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Application of Finnish phlogopite to reduce radiocesium uptake by paddy rice

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ABSTRACT

Field and pot experiments were conducted to evaluate the effectiveness of coarse Finnish phlogopite application to reduce radiocesium uptake by paddy rice (*Oryza sativa* L.). The application of phlogopite was expected to reduce radiocesium uptake by crops through K supply and radiocesium retention. Three fields were set in Fukushima Prefecture, and coarse (mean particle size of 450 μm) phlogopite from Siilinjärvi (Finland) was applied at a rate of 5 t ha^{-1}. Paddy rice was cultivated for 2–4 successive years. In all fields, the average ^{137}Cs transfer factor (TF) of brown rice harvested from plots with added phlogopite was significantly lower than that of brown rice from plots without added phlogopite over the 2–4-year experiments. TF was decreased by up to 80% following phlogopite application, without an adverse effect on yield. Exchangeable K and soil solution K were higher in the soils with added phlogopite, suggesting K released from phlogopite reduced ^{137}Cs uptake by paddy rice. Moreover, in a pot cultivation experiment, even when 55% of the total K was removed from phlogopite prior to application, the TF in pots with phlogopite application was less than half of that in pots without added phlogopite. The results from the field study and the pot cultivation experiment suggested that the application of Finnish phlogopite is effective to reduce the TF of brown rice. Exchangeable K and tetraphenylborate-extractable-K (TPB-K) at rooting stage, and soil solution K at tillering and heading stages showed significant negative correlation with TF. TPB-K was significantly positively correlated with soil solution K at tillering stage and heading stage, whereas exchangeable K at rooting stage did not exhibit significant correlation with soil solution K at heading stage. The results suggest that TPB-K is more reliable than exchangeable K, which could facilitate as a basis of K fertilizer recommendation for radiocesium-contaminated fields.

1. Introduction

Following the Tokyo Electric Power Company's Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident in Fukushima Prefecture, Japan, in March 11, 2011, it is critical to establish measures to reduce the uptake of radiocesium by crops. In paddy fields, such measures are of the highest priority, because paddy rice is the staple food in Japan (Ministry of Agriculture, Fishery and Forestry, 2015). Potassium inhibits the uptake of radiocesium by crops via competition for root absorption (Zhu and Smolders, 2000). Moreover, maintaining high levels of external K in soil inhibits the expression of K transporters which have high affinity for radiocesium (Fujimura et al., 2014). In addition to these direct effects, K application may reduce radiocesium uptake by plants by inhibiting the release of radiocesium once retained to high-affinity sites, and/or once fixed in interlayers of 2:1 clay minerals (Eguchi et al., 2015; Ogasawara et al., 2019). Based on the results of urgent experiments performed in 2011 (Kato et al., 2015), Fukushima Prefectural government recommended that when soil exchangeable (1 M ammonium acetate extractable) K is lower than 250 mg kg^{-1}, K fertilizer should be applied to secure soil exchangeable K over 250 mg kg^{-1} throughout the growing season of paddy rice (Fukushima Prefectural Government, 2014). In addition to KCl, K-bearing minerals, such as zeolite, vermiculite, and

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Table 1
Summary of field and pot experiments.

Field	City	Year	Variety	Plot area	Planting	Soil sampling date at each growing stage				Application rate (kg ha^{-1})		
				(m^2)	date	Rooting	Tillering	Heading	Harvest	N	P_2O_5	K_2O
A	Date	2014	Hitomebore	16	June 6	June 10	July 16	August 18	October 3	60	60	60
		2015	Koshihikari	16	May 14	May 15	July 3	August 7	September 23	63	82	0
		2016	Fukuhibiki	16	June 3	June 3	July 18	August 10	September 29	63	60	0
		2017	Fukuhibiki	16	June 2	June 9	July 7	August 7	October 6	63	60	0
B	Minami-Souma	2014	Koshihikari	24[a], 16[b]	May 21	May 26	July 7	August 11	September 26	26	27	12
		2015	Fukuhibiki	24[a], 16[b]	May 21	May 26	July 7	August 11	September 26	58	64	171
C	Iwaki	2015	Koganemochi	16	May 15	May 21	July 10	July 30	September 14	32	80	72[b], 192[c]
		2016	Ten'notsubu	16	May 24	May 31	June 30	August 1	September 26	32	80	72[b], 192[c]
Pot	Date	2016	Koshihikari	0.02	June 26				October 20	40	80	80

[a] Control plot.
[b] Phlogopite applied plot.
[c] Additional-K applied plot.

mica have been examined as a source of plant-available K (Saito et al., 2012; Fujimura et al., 2013, 2016; Kato et al., 2015; Eguchi et al., 2015). Shortly after the FDNPP accident, zeolite and vermiculite were expected to reduce the uptake of radiocesium by crops because of their high Cs selectivity. However, the effectiveness of this method would mainly be due to the supply of K from the applied minerals, rather than the fixation of radiocesium (Ministry of Agriculture, Fishery and Forestry, 2015). Eguchi et al. (2015) reported that, in addition to exchangeable K, nonexchangeable K (which was evaluated as tetraphenylboron (TPB)-extractable K of trioctahedral mica, such as biotite (most common trioctahedral mica) and phlogopite (magnesium-rich biotite)), also contributed to reduce radiocesium uptake by paddy rice. Furthermore, a pot experiment demonstrated that TPB-extractable K before planting was significantly negatively associated with TF, whereas exchangeable K before planting was not. TPB can extract nonexchangeable K from the mica-interlayer; therefore, plant-available K by TPB-extraction has been evaluated for many crops (Jackson, 1985; Cox and Joern, 1999; Fernández et al., 2008; Li et al., 2016, 2018). However, most studies on soil-K tests by TPB involved pot cultivation experiments. Due to the limited soil volume in pot cultivation, K uptake by plants would decrease the level of K in soil solution more rapidly than in a field study (Ishikawa et al., 2017). Lower levels of K in soil solution accelerate the release of nonexchangeable K (Fanning et al., 1989). Therefore, pot cultivation might overestimate the influence of nonexchangeable K and, consequently, the usefulness of TPB-extraction as a soil test. However, the usefulness of TPB-extraction as a soil-K test to evaluate the transfer risk of radiocesium has not been investigated in a paddy rice field.

Phlogopite contains approximately 10% (by weight) of K_2O (Fanning et al., 1989), which is approximately five-fold higher than that of commercially available Japanese zeolite (Eguchi et al., 2015). Phlogopite K-supply is expected to be more durable than that from zeolite. Paasikallio (1999) noted that application of biotite from Siilinjärvi, which is used as a K fertilizer in Finland, reduced the radiocesium uptake by Italian ryegrass from peat soil at least for 5 years, whereas zeolite reduced the radiocesium uptake for only 2 years. Biotite from Siilinjärvi is dominated by phlogopite (Al Ani, 2013). Eguchi et al. (2015) reported that application of fine-ground phlogopite (10–20 μm; according to the distributer, Siilinjärvi) to a pot experiment strongly inhibited the uptake of radiocesium by paddy rice. However, due to the high cost, the application of fine-ground phlogopite to fields is not feasible. Conversely, the application of coarse phlogopite is more feasible, because of the lower cost. However, coarse phlogopite releases K from its nonexchangeable fraction slower than fine phlogopite, because the rate of release decreases with increasing particle size (Fanning et al., 1989). Moreover, phlogopite K-release rates decrease as its K contents decrease because K-release is a diffusion-controlled process (Scott and Smith, 1966). Therefore, it is unclear whether coarse phlogopite can supply soil with K at a sufficient rate to reduce radiocesium uptake by paddy rice, particularly when the K of phlogopite is depleted by root uptake.

Table 2
Particle size distribution and clay mineralogy of soils.

Field	Particle size distribution (%)			Clay mineralogy[a]
	Clay	Silt	Sand	
A	31	21	48	Kt > Sm, Mi, Vr, HIS/HIV
B	10	6	84	Kt > HIV > Mi-Vr, Mi, Vr, Sm
C	17	6	77	Mi-Vr, HIV/HIS, Kt > Sm, Vr > Mi
Pot	16	10	74	HIV/HIS > Kt > Mi-Vr, Vr, Mi

Clay < 0.002 mm, Silt 0.002–0.02 mm, Sand 0.02–2 mm.
Kt, kaoline; Sm, smectite; Mi; mica; Vr, vermiculite; HIS, Hydroxy-interlayered smectite;
HIV, Hydroxy-interlayered vermuculite; Mi-Vr, mica-vermiculite interstratified minerals.
[a] Determined by X-ray diffraction (Fig. S1).

