Comparative dynamics of potassium and radiocesium in soybean with different potassium application levels

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ABSTRACT

We conducted a field experiment in soybean with different levels of K application to elucidate the comparative dynamics of ^{137}Cs and K. The inventory of K in the shoots increased substantially from the fifth trifoliate stage to the full seed stage, and as the absorption of K increased, so too did the absorption of ^{137}Cs. Overall, the effect of K application was much greater in terms of ^{137}Cs dynamics than K dynamics or biomass production. K application reduced not only the accumulation of ^{137}Cs in the shoots, but also the distribution of ^{137}Cs to the grains. However, the decrease of ^{137}Cs distribution to the grain had a much smaller effect on ^{137}Cs accumulation in the grains than ^{137}Cs absorption. A positive correlation was also observed between the exchangeable ^{137}Cs/K ratio in the soil and the ^{137}Cs/K ratio in the shoots for each growth stage, and the ^{137}Cs/K ratios in the shoots at the full seed and full maturity stage were much higher than those at the fifth trifoliate and full bloom stage under the same exchangeable ^{137}Cs/K ratio in the soil. These findings suggest a decrease in the discrimination of ^{137}Cs from K during absorption after the full bloom stage. As a result of this and the increase in soil-exchangeable ^{137}Cs/K with growth, radiocesium was more transferable to the shoots after the full bloom stage. Overall, these results suggest that lowering the soil-exchangeable radiocesium/potassium ratio after the full bloom stage by increasing K availability could efficiently reduce the transfer of radiocesium to the grains.

1. Introduction

The accident at Tokyo Electric Company's Fukushima Dai-ichi Nuclear Power Plant (FDNPP), caused by the Great East Japan Earthquake and subsequent Tsunami in March 2011, resulted in widespread contamination of agricultural land in eastern Japan with radionuclides, particularly in parts of Fukushima Prefecture. Immediately after the accident, the major radionuclides found in plants and environmental resources, such as soil and water, were ^{131}I and radiocesium (^{134}Cs and ^{137}Cs). Although ^{131}I (half-life: 8 days) decays within a few months, radiocesium is retained in the soil for much longer because of the length of the ^{137}Cs half-life (30.2 years), and its transfer to crops is therefore a long-term problem. With some exceptions, the Japanese government commissioned physical decontamination works, which included topsoil (depth of 15 cm) removal and soil inversion for soils exceeding 5 kBq kg dry weight of radiocesium (Yamaguchi et al., 2016). Ten years have passed since the accident and all decontamination work has been completed, except in some difficult-to-return zones near the FDNPP, where the annual cumulative radiation doses are greater than 50 mSv, and entry and lodging are prohibited.

Studies on the absorption and accumulation of radiocesium in crops since the accident have shown that potassium fertilization is the most effective and practical countermeasure for reducing radiocesium transfer from the soil to the edible parts of crops. Because cesium has similar chemical and physiological characteristics to potassium, both potassium and cesium are expected to have similar behaviors in soil-plant systems, and cesium absorption from the soil is competitively decreased by the application of potassium fertilizer (Shaw and Bell 1991; Smolders et al.,

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1997). In paddy rice (*Oryza sativa* L.), radiocesium transfer to the grains decreased with increasing exchangeable potassium (1 mol L^{-1} ammonium acetate extractable potassium) concentrations in the soil (Tsukada et al., 2002a; Kato et al., 2015), with similar results obtained in buckwheat (*Fagopyrum esculentum* Moench) and vegetables (Kobayashi 2014; Kobayashi et al., 2014; MAFF 2014; Kubo et al., 2015, 2017). Thus, the additional application of potassium fertilizer has been widely implemented in low-contamination areas (<5 kBq kg^{-1} dry weight of radiocesium) and decontaminated areas. Information about the various case studies and the countermeasures published on websites and in technical reports in the early stages after the accident has been summarized by Yamaguchi et al. (2016).

Soybean (*Glycine max* (Merr.) L.) is one of the most important crops in Japan, with almost all being used in food processing, notably tofu production, which is made by concentrating soy protein. Other important soybean products, such as miso and soy sauce, which are essential seasonings in traditional Japanese cooking, are produced via fermentation. Immediately after the accident, we conducted a survey of exchangeable potassium concentrations in the soil and radiocesium concentrations in soybean grains to determine the level of soil-exchangeable potassium required to minimize the radiocesium concentrations. Accordingly, a negative correlation was observed between the soil-exchangeable potassium concentration and the soil-to-grain transfer factor (TF) of radiocesium, which is expressed as a ratio of radioactivity in the grains to that in the soil (Bq kg^{-1} (Bq kg^{-1})$^{-1}$) (MAFF 2015). Based on these results, an exchangeable potassium concentration of ≥ 250 mg kg^{-1} of K$_2$O in the soil prior to the use of basal fertilizer at a conventional rate is now widely recommended in soybean cultivation, except during initial cultivation after the accident or in fields with a high TF, where the recommended application level was increased to 500 mg kg^{-1} of K$_2$O (MAFF 2015). The rate of additional potassium fertilizer application is calculated from the exchangeable potassium concentration of the soil before cultivation, assuming a soil bulk density of 1.0 Mg m^{-3} and soil depth of 15 cm. As a result, the percentage of soybean grains exceeding the present standard limits (100 Bq kg^{-1} fresh weight for radiocesium in food, established by the Japanese Ministry of Health, Labor and Welfare, 2011 in April 2012) has decreased annually, and the radiocesium concentration in soybean grains has not exceeded this limit since 2015 (Japanese Ministry of Agriculture, Forestry and Fisheries). According to MAFF (2015), the exchangeable potassium concentration of the soil is relatively low in fields where the radiocesium concentration of the soybean grains exceeds the above limit before 2015. Although additional potassium fertilization is a primary factor in reducing the percentage of soybean grains exceeding this limit, the decrease in soil-exchangeable radiocesium concentrations due to physical decay and fixation to clay minerals in the soil (Tsumura et al., 1984; Ehlken and Kirchner 2002) has also contributed to the reduction.

