g^{-1}, TF was less than 0.020 (maximum 0.012) (Fig. 5 c). Soils with TPB_M-TEr-K higher than 65 mg K_2O 100 g^{-1} (48 soils) were more common than soils with Ex-K higher than 50 mg K_2O 100 g^{-1} (38 soils), indicating that it is easier to achieve. Among soils showing considerably low TF (< 0.005), three-quarters (36 soils) had TPB_M-TEr-K values higher than the threshold. The criterion TPB_M-TEr-K ≥ 65 mg K_2O 100 g^{-1} distinguished more effectively between fields with low and higher RCs transfer risks compared to the Ex-K criterion.

TPB_M-Ne-K was more strongly correlated with TF than TPB_M-TEr-K among the different fields (Fig. 5 f). With a TPB_M-Ne-K content higher than 25 mg K_2O 100 g^{-1}, TF was lower than 0.02, regardless of the Ex-K content; for example, in a soil with only 9 mg K_2O 100 g^{-1} of Ex-K and 36 mg K_2O 100 g^{-1} of TPB_M-Ne-K, TF was 0.006 (F43 in Appendix Table A2). Compared with TPB_M-Ne-K, however, TPB_M-TEr-K is more convenient as a diagnostic criterion for managing RCs transfer risk for two reasons. First, TPB_M-TEr-K can be quantified by a single extraction of the TPB_M method, while the calculation of TPB_M-Ne-K requires further extraction with CH_3COONH_4. Second, the TPB_M-TEr-K content can be increased by K fertilizer application in any soil and can be applied as a fertilizer improvement criterion, but TPB_M-Ne-K content cannot be increased in soils that do not contain K-fixing minerals. Applying K fertilizer aimed at increasing TPB_M-TEr-K to 65 mg K_2O 100 g^{-1} can effectively control RCs concentration in soybeans grown in areas with high soil RCs concentrations, regardless of soil mineralogy. In soils without K-fixing minerals and soils rich in K-fixing minerals, increases in Ex-K and Ne-K, respectively, can play a major role in inhibiting RCs transfer to soybean, and TPB_M-TEr-K content reflects both increases.

The inhibitory effect of K fertilizer application on RCs transfer may be greater in soils rich in K-fixing minerals than in soils without K-fixing minerals. This is because K fixed by the minerals not only acts as a source of K for plants but also causes RCs fixation. In fact, it has been reported that when K fertilizer application was increased in a soil rich in vermiculite, the RCs transfer to buckwheat was greatly reduced, even though Ex-K was only slightly increased (Kubo et al., 2018). Previously, K fixation by vermiculites has been regarded as a troublesome matter preventing the increase of Ex-K since the FDNPP accident (Nihei and Hamamoto, 2019). In this study, we showed that fixed K inhibits the transfer of RCs to soybean. Although fixed K could not be quantified with the conventional diagnosis of soil K status by CH_3COONH_4 extraction, the TPB_M method can extract fixed K with Ex-K. It should be noted that our discussion of the factors of TPB_M-Ne-K and the mechanism reducing RCs transfer to soybean are inferences based on the field observations and have not been experimentally proved. The extent to which K fixed by vermiculites and smectites reflects on the values of TPB_M-Ne-K should be examined by chemical experiments using pure mineral samples. The direct relationship between these minerals and the TF of soybean should also be examined in the future by pot cultivation experiments. These validation works could provide important information for establishing a diagnostic protocol for producing soybean in contaminated areas.

4. Conclusions

After comparing the three extraction methods, we found that the Ne-K extracted by the TPB_M method was the most correlated with RCs transfer from soil to soybean. TPB_M-Ne-K was significantly affected by the amount of K fertilizer applied and K-fixing minerals (vermiculites and smectites), but not by mica amount, indicating that the variation reflects K fixation. We observed that the increase in TPB_M-Ne-K due to K fixation caused RCs fixation. Statistical analyses showed that TPB_M-Ne-K inhibited RCs transfer to soybean better than Ex-K through both RCs fixation and K supply. TPB_M-TEr-K included recently fixed K and Ex-K, both of which play a role in reducing RCs contamination in soybean, and it showed stronger correlations with the TF of soybean than Ex-K in every case. The criterion of TPB_M-TEr-K as 65 mg K_2O 100 g^{-1} was able to differentiate the soils with considerably low TF values (< 0.005) for soybeans from the soils with relatively high TF values (> 0.020) more effectively than Ex-K. In conclusion, the application of TPB_M-TEr-K instead of Ex-K as a fertilizer improvement criterion reduces risk of RCs transfer to soybean regardless of soil mineral composition. This application will help to reduce the amount of K fertilizer consumed for controlling soybean contamination and maintain food safety at a lower cost.

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CRediT authorship contribution statement

Shokichi Wakabayashi: Conceptualization, Data curation, Formal analysis, Methodology, Investigation, Writing – original draft, Visualization, Project administration. **Tetsuya Eguchi:** Conceptualization, Funding acquisition, Methodology. **Atsushi Nakao:** Conceptualization, Funding acquisition, Methodology, Project administration, Writing – review & editing. **Kazuki Azuma:** Investigation. **Shigeto Fujimura:** Project administration, Writing – review & editing. **Katashi Kubo:** Resources. **Masaaki Saito:** Resources, Writing – review & editing. **Hisaya Matsunami:** Funding acquisition, Investigation, Project administration. **Junta Yanai:** Conceptualization, Funding acquisition, Methodology, Project administration.

Declaration of competing interest

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

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Appendix A. Supplementary data

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