In this study, we applied coarse Siilinjärvi phlogopite to three paddy fields contaminated by radiocesium, and cultivated paddy rice to evaluate the usefulness of Siilinjärvi phlogopite application to reduce the uptake of radiocesium. We also conducted a pot cultivation experiment to evaluate the effectiveness of phlogopite when its K content was depleted following adsorption by paddy rice. Additionally, the reliability of TPB-extractable K as the basis for fertilizer recommendations was also investigated under field conditions.

2. Materials and methods

2.1. Field experiments

Three fields contaminated with radiocesium in Fukushima Prefecture were selected for this study (Table 1). Fields A, B, and C were set in Date City, Minami-Souma City, and Iwaki City, respectively. The distribution of particle size and the clay mineralogy of soils are shown in Table 2. X-ray diffraction patterns revealed that all the soils contained 2:1 type clay minerals (Fig. S1 in supplementary materials).

Plants were cultured in field A from 2014 to 2017, in field B from 2014 to 2015, and in field C from 2015 to 2016. In fields A and B, phlogopite plots (application of phlogopite at a rate of 5 t ha^{-1}) and control plots were set in duplicate. The details of the cultivation and the results of control plots have been reported elsewhere (as field C in Date City and field D as Minami-Souma City; Fujimura et al., 2016). Consistent with control plots, phlogopite plots were not subjected to decontamination.

The coarse Siilinjärvi phlogopite (SC-30; mean particle size of 450 μm) containing 9.6% of K_2O was purchased from Repco Inc., Tokyo, Japan. Phlogopite was applied at 5 t ha^{-1} with basal fertilizer, also known as pre-planting fertilization, before tillage in the first year of the experiment. In field C, a phlogopite plot (application rate of 5 t ha^{-1})

and an additional K plot (application rate of 120 $kgK_2O\ ha^{-1}$ by KCl (192 $kgK_2O\ ha^{-1}$) including basal fertilizer) without phlogopite were set in triplicate. Due to the narrowness of field C, no control (without phlogopite nor additional K) plot was set. In 2016, an additional K plot was set in duplicate, because one plot was omitted due to an accidental K supply from rice straw stacked on the plot during the previous winter in 2015. Basal fertilizer was applied as mixed fertilizer with slow release N to fields A and C. For field B, out-of-spec soybeans and rice bran, corresponding to 12 kg $K_2O\ ha^{-1}$, were applied as basal fertilizer in 2014. In 2015, in addition to out-of-spec soybeans and rice bran, corresponding to 11 kg $K_2O\ ha^{-1}$, oil palm-derived K fertilizer was applied for conventional commercial rice production in field B. Top dressing was not applied, except for nitrogen-fertilizer in field B. Due to the high exchangeable K level in field A, K-fertilizer was only applied in the first year. After the harvest, rice straw was left in the fields and plunged before the next cultivation.

Soils were sampled at the rooting, tillering, and heading stages, and at harvest. Soil solutions were also sampled when soil were sampled at rooting, tillering, and heading stages, using a sampler with a porous part (2.5 mm diameter, 10 cm length) connected to a 10 cm polypropylene tube (DIK-301 B, Daiki Rika Kogyo, Tokyo, Japan), except for field A in 2016 and 2017.

2.2. Pot cultivation

K-depleted phlogopite was prepared by contact with plant roots in K-free culture solution (Hinsinger and Jaillard, 1993). Paddy rice seed (*Oryza sativa* L. 'Koshihikari'; 120 g) was immersed in pure water for 5 days at 20 °C for germination. Phlogopite (SC-30; 15 g) was thinly spread on a plastic tray (445 × 323 × 70 mm) and germinated seeds were sown. Trays were placed in a greenhouse without air-conditioning in early summer (11 May 2016), and irrigated by K-free half-strength Kimura B solution (Nobori et al., 2014). The solution was placed at a depth of a few millimeters and then increased to 60–70 mm as the rice grew. The solution was renewed at 28 days after the start of irrigation. The K-depleted phlogopite was recovered at 4, 8, 14, 23, and 38 days after the start of irrigation by disentangling roots in pure water. The recovered phlogopite was rinsed in pure water and dried at 40 °C.

Pot cultivation was conducted in three replicates. Soil was collected from a paddy field, not field A, in Date City. Exchangeable K in the soil was 25 mg $K_2O\ kg^{-1}$. The clay mineralogy was dominated by hydroxy interlayered vermiculite (Table 2). Soils (3 kg) were mixed with 0.31 g of NH_4Cl, 0.39 g of $CaHPO_4{\bullet}2H_2O$, 1.35 g of KCl, and 10 g of original phlogopite or K-depleted phlogopite. The soil was then placed in a 0.02-m^2 Wagner pot. The rate of N, P, and K application corresponded to the recommendations for application in Fukushima Prefecture (Fukushima Prefectural Government, 2006), and the rate of phlogopite application was same as used for the field experiments (5 t ha^{-1}). Soils without phlogopite were also prepared as a control. Three 21-day seedling (*O. sativa* L. 'Koshihikari') were transplanted on 26 June 2016. Pots were flooded throughout the growing season. Rice was harvested on 20 October 2016.

2.3. Analytical methods

Soil samples were air-dried and then passed through a 2-mm sieve. Exchangeable K was extracted by shaking for 1 h using 1 M ammonium acetate at soil:solution ratio of 1:20. TPB-extractable K (TPB-K) was also determined as an indicator of plant-available K. TPB-K was extracted using the method of Carey et al. (2010). One-gram of air-dried soil was extracted by 3-mL mixed solution of 0.1 M sodium-TPB/1.7 M sodium chloride/0.01 M disodium ethylenediaminetetraacetic acid (EDTA) for 4 h at 20 °C in a 50-mL polypropylene tube. The reaction was stopped by adding 10 mL of 0.5 M ammonium chloride solution and 1.5 mL of 0.5 M cupper chloride solution. Samples were gently boiled for 30 min to destroy K-TPB precipitates, allowed to cool to 20–25 °C, made up to 50 mL with the addition of 1 mL of 3 M hydrochloric acid, and left overnight to settle. The supernatant was filtered using a 0.45-μm membrane filter prior to K analysis.

The K concentrations of soil extracts and soil solutions were determined by atomic adsorption spectrometry (ZA-3000, Hitachi High-Technologies Corporation, Tokyo, Japan). The contents of exchangeable and TPB-K were expressed in oxide form (K_2O). Exchangeable ^{137}Cs was extracted from soils sampled at the rooting stage and harvest, in the same manner as for exchangeable K, but with a soil: solution ratio of 1:10 (Kondo et al., 2014).

Table 3
Average yield and ^{137}Cs transfer of brown rice in field experiments.

Field	Year	Treatment	n	Yield of brown rice[a]	^{137}Cs concentration		^{137}Cs transfer factor
				(g m^{-2})	Brown rice[a] (Bq kg^{-1})	Soil (kBq kg^{-1})	($\times 10^3$)
A	2014	Control	2	518 ± 3	1.9 ± 0.5	4.14 ± 0.22	0.46 ± 0.13
		Phlogopite	2	542 ± 11	1.4 ± 0.0	3.90 ± 0.46	0.37 ± 0.05
	2015	Control	2	627 ± 12	7.3 ± 0.2	3.70 ± 0.19	2.0 ± 0.2
		Phlogopite	2	634 ± 11	1.5 ± 0.0	3.82 ± 0.44	0.38 ± 0.01
	2016	Control	2	749 ± 12	10.8 ± 0.9	4.19 ± 0.30	2.6 ± 0.0
		Phlogopite	2	759 ± 7	5.1 ± 0.0	4.17 ± 0.32	1.2 ± 0.1
	2017	Control	2	687 ± 8	22.3 ± 5.2	3.85 ± 0.29	5.7 ± 0.9
		Phlogopite	2	691 ± 0	12.4 ± 1.9	4.11 ± 0.15	3.0 ± 0.3
	ANOVA	Year		***	***	ns	***
		Treatment		ns	**	ns	***
		Year × Treatment		ns	ns	ns	**
B	2014	Control	2	481 ± 51	10.0 ± 2.4	1.23 ± 0.13	8.0 ± 1.1
		Phlogopite	2	497 ± 38	3.1 ± 0.3	1.11 ± 0.06	2.8 ± 0.1
	2015	Control	2	498 ± 20	1.4 ± 0.2	1.06 ± 0.02	1.3 ± 0.2
		Phlogopite	2	452 ± 17	1.1 ± 0.1	1.14 ± 0.03	1.0 ± 0.0
	ANOVA	Year		ns	*	ns	**
		Treatment		ns	*	ns	**
		Year × Treatment		ns	ns	ns	*
C	2015	Additional K	3	404 ± 83	4.2 ± 1.8	0.193 ± 0.012	22 ± 10
		Phlogopite	3	420 ± 30	2.9 ± 0.6	0.187 ± 0.018	16 ± 2
	2016	Additional K	2	423 ± 30	4.4 ± 1.2	0.223 ± 0.011	20 ± 5
		Phlogopite	3	453 ± 35	2.1 ± 0.1	0.240 ± 0.039	9.0 ± 0.9
	ANOVA	Year		ns	ns	**	ns
		Treatment		ns	ns	ns	ns
		Year × Treatment		ns	ns	ns	ns

^{137}Cs, cesium-137; K, potassium; numbers after ± denote the standard deviation (n = 3) or difference between the average value and each duplicate value.
*, $p < 0.05$; **, $p < 0.01$; ***, $p < 0.001$; ns, not significant; in two-way analysis of variance (ANOVA).
[a] 15% moisture base, > 1.8 mm.