Radiocesium transfer from the soil to the edible parts of a plant has been widely studied, but most research has focused on radiocesium, with few reports on the comparative dynamics of radiocesium and potassium. Cesium and potassium are expected to have similar behaviors in soil-plant systems; however, the radiocesium/potassium ratio is not uniform in paddy rice, indicating different behaviors (Tsumura et al., 1984; Tsukada et al., 2002b; Kondo et al., 2015). Soybean and buckwheat show considerably high TF of ^{134}Cs (Broadley and Willey 1997), and monitoring inspections after the accident also indicated that the percentage of soybean grains exceeding the limit was higher compared with other crops (Nihei and Hamamoto 2019). Potassium application is costly and labor-intensive; therefore, it is important to understand the behavior of potassium and radiocesium in soybean soil-plant systems to establish strategies for reducing radiocesium concentrations with minimal labor and cost. In the present study, we therefore examined the comparative dynamics of radiocesium (^{137}Cs) and potassium (K) in soybean. To do so, we conducted a field experiment with five levels of soil-exchangeable K and examined the variability in the absorption and distribution of ^{137}Cs compared with K.

2. Materials and methods

2.1. Field management and experimental design

The field experiment was conducted in the city of Date in Fukushima Prefecture in 2015. The soil type in the field was gray lowland soil based on the classification of cultivated soils in Japan. The soil texture was sandy clay (27.0% clay, 7.5% silt, and 65.5% sand), and the ^{137}Cs concentration from the surface to a depth of 15 cm was 2.74 kBq kg^{-1} in mid-May 2015. The physical and chemical properties of the soil are shown in Table 1.

Although the additional application of K fertilizer had been widely implemented before the conventional rate of basal fertilizer was applied, the additional K fertilizer was applied together with the basal dressing of K fertilizer (80 mg kg^{-1} soil of K$_2$O, assuming that the soil bulk density was 1.0 Mg m^{-3} and soil depth was 15 cm) in this study. One of the following five levels of soil-exchangeable K was applied to the plots prior to cultivation: no K fertilization (K0), 230 (K230), 380 (K380), 580 (K580), and 780 mg kg^{-1} (K780) of K$_2$O. For all plots, a conventional rate of N and P were applied as follows: 20 mg kg^{-1} of N and 80 mg kg^{-1} of P$_2$O$_5$ as basal dressing on June 3, 2015, followed by 40 mg kg^{-1} of N as top dressing on 29 July.

The experimental design was a randomized complete block with three replications. The area of each plot was 27.3 m^2. The soybean cultivar 'Tachinagaha' was sown at a density of 50 kg seed ha^{-1} just after the application of basal dressing (3 June). The hill space between planted rows was 0.7 m.

2.2. Sample collection, sample preparation, and chemical analysis

The soybean shoots were sampled at the fifth trifoliate stage (9 July), the full bloom stage (28 July), the full seed stage (17 September), and the full maturity stage (20 October), hereafter referred to as V5, R2, R6, and R8, respectively. After sampling at the R6 growth stage, the shoots were covered with a polyethylene mesh bag (Nandemo-kaisyubukuro, Nihon Matai Co., Ltd., Japan; 6 mm mesh, 100 × 120 cm) to collect fallen leaves. The leaves turned yellow after the R6 growth stage, and most had fallen by the R8 growth stage; thus, the leaves collected at this late stage all represented fallen leaves. The shoots were divided into three parts (leaf blade, petiole, and stem) at the V5 and R2 growth stages, and into five parts (leaf blade, petiole, stem, pod, and grain) at the R6 and R8 growth stages. Then, the samples were washed with tap water and dried for at least 48 h at 80 °C in a ventilated oven. After drying, the dry weight of the samples was measured. The dried samples were cut using a cutting mill (SM300, Retsch, Germany) with a 4-mm bottom sieve, after which a portion of the samples was pulverized with a blender (D3V-10, Osaka Chemical Co., Japan) and used for gamma-ray spectrometry. About 0.2 mg of the pulverized samples was digested with 2.5 mL of nitric acid and 0.5 mL of hydrogen peroxide

Table 1
Physical and chemical properties of the soil.

Soil texture		Sandy clay
Major clay minerals		Illite, smectite
Soil texture (%)	Clay	27.0
	Silt	7.5
	Coarse sand	44.2
	Fine sand	21.2
pH (H$_2$O)		5.2
Phosphate absorption coefficient (mg Kg^{-1})		4940
Cation exchange capacity (cmol kg^{-1} dry weight)		12.1
^{137}Cs concentration (kBq kg^{-1})		2.74

using the heat block acid digestion system (DigiPREP LS, SCP SCIENCE, Canada). The acid digests were used to determine the K concentrations of the plant samples using inductively coupled plasma-atomic emission spectroscopy (ICP-AES; Vista-MPX, Varian, USA).

Soil from the surface to a depth of 15 cm was sampled with a round worm scoop (Fujiwara Scientific Company Co., Ltd., Japan) at the same time the plants were sampled. Soil samples were collected from 10 points in each plot and combined to make a composite sample. After visible plant residues and stones were removed, the soil samples were air-dried and passed through a 2-mm sieve using a Dust Shield Automatic Mill and Screen for Soil RK4II (DIK-2610, Daiki Rika Kogyo Co., Ltd., Japan). Exchangeable ^{137}Cs was extracted at a soil-to-solution ratio of 1:10 in 1 mol L^{-1} ammonium acetate (pH 7.0) with shaking for 1 h, according to Tsukada et al. (2008). The sieved soil samples and the ammonium acetate extracts were then used for gamma-ray spectrometry. Exchangeable K was extracted using 1 mol L^{-1} ammonium acetate at a soil-to-solution ratio of 1:20 with 1 h shaking and the concentration of K was determined via ICP-AES.

2.3. Gamma-ray spectrometry

The ^{137}Cs concentrations of the plant and soil samples were determined using high-purity germanium detectors (GC2520-7500SL and GC4020-7500SL, Canberra, USA). The ^{137}Cs concentrations of the plant samples were measured using a 0.7 L Marinelli beaker or cylindrical polypropylene container (U-8 container, 65 mm in height and 50 mm in diameter; RIG, Japan). The ^{137}Cs concentrations of the soil and ammonium acetate extracts (for determining exchangeable ^{137}Cs) were measured directly in a U-8 container and 0.7 L Marinelli beaker, respectively, using gamma lines at 661.6 keV. The counting uncertainties of ^{137}Cs were kept lower than 10% and concentrations were time-corrected on each sampling day.

2.4. Statistical analysis

All statistical analyses were conducted using statistical software (BellCurve for Excel ver. 3.00, Social Survey Research Information Co., Ltd., Japan). Analysis of variance (ANOVA) followed by Tukey's multiple comparison test at $P < 0.05$ was used to determine the significance of differences between treatments.

3. Results

3.1. Plant growth

Fig. 1 shows the dry weights and distribution patterns of the dry weights of the shoots. The dry weight of the shoots increased dramatically from the R2 to the R6 growth stage and the weight of the grains at the R8 growth stage was 1.7–2.2 times higher than that at the R6 growth stage (Fig. 1a, Table S1 in supplementary material), with 42% of the total weight of the shoots represented by the grains at the R8 stage (Fig. 1b). K application did not affect the dry weight or distribution patterns of the dry weights of the shoots at any growth stage.

3.2. K and ^{137}Cs inventory and distribution in the shoots

Figs. 2 and 3 show the inventory and distribution patterns of K and ^{137}Cs in the shoots, respectively, while the corresponding numerical data are in Tables S2 and S3 in supplementary material, respectively. The inventory of K in the shoots increased greatly from the V5 to the R6 growth stage (Fig. 2a). From the R6 to the R8 growth stages, the inventory of K increased in the grains and decreased in all other parts of the shoot, regardless of the K application level. The decrease in the inventory of K was higher in the leaf blades and stems than in the petioles and pods.

The inventory of K in the shoots increased with K application, and was 1.46 times greater under K780 treatment than under K0 treatment (Fig. 2a). K application did not affect the distribution pattern of K in the shoots until the R2 growth stage (Fig. 3a); however, at the R6 and R8 growth stages, as the application level of K increased, the distribution of K tended to decrease in the grains and increase in all other parts of the shoot. At the R8 growth stage, 47%–56% of the total inventory of K in the shoots was distributed in the grains.

The inventory of ^{137}Cs in the shoots increased sharply from the R2 to the R6 growth stage (Fig. 2b). Compared with K, ^{137}Cs was distributed more in the leaf blades and less in the grains at the R6 and R8 growth stages, and the distribution percentage of ^{137}Cs to the grains was less than 38% of the total inventory (Fig. 3b). At all growth stages, the total inventory of ^{137}Cs in the shoots decreased with the increase in K application level (Fig. 2b). Although K application reduced the inventories of ^{137}Cs in each part of the shoot, the sensitivity to K application differed among parts. The rates of decrease in the inventories of ^{137}Cs at the R6 and R8 growth stages were highest in the grains and pods, followed by the petioles and stems, and were lowest in the leaf blades (Fig. 2b). Therefore, as the K application level increased, ^{137}Cs was distributed to the leaf blades rather than to the grains and pods (Fig. 3b).

3.3. K and ^{137}Cs concentrations in the grains

Table 2 shows the concentrations of K and ^{137}Cs in the grains. The K concentration in the grains was about 20 g kg^{-1} dry weight at the R6 and
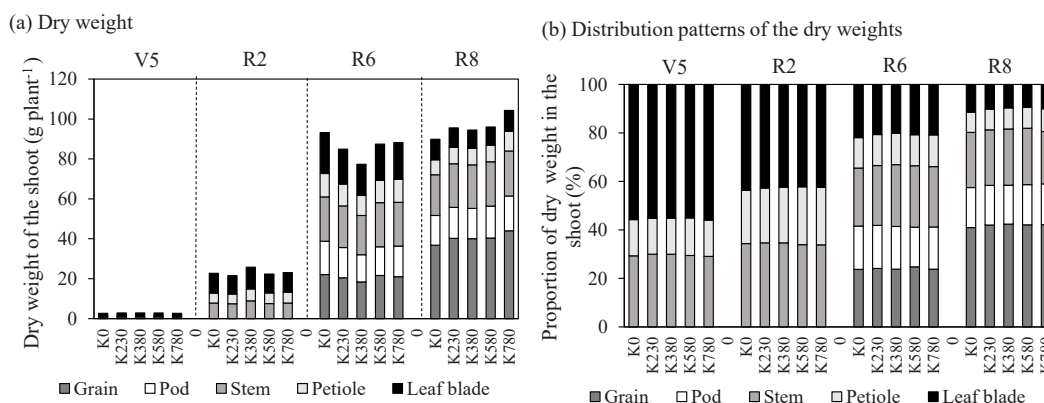

Fig. 1. Dry weights and distribution patterns of the dry weights of the shoots. (a) Dry weight. (b) Distribution patterns of the dry weights. V5, fifth trifoliate stage; R2, full bloom stage; R6, full seed stage; R8, full maturity stage. No significant differences in the dry weights of the shoot were observed between K treatment levels (Tukey's multiple comparison test at $P < 0.05$).
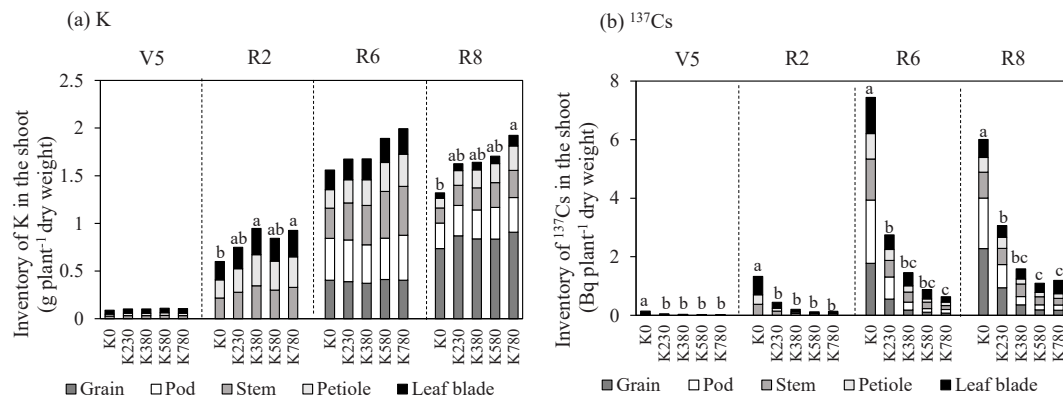
Fig. 2. Inventories of K and ^{137}Cs in the shoots. (a) K and (b) ^{137}Cs. V5, fifth trifoliate stage; R2, full bloom stage; R6, full seed stage; R8, full maturity stage. Different letters at the same growth stage indicate significant differences in the total inventories of K and ^{137}Cs in the shoots (Tukey's multiple comparison test at $P < 0.05$).
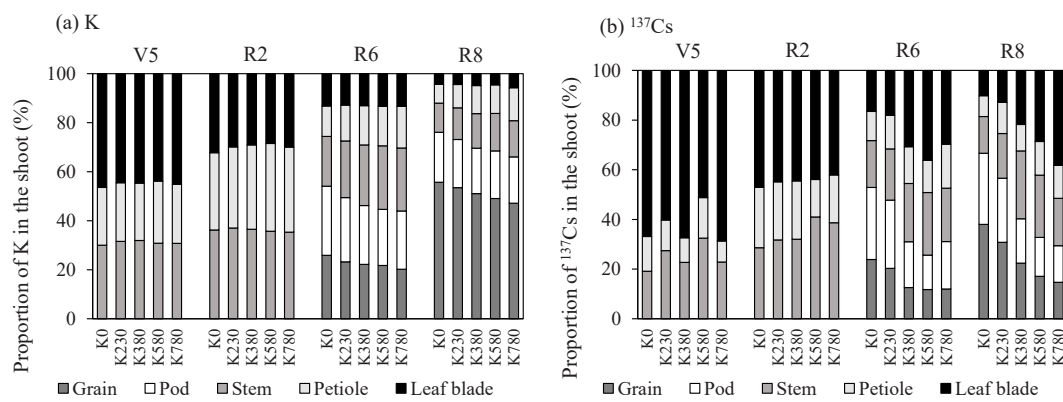