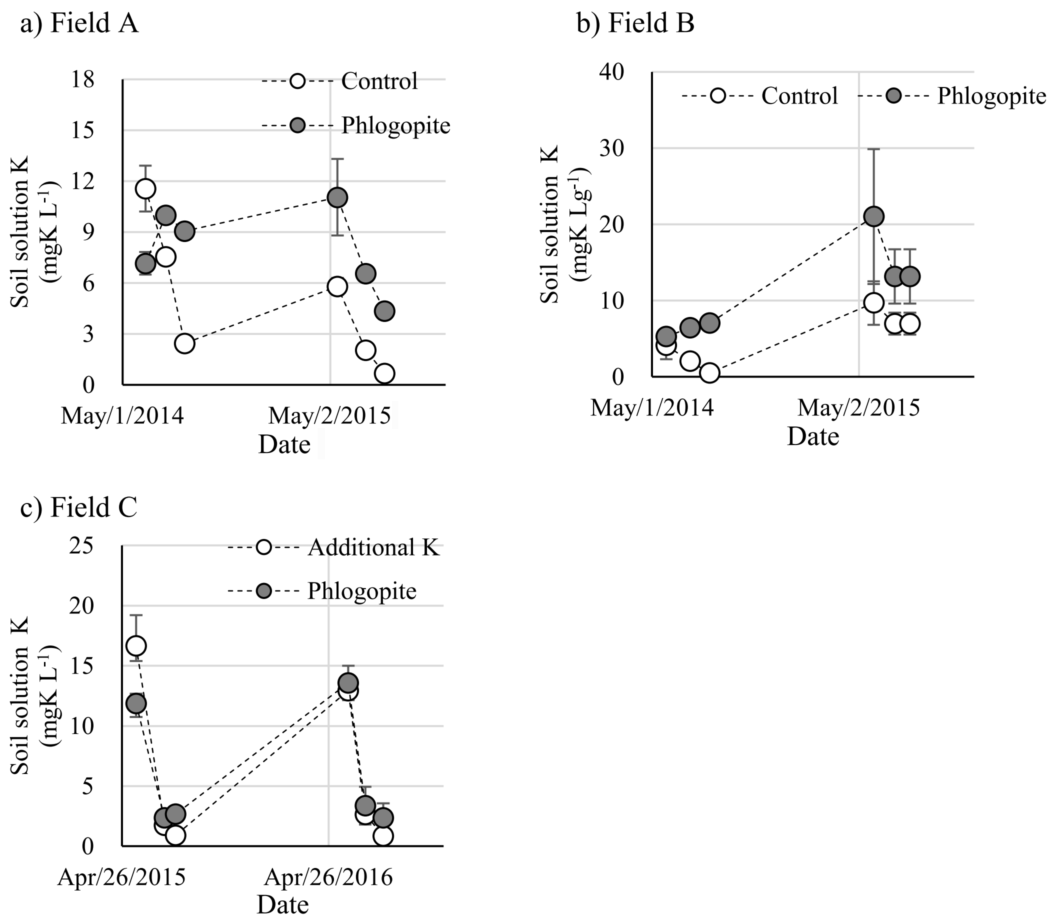

Fig. 1. Changes in soil solution K in field experiments in a) field A, b) field B, and c) field C. Open symbols indicate soils without phlogopite-application and closed symbols indicate phlogopite-applied soils. Error bars indicate the highest and lowest values.

The ^{137}Cs concentration of soil, soil extracts, and harvested brown rice was determined by gamma spectrometry (GC2520 or GC4520, Canberra, Meriden, CT, USA). Counting time was set to ensure that the counting error of each samples was below 5% for soils and soil extracts in 100 mL polypropylene cylindrical containers, and below 10% for brown rice in 2-L Marinelli beakers. The transfer factor (TF) of ^{137}Cs from soil to brown rice was determined as follows: TF = ^{137}Cs concentration of 15% moisture base brown rice (Bq kg^{-1})/^{137} Cs concentration of oven-dried soil (Bq kg^{-1}).

2.4. Statistical analysis

All statistical analyses were performed using SAS University Edition (SAS Studio 3.8; SAS Institute Inc., Cary, NC, USA). Two-way analysis of variance (ANOVA) was conducted to evaluate the effect of phlogopite application and year on field experiments. Multiple comparisons for the pot experiment results were performed using the Tukey-Kramer method. Spearman rank correlation coefficient (ρ) was calculated to analyze the relationship between soil K status and TF. Pearson correlation coefficient (r) was calculated to analyze the relationship between several soil K fractions, and between the concentration of soil exchangeable ^{137}Cs and brown rice ^{137}Cs.

3. Results

3.1. Yield and ^{137}Cs transfer of brown rice in field experiments

The yield and TF of brown rice are summarized in Table 3. Application of phlogopite did not adversely affect the yield of paddy rice. The TF of brown rice harvested in phlogopite plots was significantly lower than that of rice harvested in plots without added phlogopite in fields A and B. TF decreased up to 80% following phlogopite application. In field A, TF increased throughout the 4-year experiment in the control plot, whereas there was no change in the first 2 years, followed by an increase in the phlogopite-added plots. In field B, the TF in the second year, when basal-K-fertilizer was applied to resume conventional cultivation, was much lower than that in the first year. A difference in TF in the first and second year was unclear in field C.

3.2. Changes in soil solution K, exchangeable K, TPB-K, and exchangeable ^{137}Cs in field experiments

Changes in soil solution K, exchangeable K, and TPB-K trends are shown in Figs. 1–3. The levels of these K forms were higher in soils with added phlogopite than in soils without it, except for some observations during the first year in field C. In all fields, soil solution K, exchangeable K, and TPB- in soils without added phlogopite decreased until the heading stage. In some cases (e.g., field A in 2015), exchangeable K and TPB-K were slightly increased from the heading stage to harvest.

In soil with added phlogopite in fields A and B, there was a slight increase in soil solution K during the cultivation period in the first year, while there was no considerable change in exchangeable K and TPB-K. Conversely, in field C, the levels of soil solution K in soil with added phlogopite and soil with additional-K decreased. Exchangeable K in soil with added phlogopite also decreased until the heading stage, as observed for soil with additional K. However, there was a marked increase afterward, from the heading stage to harvest, resulting in almost similar exchangeable K in soil with added phlogopite at rooting stage