Fig. 3. Distribution patterns of K and ^{137}Cs in the shoots. (a) K and (b) ^{137}Cs. V5, fifth trifoliate stage; R2, full bloom stage; R6, full seed stage; R8, full maturity stage.

Table 2
Concentration of K and ^{137}Cs in the grains.

K treatment	K (g kg^{-1} dry weight)		^{137}Cs (Bq kg^{-1} dry weight)	
	R6	R8	R6	R8
K0	18.3	20.0	79.5 [a]	61.8 [a]
K230	19.0	21.6	27.3 [b]	23.7 [b]
K380	20.2	20.9	9.9 [bc]	8.7 [c]
K580	19.1	20.7	4.8 [c]	4.6 [c]
K780	19.2	20.6	3.6 [c]	4.0 [c]
ANOVA				
K treatment		ns		**
Growth stage		**		ns
K treatment × Growth stage		ns		ns

R6, full seed stage; R8, full maturity stage.
Different letters at the same growth stage indicate significant differences (Tukey's multiple comparison test at $P < 0.05$).
** and * show significant difference at $P < 0.01$ and 0.05, respectively. ns indicates not significant.

the R8 growth stages, regardless of the K application level. In contrast, the ^{137}Cs concentration in the grains at the R6 and R8 growth stages decreased with the increase in K application level, and the effect of K application on the ^{137}Cs concentration in the grain was unclear when the targeted soil-exchangeable K concentration before the cultivation was greater than 380 mg kg^{-1} of K$_2$O. The TF ranged from 0.0015 to 0.0237 at the R8 growth stage, and was dependent on the K application level (data not shown).

3.4. ^{137}Cs/K ratios in the plants

Table 3 shows the ^{137}Cs/K ratio of each plant part. The ratio was highest in the leaf blade throughout cultivation, and although values at the R6 growth stage were similar in all parts of the shoot except the leaf blade, the ratio of the grains tended to be lower than all other parts at the R8 growth stage. The ^{137}Cs/K ratio of the grains at the R8 growth stage was slightly lower than that at the R6 growth stage, whereas in all other parts of the shoot, the ratio increased. K application caused a reduction in the ^{137}Cs/K ratio in all parts of the shoot.

3.5. Exchangeable K and ^{137}Cs concentrations in the soil

Table 4 shows the concentrations of exchangeable K and ^{137}Cs in the soil, both of which can be absorbed by crops. As growth progressed, the exchangeable K concentration decreased except under K0 treatment, whereas exchangeable ^{137}Cs tended to increase. At all growth stages, higher K application levels increased the exchangeable K concentration and decreased the exchangeable ^{137}Cs concentration. Consequently, the exchangeable ^{137}Cs/K ratio tended to increase with growth and decrease with increasing K application level at all growth stages.

3.6. Relationship between the ^{137}Cs/K ratio in the shoots and exchangeable ^{137}Cs/K ratio in the soil

The relationship between the ^{137}Cs/K ratio in the shoots (Table 3) and exchangeable ^{137}Cs/K ratio in the soil (Table 4) is shown in Fig. 4. A positive correlation was observed between the ^{137}Cs/K ratio in the shoots and the exchangeable ^{137}Cs/K ratio in the soil for each growth stage ($r^2 > 0.98$), and the relationship was expressed as a linear

Table 3
The ^{137}Cs/K ratio in each plant part.

Growth stage	Plant part	^{137}Cs/K (Bq g^{-1} dry weight)													
		K0			K230			K380			K580			K780		
V5	Shoot	1.60	A		0.49	B		0.32	B		0.28	B		0.21	B	
	Leaf blade	2.29	A	a	0.67	B	a	0.48	B	a	0.32	B		0.32	B	a
	Petiole	0.94	A	b	0.26	B	c	0.13	BC	b	0.18	BC		0.07	C	b
	Stem	1.03	A	b	0.43	B	b	0.22	B	b	0.30	B		0.15	B	b
	Pod	–														
	Grain	–														
	ANOVA															
	K treatment							**								
	Plant parts							**								
	K treatment × Plant parts							**								
R2	Shoot	2.19	A		0.59	B		0.22	C		0.15	C		0.15	C	
	Leaf blade	3.20	A	a	0.89	B	a	0.34	BC	a	0.21	C		0.21	C	
	Petiole	1.68	A	b	0.41	B	b	0.15	B	b	0.06	B		0.08	B	
	Stem	1.73	A	b	0.51	B	b	0.19	B	ab	0.18	B		0.17	B	
	Pod	–														
	Grain	–														
	ANOVA															
	K treatment							**								
	Plant parts							**								
	K treatment × Plant parts							**								
R6	Shoot	4.74	A		1.67	B		0.87	C		0.47	C		0.32	C	
	Leaf blade	5.97	A	a	2.37	B		2.10	BC	a	1.30	BC	a	0.73	C	a
	Petiole	4.50	A	ab	1.55	B		0.81	BC	b	0.38	C	b	0.34	C	ab
	Stem	4.42	A	ab	1.48	B		0.83	BC	b	0.45	C	b	0.28	C	ab
	Pod	4.87	A	ab	1.74	B		0.66	C	b	0.28	C	b	0.26	C	ab
	Grain	4.36	A	b	1.45	B		0.49	C	b	0.25	C	b	0.19	C	b
	ANOVA															
	K treatment							**								
	Plant parts							**								
	K treatment × Plant parts							ns								
R8	Shoot	4.53	A		1.90	B		0.96	BC		0.65	C		0.62	C	
	Leaf blade	10.68	A	a	5.57	AB	a	4.14	B	a	4.28	B	a	4.45	B	a
	Petiole	4.91	A	b	2.45	B	b	0.91	BC	bc	0.76	C	b	0.62	C	b
	Stem	5.70	A	b	2.62	B	b	1.88	BC	b	1.05	BC	b	0.80	C	b
	Pod	6.38	A	b	2.50	B	b	0.92	C	bc	0.52	C	b	0.49	C	b
	Grain	3.09	A	b	1.10	B	b	0.42	C	c	0.22	C	b	0.19	C	b
	ANOVA															
	K treatment							**								
	Plant parts							**								
	K treatment × Plant parts							ns								

V5, fifth trifoliate stage; R2, full bloom stage; R6, full seed stage; R8, full maturity stage.

Different letters at the same growth stage indicate significant differences between K treatments (uppercase letters) and between plant parts (lowercase letters) (Tukey's multiple comparison test at $P < 0.05$).

** and * show significant differences at $P < 0.01$ and 0.05, respectively. ns indicates not significant. ^{137}Cs/K ratios in each part of the shoot were significantly different among growth stages (ANOVA at $P < 0.01$).

From the R6 to the R8 growth stages, the ^{137}Cs/K ratios of the leaf blades, petioles, stems, and pods increased, whereas those in the grains decreased (ANOVA at $P < 0.05$).

regression equation (Y and X representing the ^{137}Cs/K ratio in the shoots and exchangeable ^{137}Cs/K ratio in the soil, respectively) passing through the origin with a specific regression coefficient (slope). Moreover, the slope of the regression line for the shoots was almost twice as high at the R6 and R8 growth stages than the V5 and R2 growth stages.

4. Discussion

The dry weight of the shoots increased significantly from the R2 to the R6 growth stage (Fig. 1a). The inventories of K and ^{137}Cs in the shoots increased remarkably with this increase in the dry weight (Figs. 1a and 2 and). Harper (1971) reported that the maximum rate of nutrient uptake occurred during the R2 and R5 growth stages. Thus, nutrient accumulation in the shoots coincides closely with dry matter accumulation. The rapid increase in K demand after the R2 growth stage

(full bloom stage) reflects the overlap of vegetative growth (the production of leaves and stems) and reproductive growth (production of grains) (Fig. 1a). Both K and Cs belong to the same alkali metal group and are absorbed competitively (Shaw and Bell 1991), suggesting that Cs is absorbed by the K transport system (Zhu and Smolders 2000; White and Broadley 2000). Thus, as the absorption of K increased, the absorption of ^{137}Cs may also have increased.

The K concentration in the grains was relatively constant, regardless of the K application level (Table 2). In line with this, a previous study showed that the nutrient composition (K, Mg, Ca, Na, and P) and nutritional value (proteins, lipids, and total sugars) of soybean grains were not affected by K application (Hirayama et al., 2018), with a similar result also reported in paddy rice (Tsukada et al., 2002a). These results suggest that the dynamics of K in soybean is determined by stoichiometric homeostasis, that is, the ability of plants to maintain a

Table 4
Concentration of exchangeable K and ^{137}Cs in the soil.

K treatment	Exchangeable K (mg kg⁻¹ dry weight)				Exchangeable ^{137}Cs (Bq kg⁻¹ dry weight)				Exchangeable ^{137}Cs/K (Bq mg⁻¹ dry weight)			
	V5	R2	R6	R8	V5	R2	R6	R8	V5	R2	R6	R8
K0	86 d	72 e	73 d	86 d	81 a	85 a	90 a	96 a	0.96 a	1.21 a	1.24 a	1.16 a
K230	174 cd	145 d	116 cd	122 cd	57 b	64 b	61 ab	68 ab	0.35 b	0.44 b	0.53 b	0.58 ab
K380	251 bc	215 c	150 c	184 c	48 b	56 b	51 b	55 ab	0.18 b	0.25 b	0.34 b	0.30 b
K580	342 b	327 b	260 b	256 b	37 b	40 b	41 b	41 b	0.11 b	0.12 b	0.16 b	0.16 b
K780	564 a	521 a	334 a	328 a	43 b	43 b	41 b	46 b	0.08 b	0.08 b	0.12 b	0.14 b
ANOVA												
K treatment	**				**				**			
Growth stage	**				ns				ns			
K treatment × Growth stage	**				ns				ns			

V5, fifth trifoliate stage; R2, full bloom stage; R6, full seed stage; R8, full maturity stage.
Different letters at the same growth stage indicate significant differences (Tukey's multiple comparison test at $P < 0.05$).
** and * show significant difference at $P < 0.01$ and 0.05, respectively. ns indicates not significant.
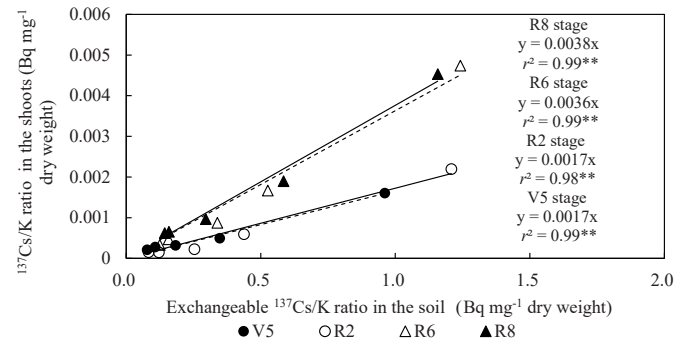

Fig. 4. Relationship between the ^{137}Cs/K ratio in the shoots and exchangeable ^{137}Cs/K ratio in the soil. V5, fifth trifoliate stage; R2, full bloom stage; R6, full seed stage; R8, full maturity stage. ** indicates $P < 0.01$. ANCOVA shows a significant difference of the regression line slopes before and after R2 growth stage ($P < 0.01$).

relatively stable element composition regardless of changes in nutrient availability. The K concentration of the soybean grains was approximately 20 g kg⁻¹ dry weight (Table 2), whereas that of paddy rice grain was about 2 g kg⁻¹ dry weight (Kato et al., 2015). Furthermore, 47%–56% of the total inventory of K in the soybean shoots was distributed to the grains (Fig. 3a), whereas in paddy rice, the distribution of K to the grains was less than 13% of the shoots (Ishikawa et al., 2018). This is probably because paddy rice grains accumulate K in the embryo, which is only a small part of the grains, whereas soybean grains accumulate K in the cotyledon, the largest part of the grain (Nihei et al., 2017). When grown under identical soil, the radiocesium concentration of the soybean grains is likely to be higher than that of paddy rice given that the localization of Cs and K in the soybean grains is similar (Nihei et al., 2017).