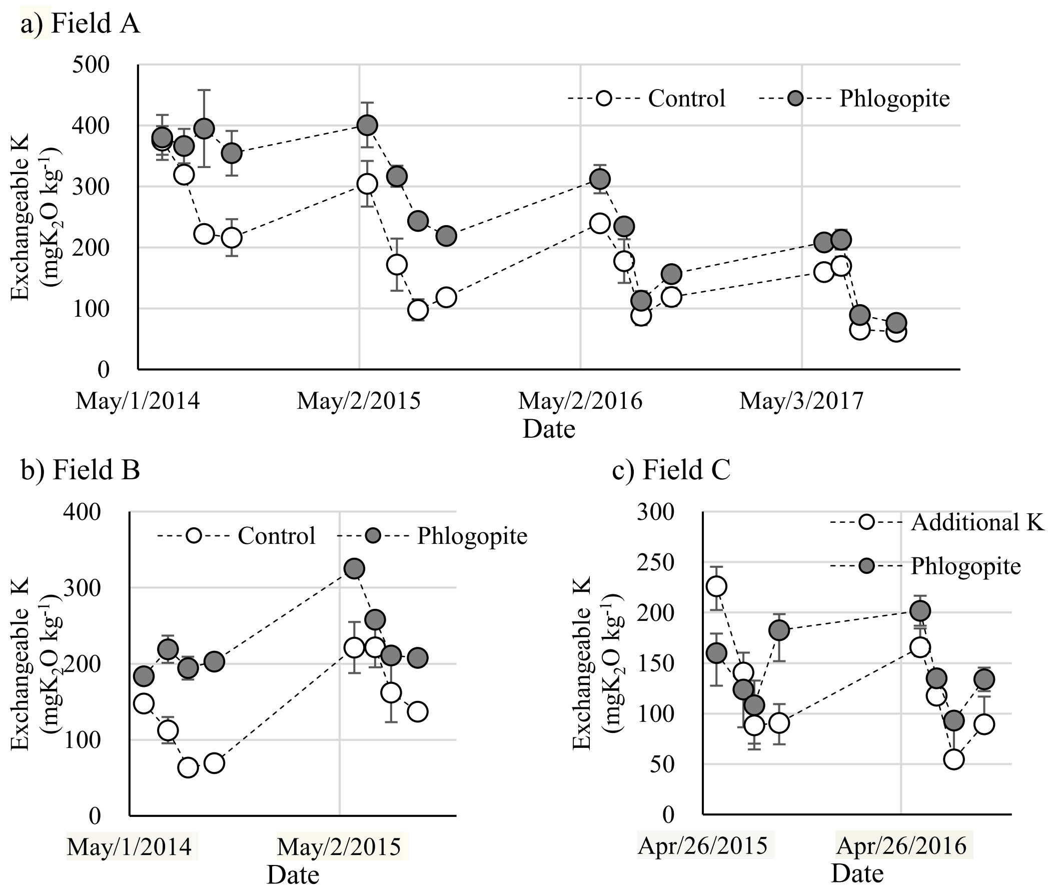

Fig. 2. Changes in soil exchangeable K in field experiments in a) field A, b) field B, and c) field C. Open symbols indicate soils without phlogopite application and closed symbols indicate soils with phlogopite application. Error bars indicate the highest and lowest values.

and at harvest. In summary, in all the fields in the first year, exchangeable K in each of the soils with added phlogopite did not considerably differ at rooting stage and at harvest.

From the second year, soil solution K, exchangeable K, and TPB-K in soil with added phlogopite tended to decrease in the fields during the growing period, as observed with control soils. However, these K levels in soil with added phlogopite were always higher than in those without added phlogopite. Notably, the levels of these K forms increased from harvest to the rooting stage in the following year, even in field A, where K fertilizer was only applied in the first year.

The exchangeable ^{137}Cs concentration increased at harvest compared with the rooting stage, and then decreased at the rooting stage in the following year (Fig. 4). The extent of the increase at harvest was smaller in soils with added phlogopite than in soils without phlogopite (Fig. 4). Spearman rank-correlation coefficients between TF and selected soil properties in field experiments are shown in Table 4. The TF was significantly negatively correlated with exchangeable K at all growing stages, TPB-K at the rooting and tillering stages, and soil solution K at the tillering and heading stages. There was a significant negative correlation between soil solution K and exchangeable ^{137}Cs concentration (Fig. 5). Moreover, there was a significant positive correlation between exchangeable ^{137}Cs and brown rice ^{137}Cs concentration (Fig. 6).

3.3. Results of the pot cultivation

The results of the pot cultivation experiment are shown in Table 5. Following 38-days' contact with paddy rice root, the K of phlogopite decreased by more than 90%. There was no significant difference between treatments on the total and exchangeable ^{137}Cs concentration in soils after harvest. The application of phlogopite, except for the condition with rice root contact for 38 days, significantly increased exchangeable K after harvest and decreased the TF of brown rice.

4. Discussion

4.1. Effect of phlogopite application

Maintaining an adequate level of external K mitigates the uptake of radiocesium by crops through competition with radiocesium for root uptake (Zhu and Smolders, 2000). Moreover, an adequate level of external K inhibits the expression of K transporters with high affinity for Cs (Fujimura et al., 2014). The added phlogopite would have maintained the external K level during the growing season of paddy rice by releasing nonexchangeable K to the exchangeable fraction and soil solution K, and reducing TF (Eguchi et al., 2015).

There was a negative correlation between exchangeable ^{137}Cs and soil solution K in all fields (Fig. 5), as observed by Eguchi et al. (2015). Kondo et al. (2014) reported that exchangeable radiocesium contributes to radiocesium transfer to brown rice as well as exchangeable K. In the present study, there was a significant correlation between the concentration of soil exchangeable ^{137}Cs and the concentration of brown rice ^{137}Cs (Fig. 6). However, both soil exchangeable ^{137}Cs and ^{137}Cs uptake by paddy rice were influenced by soil K status (Fig. 5, Table 4).

All the soils contained 2:1-type mica and vermiculite (Table 2), which plays a major role in Cs-fixation in soils in the Fukushima area (Okumura et al., 2018). These minerals can fix K by collapse of the interlayer (Huang, 2005). Excluding soils with added phlogopite at the first year in fields A and B, soil K fractions decreased and exchangeable ^{137}Cs increased from the rooting stage to the heading stage (Figs. 1–4). Moreover, soil exchangeable K and TPB-K were always increased and

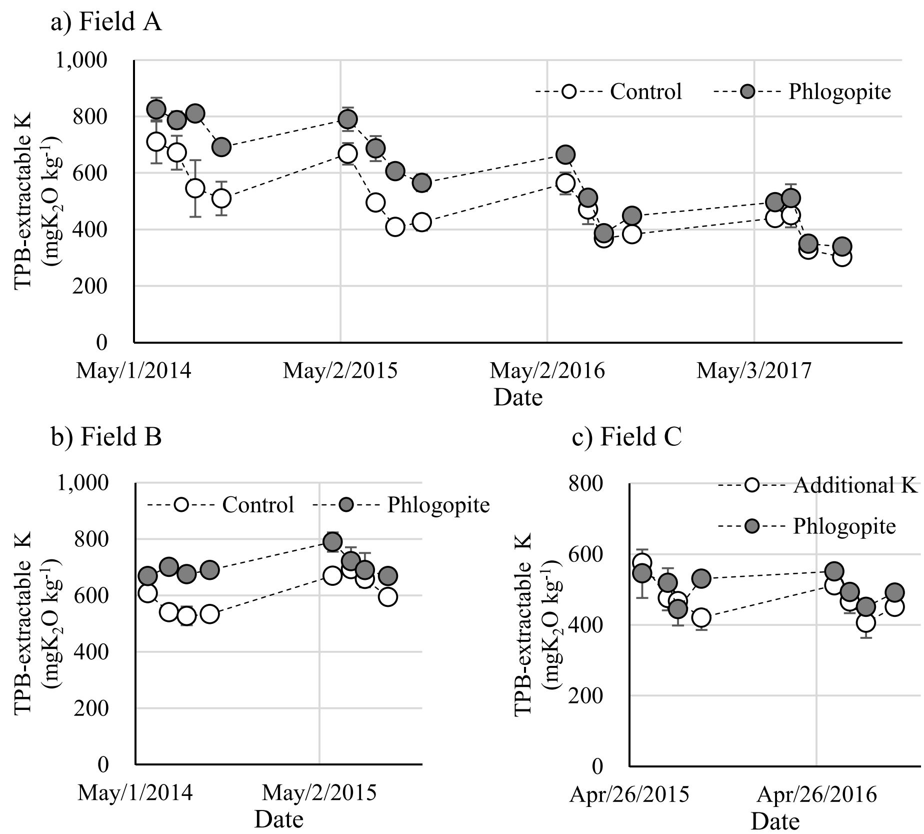

Fig. 3. Changes in soil TPB-extractable K in field experiments in a) field A, b) field B, and c) field C. Open symbols indicate soils without phlogopite application and closed symbols indicate soils with phlogopite application. Error bars indicate the highest and lowest values.

exchangeable ^{137}Cs was always decreased from harvest to rooting stage in the following year (Figs. 1–4). The K supply from K-fertilizer and/or rice residue (i.e., straw, stump, and root) would have induced the collapse of interlayers of 2:1-type clay minerals, and some part of exchangeable Cs would have been strongly retained at newly-formed Cs-selective sites (Absalom et al., 1995; Ogasawara et al., 2019; Kitayama et al., 2020) or fixed into collapsed interlayers (Absalom et al., 1995).