The effect of K application was much greater in terms of the ^{137}Cs dynamics than those of K or biomass production (Figs. 1–3). At all growth stages, the total inventory of ^{137}Cs in the shoots decreased with the increase in K application level (Fig. 2b), and no apparent effect of K application on ^{137}Cs accumulation in the shoots was observed when the targeted soil-exchangeable K concentration before the cultivation was higher than 380 mg kg⁻¹ of K$_2$O (Fig. 2b). Moreover, ^{137}Cs was distributed to the leaf blades rather than the grains as the application level of K increased (Fig. 3b). In paddy rice, the ratio of Cs in the brown rice to that in the straw was negatively correlated with the soil K level, suggesting that the proportion of Cs accumulation in the brown rice to that in the whole plant increased under low soil K conditions (Ishikawa et al., 2018). The present results suggest that K application decreased not only the accumulation of ^{137}Cs in the shoots but also the distribution of ^{137}Cs to the grains. However, the decrease of ^{137}Cs distribution to the grain had a much smaller effect than that of ^{137}Cs root absorption (Figs. 2b and 3b). Overall, the main factor affecting the reduction in ^{137}Cs accumulation in the grains was the reduction in ^{137}Cs absorption with K application.

At all growth stages, higher K application levels increased the exchangeable K concentration and decreased the exchangeable ^{137}Cs concentration (Table 4). This result suggests that the exchangeable ^{137}Cs concentration increases as the exchangeable K concentration in the soil decreases, which is line with previous findings (Kubo et al., 2015; Ishikawa et al., 2018). Cs in the soil is selectively fixed at frayed edge sites (FES) (Sawhney 1972; Cremers 1988), and when the exchangeable K concentration in the soil decreases, Cs fixed in the FES should be released (Gommers et al., 2005; Thiry et al., 2005). Here, the exchangeable ^{137}Cs/K ratio in the soil was positively correlated with the ^{137}Cs/K ratio in the shoots (Fig. 4), similar to results obtained in paddy rice (Kondo et al., 2015). These results also suggest that the exchangeable ^{137}Cs/K ratio in the soil is an important factor determining the ^{137}Cs/K ratio in shoots. The regression line slopes were similar at the V5

and R2 growth stages and the R6 and R8 growth stages (Fig. 4). In contrast, the regression line slopes at the R6 and R8 growth stages were much higher than those at theV5 and R2 growth stages. These results suggest that the discrimination of ^{137}Cs from K at the R6 and R8 growth stages was lower than that at the V5 and R2 growth stages, despite similar exchangeable ^{137}Cs/K ratios in the soil, which indicates that the absorption mechanisms of K and ^{137}Cs in soybean differ before and after the R2 growth stage. The absorption of Cs is mediated mainly by K transporters and channels, and the discrimination of Cs from K differs among these systems (White and Broadley 2000; Zhu and Smolders 2000; Broadley et al., 2001; Qi et al., 2008). High-affinity K transporters function in low-K conditions with low discrimination of Cs from K, whereas K channels, such as voltage-independent cation channels, function in high-K conditions with high discrimination (Zhu and Smolders 2000). Meanwhile, K starvation can also upregulate the expression of high-affinity K transporters, which may have reduced the discrimination of ^{137}Cs from K after the R2 growth stage. As a result of the decrease in discrimination of ^{137}Cs from K during absorption (Fig. 4) and the increase in soil-exchangeable ^{137}Cs/K (Table 4) with growth, ^{137}Cs became more transferable to the shoots after the R2 growth stage.

The chemical behavior of Cs is likely to be similar to that of K given that both are alkali elements with similar physicochemical properties (Shaw and Bell 1991; Smolders et al., 1997). However, they appear to behave differently in soil-plant systems. The exchangeable ^{137}Cs/K ratio in the soil was much higher than the ^{137}Cs/K ratio in the shoots at all growth stages (Fig. 4), and the ratio was not uniform in different parts of the shoot (Table 3). In soybean, ^{137}Cs was distributed more in the leaf blades and less in the grains compared with K, regardless of the K application level, and this tendency was promoted by K fertilization (Fig. 3). Accordingly, the ^{137}Cs/K ratio was highest in the leaf blades and tended to be lowest in the grains at the R8 growth stage, and the rates of decrease in the ^{137}Cs/K ratio with K application were larger in the grains than the leaf blades at the same stage (Table 3). K is a mobile nutrient, translocated from the older to younger parts and from the source (plant parts other than the grains: leaf blade, stem, and so on) to the sink (grains). Tsukada et al. (2002b) reported that, in paddy rice, the ^{133}Cs concentration at maturity was higher in older leaf blades, whereas the K concentration was higher in younger leaf blades. These results suggest that K is more efficiently absorbed from the soil and more translocatable during source-to-sink translocation than Cs. The differences in the ^{137}Cs/K ratio among different parts of the shoot are thought to be related to the discrimination of Cs from K.