Notably, the concentrations of radiocesium in soil solution directly influence the uptake of radiocesium (Zhu and Smolders, 2000). An increase in exchangeable ^{137}Cs would have reflected an increase in soil solution ^{137}Cs. In fact, Absalom et al. (1995) reported an increase in the solid/liquid distribution coefficient of radiocesium with increasing solution K. Their results strongly suggest that the decrease in soil solution K increases the soil solution radiocesium. Furthermore, an increase in soil solution radiocesium proportionally influences the uptake of radiocesium by crops, whereas the decrease in soil solution K exponentially influences the uptake of radiocesium (Zhu and Smolders, 2000). However, it is not practical to determine soil solution radiocesium in a field study because of low concentrations. In the present study, exchangeable ^{137}Cs were determined as an index of labile ^{137}Cs fractions. In addition to concentrations of labile radiocesium, its solid/liquid distribution is also important for the evaluation of the risk of radiocesium uptake by crops (Zhu and Smolders, 2000). Future studies should elucidate changes in solid/liquid distribution of labile radiocesium induced by K uptake by crop roots.

Mica and K-depleted mica (i.e., vermiculite) fix radiocesium into the interlayer, and reduce the availability of radiocesium to plants (Fanning et al., 1989). It is probable that the release of interlayer K increases the radiocesium selectivity of applied phlogopite, at least under low levels of K-depletion (Le Roux and Rich, 1969; Ogasawara et al., 2017). Eguchi et al. (2015) reported that the application of silt-sized vermiculite prepared from Siilinjärvi phlogopite reduced the concentration of soil-exchangeable ^{137}Cs. In our field experiments, the concentration of exchangeable ^{137}Cs was lower in soils with added phlogopite (Fig. 4). However, the fixation of radiocesium by applied phlogopite was unclear. Because the intensity of K in the soil solution (activity ratio of K to Ca and Mg) controls fixation and/or the strong retention of Cs to 2:1-type clay minerals (Absalom et al., 1995), if applied phlogopite considerably reduces exchangeable radiocesium, it is expected that soils with phlogopite will have lower levels of exchangeable radiocesium compared to soils without phlogopite at comparable soil solution K levels. However, the exchangeable ^{137}Cs of soils with and without added phlogopite did not appear to differ at comparable levels of soil solution K (Fig. 5).

Although the effectiveness of phlogopite on radiocesium fixation was obscure, the concentration of exchangeable ^{137}Cs in soils with added phlogopite was always lower than that in soils without added phlogopite, except in the heading stage in field C during the first year (Fig. 4). This is due to the inhibitory effect of K supply from phlogopite on the release of radiocesium once fixed by indigenous clay minerals (Eguchi et al., 2015), rather than the fixation of radiocesium to the added phlogopite. During the growing season, K adsorption by paddy rice depletes nonexchangeable K in indigenous 2:1-type clay minerals, inducing the simultaneous release of radiocesium (Gommers et al., 2005; Thiry et al., 2005). The K released from added phlogopite would have reduced the uptake of radiocesium through competition with Cs on root adsorption, inhibited gene expression of the high-Cs-affinity transporter, and inhibited radiocesium release from indigenous clay minerals.

Interestingly, there was no significant difference in exchangeable ^{137}Cs between treatments in the pot experiment, even though the level of exchangeable K differed significantly over the range in which exchangeable ^{137}Cs concentration was decreased in field experiments

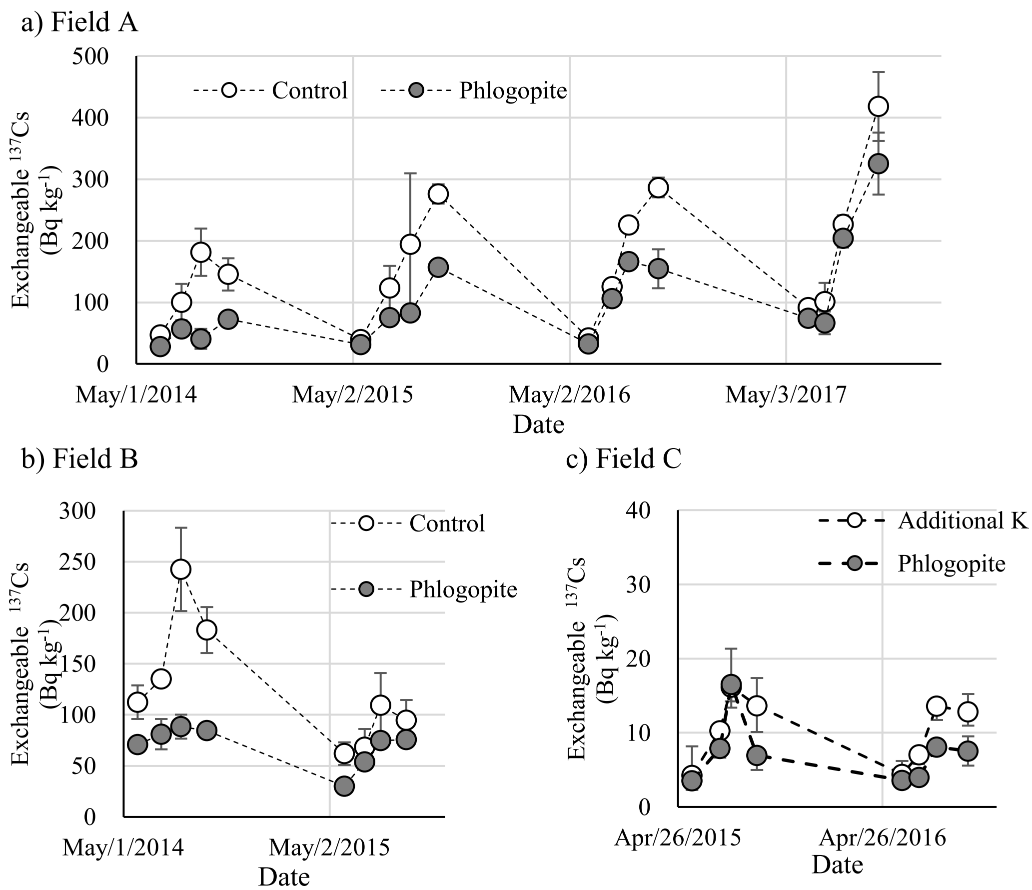

Fig. 4. Changes in soil exchangeable ^{137}Cs in field experiments in a) field A, b) field B, and c) field C. Open symbols indicate soils without phlogopite application and closed symbols indicate soils with phlogopite application. Error bars indicate the highest and lowest values.

Table 4
Spearman correlation coefficients between soil K status and transfer factor on field experiments.

Soil property	Period	Transfer factor
Exchangeable K	Rooting	−0.82 ***
	Tillering	−0.92 ***
	Heading	−0.81 ***
	Harvest	−0.74 **
TPB-extractable K	Rooting	−0.82 ***
	Tillering	−0.67 **
	Heading	−0.47
	Harvest	−0.51 *
Soil solution K	Rooting	0.31
	Tillering	−0.79 **
	Heading	−0.59 *

n = 12 for soil solution K, n = 16 for other factors.
*, $p < 0.05$; **, $p < 0.01$; ***, $p < 0.001$.

(Figs. 2 and 4, Table 5). The effectiveness of phlogopite to fix ^{137}Cs was unclear, as observed in the field experiments, even though Eguchi et al. (2015) reported ^{137}Cs fixation by fine (10–20 μm) vermiculite prepared from same Finnish phlogopite by K-depletion. The ability of micaceous 2:1-type clay mineral to selectively retain radiocesium, and the consequent fixation of radiocesium, is influenced by particle size (Sato et al., 2016) and the degree of K-depletion (Kitayama et al., 2020). In the present study, in field and pot experiments, coarse particle size (450 μm on average), rather than the degree of K-depletion, would be the major reason for the ineffectiveness of reducing exchangeable radiocesium with phlogopite application, because the phlogopites used in the pot experiment covered a wide range of K-contents (8–100% of the original phlogopite, Table 5). In fact, Sato et al. (2016) reported that fine vermiculite (<25 μm) had a higher capacity for the selective retention of radiocesium than coarser vermiculite. Moreover, this suggests that, in contrast to soils in field experiments, the concentration of exchangeable ^{137}Cs in the soil used in pot experiments was not influenced by exchangeable K.