Physiological processes that increase nutrient accumulation in the grains include direct transport (absorption) from the soil and remobilization from plant parts other than the grains. The increase in the inventory of K in the grains between the R6 and the R8 growth stages was similar to or slightly lower than the total decrease in K in all parts of the shoot except the grains, whereas the inventory of K decreased sharply in the leaf blades and stems (Fig. 2a). In addition, the concentration of K in these parts also decreased substantially during this period (data not shown). Thus, these parts are the primary source of K for soybean grains. Bender et al. (2015) suggested that the leaf provides the primary source of remobilized N and P, but the stem seems to serve as temporary storage for K in soybean. However, the source of ^{137}Cs in the grains could not be explained fully by translocation from the parts of the shoot other than the grains, except under K0 treatment. This was because the increase in the inventory of ^{137}Cs in the grains from the R6 to R8 growth stages was greater than the apparent total decrease in all other parts of the shoot (Fig. 2b). Therefore, it is thought that the increase in the inventory of ^{137}Cs in the grains from the R6 to R8 growth stages depends on whether the ^{137}Cs accumulated in the grains was newly absorbed from the soil or had already accumulated in the roots and was translocated. Given that the two were indistinguishable in this study due to the lack of root samples, further studies are needed to elucidate the dynamics of ^{137}Cs in the roots under minimal soil contamination.

It is difficult to remove all radioactivity from fields by decontamination. Furthermore, the decrease in radiocesium concentrations in the soil is slow because of the length of the ^{137}Cs half-life. Because there is still a high risk of an increased TF when the soil-exchangeable K concentration decreases substantially (NARO and Fukushima Prefecture 2019), continuous countermeasures against radiocesium transfer to crops are required. In soybean, the demand for K increased after the R2 growth stage (Fig. 2a) and the exchangeable ^{137}Cs/K ratio in the soil tended to increase with growth (Table 4). Moreover, the discrimination of ^{137}Cs from K was lower at the R6 and R8 growth stages than at the V5 and R2 growth stages under the same exchangeable ^{137}Cs/K ratio in the soil (Fig. 4). Therefore, lowering the soil-exchangeable radiocesium/potassium ratio after the R2 growth stage by increasing K availability may efficiently reduce the transfer of radiocesium from the soil to the grains. The optimal strategy for increasing K availability after the R2 growth stage in the field remains unclear, and thus further studies are needed.

5. Conclusions

We conducted a field experiment in soybean with different levels of K application to elucidate the comparative dynamics of ^{137}Cs and K. The inventory of K in the shoots increased greatly from the fifth trifoliate stage to the full seed stage, and as the absorption of K increased, so too did the absorption of ^{137}Cs. The effect of K application was much greater in terms of the dynamics of ^{137}Cs than those of K or biomass production. Although K application reduced not only the accumulation of ^{137}Cs in the shoots but also the distribution of ^{137}Cs to the grains, the ^{137}Cs distribution to the grain had a much smaller effect on ^{137}Cs accumulation in the grains than ^{137}Cs absorption. Moreover, the exchangeable ^{137}Cs/K ratio in the soil was positively correlated with the ^{137}Cs/K ratio in the shoots for each growth stage, and the slope of the regression line at the full seed and full maturity stage was almost twice that at the fifth trifoliate and full bloom stage. These results suggest that the discrimination of ^{137}Cs from K decreases after the full bloom stage. As a result of the decrease in the discrimination of ^{137}Cs from K during absorption and the increase in soil-exchangeable ^{137}Cs/K with growth, ^{137}Cs was more transferable to the shoots after the full bloom stage. Therefore, as a future strategy, lowering the soil-exchangeable radiocesium/potassium ratio after the full bloom stage by increasing potassium availability could efficiently reduce the transfer of radiocesium to the grains.

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.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvrad.2021.106609.

References

Bender, R.R., Haegele, J.W., Below, F.E., 2015. Nutrient uptake, partitioning, and remobilization in modern soybean varieties. Agron. J. 107, 563–573. https://doi.org/10.2134/agronj14.0435.

Broadley, M.R., Willey, N.J., 1997. Differences in root uptake of radiocaesium by 30 plant taxa. Environ. Pol. 97, 11–15. https://doi.org/10.1016/S0269-7491(97)00090-0.

Broadley, M.R., Escobar-Gutiérrez, A.J., Bowen, H.C., Willey, N.J., White, P.J., 2001. Influx and accumulation of Cs$^+$ by the *akt 1* mutant of *Arabidopsis thaliana* (L.) Heynh. lacking a dominant K$^+$ transport system. J. Exp. Bot. 52, 839–844. https://doi.org/10.1093/jexbot/52.357.839.

Cremers, A., 1988. Quantitative analysis of radiocaesium retention in soils. Nature 335, 247–249. https://doi.org/10.1038/335247a0.

Ehlken, S., Kirchner, G., 2002. Environmental processes affecting plant root uptake of radioactive trace elements and variability of transfer factor data: a review. J. Environ. Radioact. 58, 97–112. https://doi.org/10.1016/S0265-931X(01)00060-1.

Gommers, A., Thiry, Y., Delvax, B., 2005. Rhizospheric mobilization and plant uptake of radiocesium from weathered micas: I. Influence of Potassium Depletion. J. Environ. Qual. 34. https://doi.org/10.2134/jeq2004.0406.

Harper, J.E., 1971. Seasonal nutrient uptake and accumulation patterns in soybeans. Crop Sci. 11, 347–350. https://doi.org/10.2135/cropsci1971.0011183X001100030011x.

Hirayama, T., Takeuchi, T., Nakayama, H., Nihei, N., 2018. Effects of decreasing radiocesium transfer from the soil to soybean plants and changing the seed nutrient composition by the increased application of potassium fertilizer. Bulletin of the Fukushima Agricultural Technology Centre 9, 1–10 (in Japanese with English abstract).

Ishikawa, J., Fujimura, S., Kondo, M., Murai-Hatano, M., Goto, A., Shinano, T., 2018. Dynamic changes in the Cs distribution throughout rice plants during the ripening period, and effects of the soil-K level. Plant Soil 429, 503–518. https://doi.org/10.1007/s11104-018-3700-z.

Japanese Ministry of Agriculture, Forestry and Fisheries, Results of radioactive cesium concentration in agricultural products (in Japanese) (accessed 17 March 2021). http://www.maff.go.jp/j/kanbo/joho/saigai/s_chosa/index.html.

Japanese Ministry of Health, Labor, and Welfare, 2011. New standard limits for radionuclides in foods (accessed 17 March 2021). http://www.mhlw.go.jp/english/topics/2011eq/dl/food-120821_1.pdf.

Kato, N., Kihou, N., Fujimura, S., Ikeba, M., Miyazaki, N., Saito, Y., Egichi, 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. https://doi.org/10.1080/00380768.2014.995584.