Compared to the soils in field experiments, the mineralogy of the clay fraction of soil used in pot experiments has higher degree of Al-interlayering of 2:1-type clay minerals (Table 2). As previously stated, the fixation/release of K by 2:1-type clay minerals would have altered the concentration of exchangeable ^{137}Cs. In the pot experiment, a high degree of Al interlayering may have inhibited K fixation by propping interlayers (Rich and Black, 1964), consequently inhibiting the change in exchangeable ^{137}Cs concentration. Therefore, in soils with 2:1-type clay minerals, with a high degree of Al-interlayering, an indirect effect of phlogopite through the inhibited release of fixed ^{137}Cs cannot be expected. However, in pot experiments, the application of phlogopite reduced TF by more than 80% (Table 5). Applying phlogopite would be effective to reduce TF, even when the indirect effects via inhibited release of fixed ^{137}Cs are not significant.

4.2. *TPB-K as a basis of fertilizer recommendations*

Exchangeable K and TPB-K at the rooting stage were strongly negatively correlated with TF (Table 4, Fig. 7), whereas soil solution K at the rooting stage was not correlated with TF (Table 4). However, soil solution K at the tillering and heading stages was significantly negatively related to TF (Fig. 8). Plant roots take up K from soil solution; hence, the absorption of radiocesium by plant roots is regulated by the concentration of K in soil solution (Zhu and Smolders, 2000). When plant roots

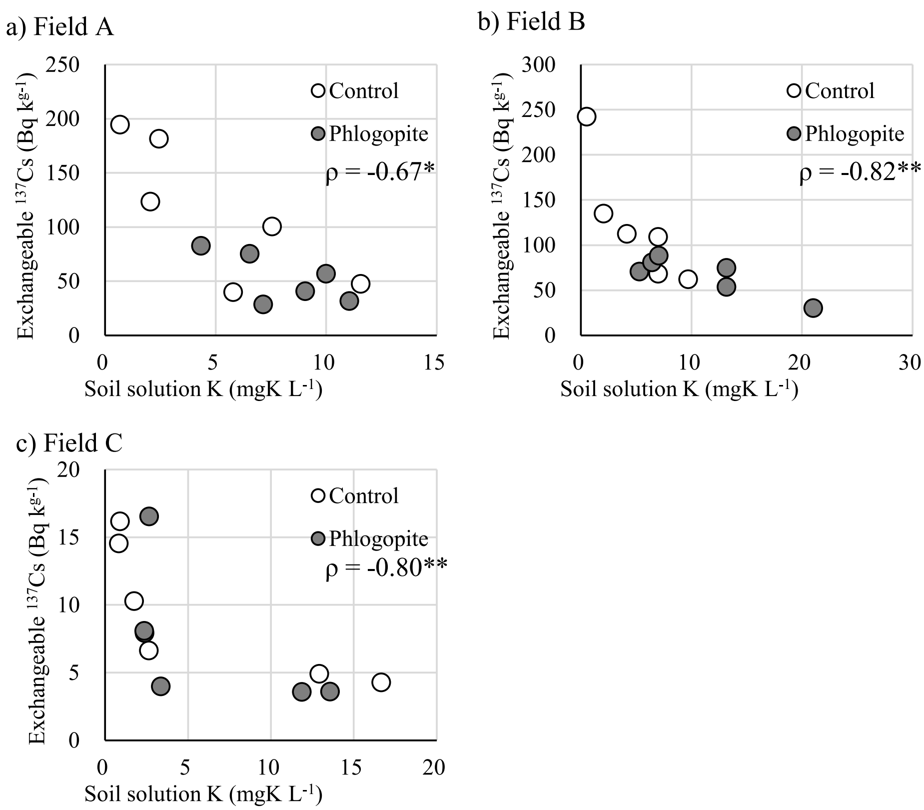

Fig. 5. Relationship between soil solution K and exchangeable ^{137}Cs. Open symbols indicate soils without phlogopite application and closed symbols indicate soils with phlogopite application in field trials. **, $p < 0.01$.

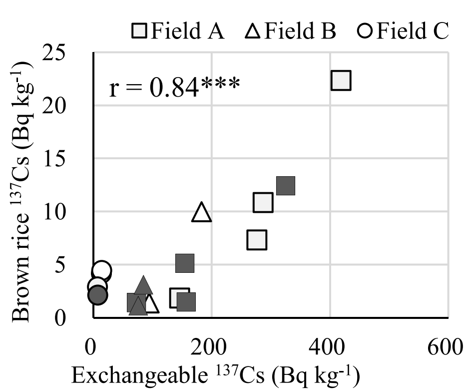

Fig. 6. Relationship between soil exchangeable ^{137}Cs concentration at harvest and brown rice ^{137}Cs concentration. Square, triangle, and circle symbols indicate field A, B and C, respectively. Open symbols indicate soils without phlogopite application and closed symbols indicate soils with phlogopite application in field trials. ***, $p < 0.001$.

take up K, the replenishment of soil solution K is affected by release from labile solid-state K fractions such as exchangeable K and TPB-K (Huang, 2005). Therefore, for radiocesium-polluted fields, a purpose of determining labile solid-state K is to evaluate the level of soil solution K level when paddy rice absorbs radiocesium. Fukushima Prefectural Government (2014) recommends maintaining the concentration of K in the soil solution above 7 mgK L^{-1} from the panicle formation to the ripening stage of paddy rice. Exchangeable K at rooting stage was significantly positively correlated with soil solution K at tillering stage; however, there was no significant correlation between exchangeable K at rooting stage and soil solution K at heading stage (Table 6). TPB-K at rooting stage was significantly positively correlated with soil solution K at both tillering and heading stages (Table 6). TPB-K might be a better indicator of plant-available K than exchangeable K as a basis of fertilizer recommendations.

Furthermore, the method used for TPB extraction in this study has been proposed for grassland (Carey et al., 2010). Some modification of the method for rice culture in areas contaminated by radiocesium would be needed, because the objective of a soil K test in radiocesium-polluted fields differs from that in unpolluted fields. In general, a soil K test is conducted to determine the adequate K application rate required to secure crop yield. In this field experiment, there was no considerable increase in yield following phlogopite application, indicating that the level of K in soil solution was sufficient to secure yield, even without phlogopite application, whereas TF was considerably decreased following phlogopite application (Table 3). These results indicate that higher levels of K in soil solution are required for crop production in fields polluted with radiocesium compared to non-polluted fields.

Moreover, the ability to acquire nonexchangeable K from mica differs among crops (Mengel and Rahmatullah, 1994; Sugiyama and Ae, 2000). The method of TPB extraction used in the present study is intended for use in grassland, where the rate of K application is low. In contrast, after the FDNPP accident, an increased rate of K-fertilizer was applied to paddy fields in Fukushima and the surrounding area. Eguchi et al. (2015) reported that TPB-K before planting showed much higher correlation with TF than exchangeable K in a pot experiment. However, in the present field experiment, the correlation coefficient between TPB-K at the rooting stage and TF was only slightly higher than that between exchangeable K and TF (Table 4, Fig, 7). In the pot cultivation experiment, the decreased level of soil K induced by the uptake of K by crops is more rapid than that observed in the field study (Ishikawa et al., 2017). Conversely, the release of nonexchangeable K would be less intensive in the field study than in the pot cultivation experiment. Further research on milder methods of extracting nonexchangeable K is needed for radiocesium-contaminated paddy field that have received an

Table 5
Summary of the pot experiment results.

Treatment[a]	Phlogopite	Soils after harvest			Brown rice[b]	
	K content	Total ^{137}Cs	Exchangeable ^{137}Cs	Exchangeable K	^{137}Cs concentration	^{137}Cs transfer factor
	(g K_2O kg^{-1})	(kBq kg^{-1})	(Bq kg^{-1})	(mg K_2O kg^{-1})	(Bq kg^{-1})	($\times 10^3$)
Control		6.31 ± 0.28 a	321 ± 35 a	26 ± 4 f	248 ± 31 a	39 ± 5 a
+Phlogopite (Original)	87	6.19 ± 0.27 a	285 ± 20 a	155 ± 8 a	40 ± 3 d	6.4 ± 0.4 d
+Phlogopite (4 days contact)	72	6.24 ± 0.33 a	285 ± 30 a	122 ± 4 b	53 ± 18 d	8.6 ± 3.4 d
+ Phlogopite (8 days contact)	50	6.19 ± 0.38 a	332 ± 35 a	90 ± 11 c	78 ± 15 cd	13 ± 3 cd
+Phlogopite (14 days contact)	38	6.37 ± 0.35 a	288 ± 6 a	70 ± 8 d	118 ± 9 bc	19 ± 2 bc
+Phlogopite (23 days contact)	20	6.20 ± 0.43 a	307 ± 25 a	44 ± 4 e	160 ± 25 b	26 ± 6 b
+Phlogopite (38 days contact)	7	6.06 ± 0.43 a	294 ± 18 a	25 ± 6 f	229 ± 27 a	38 ± 7 a

Different letters represent significant differences according to the Tukey's HSD test ($p < 0.05$).
[a] Control indicates that phlogopite was not applied. Original indicates the applied phlogopite was not subjected to the contact, days represent the duration of the contact with paddy rice root in K-deficient Kimura-B nutrient solution.
[b] 15% moisture level, no sieving.

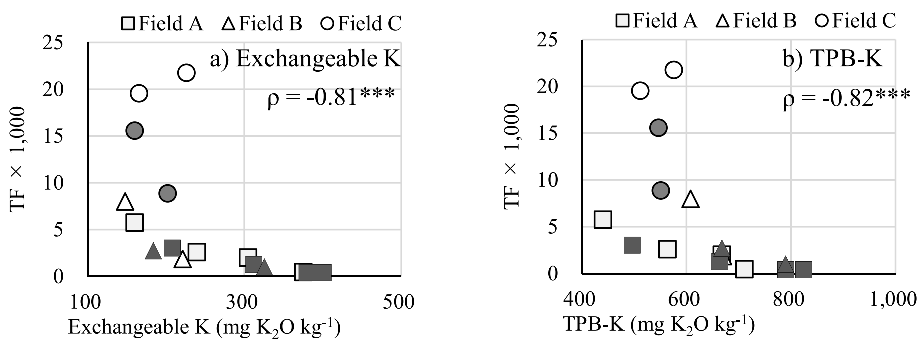

Fig. 7. Relationship between a) exchangeable K and b) TPB-K at the rooting stage, and TF in field experiments. Square, triangle, and circle symbols indicate field A, B and C, respectively. Open symbols indicate soils without phlogopite application and closed symbols indicate soils with phlogopite application. ***, $p < 0.001$.

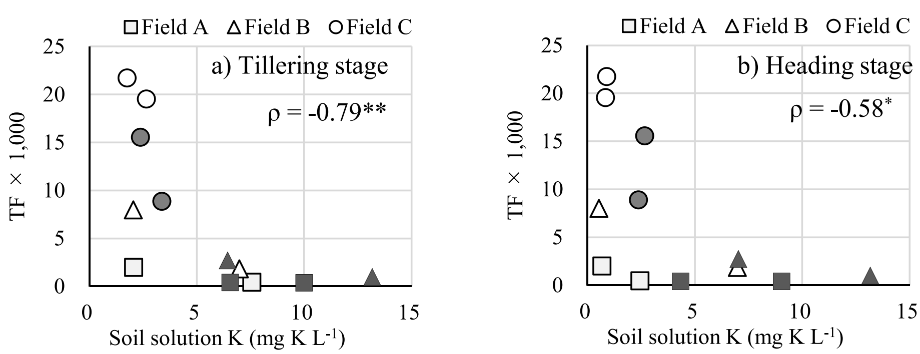

Fig. 8. Relationship between soil solution K a) at the tillering stage and b) at the heading stage, and TF in field experiments. Square, triangle, and circle symbols indicate field A, B and C, respectively. Open symbols indicate soils without phlogopite application and closed symbols indicate soils with phlogopite application. *, $p < 0.05$; **, $p < 0.01$.

increased rate of K-fertilization. Recently, Kurokawa et al. (2020) proposed a criterion of nonexchangeable K (i.e. boiling HNO_3 extractable K minus exchangeable K) > 50 mg K_2O 100 g^{-1} as a threshold to differentiate soils with low radiocesium transfer risk, despite having a low level of exchangeable K, from soils with a high radiocesium transfer risk. Various extraction methods should be compared and improved in future.

4.3. Practicality of coarse siilinjärvi phlogopite application

A phlogopite application rate of 5 t ha^{-1} corresponded to 48 kg K_2O ha^{-1}, equivalent to 6-years' conventional application in Fukushima Prefecture (Fukushima Prefectural Government, 2014). The effectiveness of phlogopite application lasted for the duration of the field experiment. In field C, where K depletion was intensive, phlogopite application was more effective than additional K application for 2 years. In field A, although K fertilizer was only applied in the first year as a basal fertilizer, the effect of the added phlogopite lasted for 4 years. In pot cultivation, even when about 55% of K (corresponding to 3-years' of K-fertilizer application) was removed from phlogopite, the TF was less than half that in the control (Table 5). The effectiveness of phlogopite application would be more durable if K fertilizer was applied appropriately.

When fine Siilinjärvi phlogopite is applied to soil, the increase in TPB-K is almost equal to the total K in the applied phlogopite (Eguchi et al., 2015). Assuming a 15-cm of top-soil depth, which is the target depth of soil improvement in Fukushima Prefecture (Fukushima Prefectural Government, 2014), and the average bulk density of paddy fields in Japan (0.95; Nakai and Obara, 2002), fine Siilinjärvi phlogopite application at a rate of 5 t ha^{-1} should increase TPB-K by 336 mg kg^{-1}. In the present study, increases in TPB-K by coarse Siilinjärvi phlogopite

Table 6
Pearson correlation coefficients between soil K status at the rooting stage and that at later stages in field experiments.

		Exchangeable K	TPB-extractable K	Soil solution K
		Rooting	Rooting	Rooting
Exchangeable K	Tillering	0.85***	0.79***	−0.02
	Heading	0.74**	0.85***	−0.07
	Harvest	0.73**	0.82***	0.03
TPB-extractable K	Tillering	0.59*	0.86***	−0.08
	Heading	0.46	0.82***	−0.09
	Harvest	0.37	0.79***	−0.10
Soil solution K	Tillering	0.62*	0.83***	0.30
	Heading	0.29	0.71*	0.29

n = 12 for soil solution K, n = 16 for exchangeable K and TPB-K.
*p, <0.05; **p < 0.01; ***p < 0.001.

application at the rooting stage in the first year were 109 mg kg^{-1} and 25 mg kg^{-1} in field A and B, respectively (data not shown). Even though it was obvious that K release from coarse Siilinjärvi phlogopite was slower than that from the fine one, the K-depletion results following contact with paddy-rice root indicated that more than 90% of K in coarse Siilinjärvi phlogopite was plant-available (Table 5). The results imply the effectiveness of coarse Siilinjärvi phlogopite application lasts longer than that of the fine one. In addition to particle size, chemical composition, which differs across localities, significantly influences the degree of binding of nonexchangeable K (Fanning et al., 1989). Therefore, the effectiveness of phlogopite application in decreasing radiocesium uptake by crops would vary across localities.

In summary, the application of coarse Siilinjärvi phlogopite found to be an effective countermeasure to reduce the uptake of radiocesium by crops.

5. Conclusion

The application of coarse Siilinjärvi phlogopite maintained soil solution K and exchangeable K at higher levels during the growing period of paddy rice, and thus decreased TF. The relationship between soil K status at the rooting and later stages suggested that TPB-K was a better indicator of soil K plant-availability as a basis for fertilizer recommendations than exchangeable K. In the 4-year field experiment without K application, except for basal fertilizer in the first year, the effectiveness of phlogopite application lasted for the duration of the experiment. Moreover, in the pot experiment, phlogopite reduced TF to less than half that of the control, even when around 55% of its K, corresponding to 3-years' K-fertilizer application, was removed prior to application. Therefore, we conclude that the application of coarse Siilinjärvi phlogopite would be an effective countermeasure to reduce the uptake of radiocesium by crops.

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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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvrad.2021.106687.