Kobayashi, 2014. Countermeasure practices for radiocesium contamination control in upland crops. Jpn. J. Soil Sci. Plant Nutr. 85, 94–98. https://doi.org/10.20710/dojo.85.2_94. (In Japanese).

Kobayashi, T., Saito, S., Hara, Y., 2014. The absorption mechanism and the measure against absorption control of radiocesium to vegetables. Jpn. J. Soil Sci. Plant Nutr. 85, 99–102. https://doi.org/10.20710/dojo.85.2_99. (In Japanese).

Kondo, M., Makino, T., Eguchi, T., Goto, A., Nakano, H., Takai, T., Arai-Sanoh, Y., Kimura, T., 2015. Comparative analysis of the relationship between Cs and K in soil and plant parts toward control of Cs accumulation in rice. Soil Sci. Plant Nutr. 61, 144–151. https://doi.org/10.1080/00380768.2014.973348.

Kubo, K., Nemoto, K., Kobayashi, H., Kuriyama, Y., Harada, H., Matsunami, H., Eguchi, T., Kihou, N., Ota, T., Keitoku, S., Kimura, T., Shinano, T., 2015. Analyses and countermeasures for decreasing radioactive cesium in buckwheat in areas affected by the nuclear accident in 2011. Field Crop. Res. 170, 40–46. https://doi.org/10.1016/j.fcr.2014.10.001.

Kubo, K., Fujimura, S., Kobayashi, H., Ota, T., Shinano, T., 2017. Effect of soil exchangeable potassium content on cesium absorption and partitioning in

buckwheat grown in a radioactive cesium-contaminated field. Plant Prod. Sci. 20, 396–405. https://doi.org/10.1080/1343943X.2017.1355737.

MAFF, NARO, NIAES, 2014. Factors and Countermeasure of Buckwheat with High Radioactive Cesium Concentration. Investigation of Factors and Result of Trial Examination (Outline, the second ed.) (In Japanese) (Accessed 17 March 2021). http://www.maff.go.jp/j/kanbo/joho/saigai/pdf/h25soba_yoin.pdf.

MAFF, NARO, NIAES, 2015. Factors and Countermeasure of Soybean with High Radioactive Cesium Concentration. Investigation of Factors and Result of Trial Examination (Outline, the third ed.) (In Japanese) (Accessed 17 March 2021). http://www.maff.go.jp/j/kanbo/joho/saigai/pdf/youin_daizu_3.pdf.

NARO, Fukushima Prefecture, 2019. Cultivation guide to reduce radiocesium concentration in soybean (in Japanese) (accessed 17 March 2021). http://www.naro.affrc.go.jp/publicity_report/publication/pamphlet/tech-pamph/130461.html.

Nihei, N., Hamamoto, S., 2019. Absorption of radiocaesium in soybean. In: Nakanishi, T. M., O`Brien, M., Tanoi, K. (Eds.), Agricultural Implications of the Fukushima Nuclear Accident (III): after 7 Years. Springer Japan, pp. 27–33. https://doi.org/10.1007/978-981-13-3218-0_4.

Nihei, N., Sugiyama, A., Ito, Y., Onji, T., Kita, K., Hirose, A., Tanoi, K., Nakanishi, T.M., 2017. The concentration distribution of Cs in Soybean Seeds. Radioisotopes 66, 235–242. https://doi.org/10.3769/radioisotopes.66.235.

Qi, Z., Hampton, C.R., Shin, R., Barkla, B.J., White, P.J., Schachtman, D.P., 2008. The high affinity K$^+$ transporter AtHAK5 plays a physiological role in planta at very low K$^+$ concentrations and provides a caesium uptake pathway in Arabidopsis. J. Exp. Bot. 59, 595–607. https://doi.org/10.1093/jxb/erm330.

Sawhney, B.L., 1972. Selective sorption and fixation of cations by clay minerals: a review. Clay Clay Miner. 20, 93–100. https://doi.org/10.1346/CCMN.1972.0200208.

Shaw, G., Bell, J.N.B., 1991. Competitive effects pf potassium and ammonium on caesium uptake kinetics in wheat. J. Environ. Radioact. 13, 283–296. https://doi.org/10.1016/0265-931X(91)90002-W.

Smolders, E., Van den Brande, K., Merckx, R., 1997. Concentration of ^{137}Cs and K in soil solution predict the plant availability of ^{137}Cs in soils. Environ. Sci. Technol. 31, 3432–3438. https://doi.org/10.1021/es970113r.

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. https://doi.org/10.2134/jeq2004.0407.

Tsukada, H., Hasegawa, H., Hisamatsu, S., Yamasaki, S., 2002a. Transfer of ^{137}Cs and stable Cs from paddy soil to polished rice in Aomori, Japan. J. Environ. Radioact. 59, 351–363. https://doi.org/10.1016/S0265-931X(01)00083-2.

Tsukada, H., Hasegawa, H., Hisamatsu, S., Yamasaki, S., 2002b. Rice uptake and distributions of radioactive ^{137}Cs, stable ^{133}Cs and K from soil. Environ. Pollut. 117, 403–409. https://doi.org/10.1016/S0269-7491(01)00199-3.

Tsukada, H., Takeda, A., Hisamatsu, S., Inaba, J., 2008. Concentration and specific activity of fallout ^{137}Cs in extracted and particle-size fractions of cultivated soils. J. Environ. Raidoact. 99, 875–881. https://doi.org/10.1016/j.jenvrad.2007.11.014.

Tsumura, A., Komamura, M., Kobayashi, H., 1984. Behavior of radioactive Sr and Cs in soils and soil-plant systems. Bull. Natl. Inst. Agric. Sci. Ser. B (Soils Fert.) 36, 57–113 (In Japanese).

White, P.J., Broadley, M.R., 2000. Mechanisms of caesium uptake by plants. New Phytol. 147, 241–256.

Yamaguchi, N., Taniyama, I., Kimura, T., Yoshioka, K., Saito, M., 2016. Contamination of agricultural products and soils with radiocesium derived from the accident at TEPCO Fukushima Daiichi Nuclear Power Station: monitoring, case studies and countermeasures. Soil Sci. Plant Nutr. 62, 303–314. https://doi.org/10.1080/00380768.2016.1196119.

Zhu, Y.G., Smolders, E., 2000. Plant uptake of radiocaesium: a review of mechanisms, regulation and application. J. Exp. Bot. 51, 1635–1645. https://doi.org/10.1093/jexbot/51.351.1635.