References

Absalom, J.P., Young, S.D., Crout, N.M.J., 1995. Radio-caesium fixation dynamics: measurement in six Cumbrian soils. Eur. J. Soil Sci. 46, 461–469.
Al Ani, T., 2013. Mineralogy and petrography of Siilinjärvi carbonatite and glimmerite rocks, eastern Finland. Geol. Surv. Finl. Arch. Rep. 164.
Carey, P.L., Curtin, D., Scott, C.L., 2010. An improved procedure for routine determination of reserve-K in pastoral soils. Plant Soil 341, 461–472.
Cox, A., Joern, B., 1999. Plant-available potassium assessment with a modified sodium tetraphenylboron method. Soil Sci. Soc. Am. J. 63, 902–911.
Eguchi, T., Ohta, T., Ishikawa, T., Matsunami, H., Takahashi, Y., Kubo, K., Yamaguchi, N., Kihou, N., Shinano, T., 2015. Influence of the nonexchangeable potassium of mica on radiocesium uptake by paddy rice. J. Environ. Radioact. 147, 33–42.
Fanning, D., Keramidas, V., El-desoky, M., 1989. Micas. In: Bigham, J., Dixon, J., Milford, M., Roth, C., Weed, S. (Eds.), Minerals in Soil Environments. Soil Science Society of America, Madison, pp. 551–634.
Fernández, F.G., Brouder, S.M., Beyrouty, C.A., Volenec, J.J., Hoyum, R., 2008. Assessment of plant-available potassium for No-till, rainfed soybean. Soil Sci. Soc. Am. J. 72, 1085–1095.
Fujimura, S., Yoshioka, K., Saito, T., Sato, Mutsuto, Sato, Makoto, Sakuma, Y., Muramatsu, Y., 2013. Effects of Applying Potassium, Zeolite and vermiculite on the radiocesium uptake by rice plants grown in paddy field soils collected from Fukushima Prefecture. Plant Prod. Sci. 16, 166–170.
Fujimura, S., Ishikawa, J., Sakuma, Y., Saito, T., Sato, M., Yoshioka, K., 2014. Theoretical model of the effect of potassium on the uptake of radiocesium by rice. J. Environ. Radioact. 138, 122–131.
Fujimura, S., Eguchi, T., Matsunami, H., Ota, T., Murakami, T., Ishikawa, T., Makino, T., Akahane, I., Kamiya, T., Aono, K., Naka, T., Okushima, S., 2016. Transfer factors of radiocesium to brown rice in the decontaminated paddy field. Jpn. J. Crop Sci. 85, 211–217.
Fukushima Prefectural Goverment, 2006. Fertilizer recommendations. https://www.pref.fukushima.lg.jp/sec/36021d/kankyou-nogyou-sehikijyun.html. (Accessed 3 November 2020).
Fukushima Prefectural Goverment, 2014. Countermeasures to reduce radiocesium uptake by paddy rice. https://www.pref.fukushima.lg.jp/uploaded/attachment/49127.pdf. (Accessed 3 November 2020).
Gommers, A., Thiry, Y., Delvaux, B., 2005. Rhizospheric mobilization and plant uptake of radiocesium from weathered micas: I. Influence of potassium depletion. J. Environ. Qual. 34, 2167–2173.
Hinsinger, P., Jaillard, B., 1993. Root-induced release of interlayer potassium and vermiculitization of phlogopite as related to potassium depletion in the rhizosphere of ryegrass. J. Soil Sci. 44, 525–534.
Huang, P., 2005. Chemistry of potassium in soils. In: Tabatabai, M., Sparks, D. (Eds.), Chemical Processes in Soils. Soil Science Society of America, Madison, pp. 227–292.
Ishikawa, T., Sakuma, Y., Saito, T., Eguchi, T., Fujimura, S., Matsunami, H., Ota, T., Nakai, M., Obara, H., 2002. Data sheets. In: Kono, E. (Ed.), Glossary of Soil Physics. Yokendo, Tokyo, Japan, pp. 177–183.
Jackson, B., 1985. A modified sodium tetraphenylboron method for the routine determination of reserve-potassium status of soil. N. Z. J. Exp. Agric. 13, 253–262.
Kato, N., Kihou, N., Fujimura, S., Ikeba, M., Miyazaki, N., Saito, Y., Eguchi, T., Itoh, S., 2015. Potassium fertilizer and other materials as countermeasures to reduce radiocesium levels in rice: results of urgent experiments in 2011 responding to the Fukushima Daiichi Nuclear Power Plant accident. Soil Sci. Plant Nutr. 61, 179–190.
Kitayama, R., Yanai, J., Nakao, A., 2020. Ability of micaceous minerals to adsorb and desorb caesium ions: effects of mineral type and degree of weathering. Eur. J. Soil Sci. 71, 641–653.
Kondo, M., Maeda, H., Goto, A., Nakano, H., Kiho, N., Makino, T., Sato, M., Fujimura, S., Eguchi, T., Hachinohe, M., Hamamatsu, S., Ihara, H., Takai, T., Arai-Sanoh, Y., Kimura, T., 2014. Exchangeable Cs/K ratio in soil is an index to estimate accumulation of radioactive and stable Cs in rice plant. Soil Sci. Plant Nutr. 61, 133–143.
Kurokawa, K., Nakao, A., Wakabayashi, S., Fujimura, S., Eguchi, T., Matsunami, H., Yanai, J., 2020. Advanced approach for screening soil with a low radiocesium transfer to brown rice in Fukushima based on exchangeable and nonexchangeable potassium. Sci. Total Environ., 140458
Le Roux, J., Rich, C., 1969. Ion selectivity of micas as influenced by degree of potassium depletion. Soil Sci. Soc. Am. Proc. 33, 684–690.
Li, T., Wang, H., Zhou, Z., Chen, X., Zhou, J., 2016. A new grading system for plant-available potassium using exhaustive cropping techniques combined with chemical analyses of soils. Sci. Rep. 6, 37327.
Li, X., Zhang, Y., Wang, W., Khan, M.R., Cong, R., Lu, J., 2018. Establishing grading indices of available soil potassium on paddy soils in Hubei province, China. Sci. Rep. 8, 16381.
Mengel, K., Rahmatullah, 1994. Exploitation of potassium by various crop species from primary minerals in soils rich in micas. Biol. Fertil. Soils 17, 75–79.
Ministry of Agriculture, Fishery and Forestry, 2015a. Factors responsible for the production of rice with high radioactive cesium concentration and its countermeasure. (Accessed 3 November 2020). second ed.

Ministry of Agriculture, Fishery and Forestry, 2015b. Summary of the basic plan for food, agriculture and rural areas. https://www.maff.go.jp/e/policies/law_plan/attach/pdf/index-2.pdf. (Accessed 15 December 2020).
Nobori, T., Kobayashi, N.I., Tanoi, K., Nakanishi, T.M., 2014. Effects of potassium in reducing the radiocesium translocation to grain in rice. Soil Sci. Plant Nutr. 60, 772–781.
Ogasawara, S., Nakao, A., Yanai, J., 2017. A stepwise change of frayed edge site content in biotite in response to the gradual release of potassium from the interlayers. Soil Sci. Plant Nutr. 63, 529–535.
Ogasawara, S., Eguchi, T., Nakao, A., Fujimura, S., Takahashi, Y., Matsunami, H., Tsukada, H., Yanai, J., Shinano, T., 2019. Phytoavailability of 137Cs and stable Cs in soils from different parent materials in Fukushima, Japan. J. Environ. Radioact. 198, 117–125.
Okumura, M., Kerisit, S., Bourg, I.C., Lammers, L.N., Ikeda, T., Sassi, M., Rosso, K.M., Machida, M., 2018. Radiocesium interaction with clay minerals: theory and simulation advances Post–Fukushima. J. Environ. Radioact. 189, 135–145.
Paasikallio, A., 1999. Effect of biotite, zeolite, heavy clay, bentonite and apatite on the uptake of radiocesium by grass from peat soil. Plant Soil 206, 213–222.
Rich, C., Black, W., 1964. Potassium exchange as affected by cation size, pH and mineral structure. Soil Sci. 97, 384–390.
Saito, T., Ohkoshi, S., Fujimura, S., Iwabuchi, K., Saito, M., Nemoto, T., Sato, Mutsuto, Sato, Makoto, Yoshioka, K., Tsukada, H., 2012. Effect of potassium application on root uptake of radiocesium in rice. In: Proceedings of International Symposium on Environmental Monitoring and Dose Estimation of Residents after Accident of TEPCO's Fukushima Daiichi Nuclear Power Station. Kyoto University Research Reactor Institute Press, Kyoto, pp. 165–169.
Sato, H., Kaneko, Y., Morita, M., Tamura, K., Yamagishi, A., 2016. Application of vermiculite clay for selective adsorption of cesium ions by high performance liquid chromatography. Clay Sci. 20, 7–11.
Scott, A.D., Smith, S.J., 1966. Suceptibility of interlayer potassium in micas to exchange with sodium. In: Clays and Clay Minerals, pp. 69–81.
Sugiyama, M., Ae, N., 2000. Uptake response of crops to potassium from andosol and potassium-bearing minerals applied to andosol. Jpn. J. Soil Sci. Plant Nutr. 71, 786–793.
Thiry, Y., Gommers, A., Iserentant, A., Delvaux, B., 2005. Rhizospheric mobilization and plant uptake of radiocesium from weathered micas: II. Influence of mineral alterability. J. Environ. Qual. 34, 2174–2180.
Zhu, Y.-G., Smolders, E., 2000. Plant uptake of radiocaesium: a review of mechanisms, regulation and application. J. Exp. Bot. 51, 1635–1645.