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## ORIGINAL ARTICLE

## Radioactive Cs transfer to vegetables after the FDNPP accident

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### ABSTRACT

To evaluate the effectiveness of potassium (K) application in mitigating <sup>137</sup>Cs transfer from soil to plants, several vegetable species were cultivated under field and pot experiments. In the field experiment, squash, sweet potato, turnip, potato, and carrot were examined in 2020 and 2021 in two different areas of Hamadori (coastal region in Fukushima Prefecture). Transfer factor (TF) was calculated by dividing harvest radioactivity (Bq kg<sup>-1</sup> dry or fresh) to soil radioactivity (Bq kg<sup>-1</sup> dry) and was negatively correlated with the amount of exchangeable K (ExK) at harvest, regardless of the species, year, and location. In the pot experiment, edamame (immature soybean seed), spinach, turnip, and komatsuna were cultivated, and it was confirmed that ExK was the most powerful factor in regulating TF. Based on the relationship between ExK and TF for each vegetable species, the amount of ExK required to keep the <sup>137</sup>Cs concentration lower than a certain level (standard limitation value and one-quarter of that value) was calculated.

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### KEY WORDS

<sup>137</sup>Cs; Fukushima Prefecture; potassium application; vegetables

### 1. Introduction

After major nuclear accidents, radionuclides can contaminate agricultural land and products. Based on the inventory of released radionuclides and their half-lives, radiocesium (<sup>134</sup>Cs, <sup>137</sup>Cs) and <sup>90</sup>Sr have been considered the most problematic for soil contamination after the Chernobyl accident (e.g., Ivanov et al. 1997). In the case of the Tokyo Electric Power Company's Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident that occurred in 2011 in Japan, radionuclides with very short half-lives, such as <sup>133</sup>Xe (half-life is 5.25 days) (Stohl et al. 2012), <sup>131</sup>I (half-life 8.02 days) (Fesenko et al. 2020), and others (Povinec, Hirose, and Aoyama), were dispersed in the environment. Among them, <sup>131</sup>I dispersed into the air and got deposited on the surface of some leafy vegetables (United Nations 2014; Onda et al. 2015). After a few months, the dispersed <sup>131</sup>I in the environment was almost completely diminished owing to its short half-life, and further dispersion from the FDNPP did not occur. Furthermore, as the FDNPP accident occurred in March, most of the agricultural activity was not initiated, so direct attachment to the crops was not a major concern. However, during the initial days after the accident, dry deposition of <sup>131</sup>I occurred on some vegetables (Ohse et al. 2015), and secondary contamination took place by radiocesium derived from debris removal operations in a few cases (Matsunami et al. 2016). Therefore, the primary concern was food contamination due to the transfer of radiocesium from soil to plants (Hachinohe and Shinano 2020a, 2020b).

As early as 2011, most research focused on rice cultivation, and at first, the cultivation area was chosen based on the radiocesium activity of the soil (lower than 5 kBq kg<sup>-1</sup> on a dry weight basis). It was based on the observation of the transfer factor (TF: radioactivity of brown rice to radioactivity of soil) after the global fallout

in the 1960s (Shinano 2021). Unfortunately, the estimated TF was not reliable enough to predict brown rice radioactivity, and the exchangeable potassium (ExK) level was found to be critical for mitigating radiocesium transfer from soil to brown rice (Fujimura et al. 2014; Kato et al. 2015). Since 2012, the application of a large amount of potassium fertilizer has been practised to decrease the radioactivity of brown rice in contaminated areas. This countermeasure has also been applied for soybean (Hirayama et al. 2018; Hatano, Shinjo, and Takata 2021; Matsunami et al. 2021), buckwheat (Kubo et al. 2015), and pastures (Yamamoto et al. 2014; MAFF (Ministry of Agriculture, Forestry and Fisheries) 2020). On the other hand, very limited work has been conducted on vegetables (Kobayashi, Saito, and Hara 2014; Tagami et al. 2020), and most studies on vegetables lack information on how plants respond to soil ExK, which is well known as a critical determinant factor to regulate Cs transfer from soil to plants.

In the area where a substantial amount of radiocesium was dispersed by the FDNPP accident (mainly the southern part of Tohoku and the northern part of Kanto, Japan), in addition to major crops (e.g., rice, soybean, and buckwheat), leafy vegetables are also important crops, especially to farmers who produce vegetables mainly for domestic consumption, and some of the products are sold in a small lot. However, research on vegetables is limited because of their high water content at harvest, which decreases the radiocesium concentration of the product in fresh weight base (water content is more than 90% in leafy vegetables, e.g., Popkin, D'Anci, and Rosenberg 2010) and because they are generally grown with high amounts of fertilizers, both of which decrease the transfer of radiocesium from soil to plants. In 2020, reports on the limited data on vegetables published after the

**Table 1.** Growth condition of a field experiment (2020 and 2021).

| Year | Vegetables   | Variety      | Sowing date | Harvesting date | Nitrogen (kg N ha <sup>-1</sup> ) | Phosphorus (kg P <sub>2</sub> O <sub>5</sub> ha <sup>-1</sup> ) | Hill spacing (cm × cm) |
|------|--------------|--------------|-------------|-----------------|-----------------------------------|---|------------------------|
| 2020 | Squash       | Kokonoeguri  | 28 May      | 11 Aug          | 150                               | 150   | 40 × 200               |
|      | Sweet potato | Beniazuma    | 1 Jun       | 26 Oct          | 150                               | 150   | 30 × 200               |
|      | Turnip       | Natsuhakurei | 28 May      | 21 Jul          | 150                               | 150   | 15 × 200               |
| 2021 | Squash       | Kokonoeguri  | 19 May      | 2 Aug           | 150                               | 150   | 40 × 180               |
|      | Sweet potato | Beniazuma    | 26 May      | 5 Oct           | 30                                | 100   | 30 × 120               |
|      | Komatsuna    | Rakuten      | 6 May       | 7 Jun           | 150                               | 150   | 10 × 120               |
|      | Potato       | Kitaakari    | 31 Mar      | 1 Jul           | 150                               | 150   | 30 × 120               |
|      | Carrot       | Koyo 2go     | 16 Jul      | 8 Nov           | 150                               | 150   | 15 × 120               |

FDNPP were summarized in TECDOC from International Atomic Energy Agency (IAEA) (Tagami et al. 2020). The transfer of radionuclides from soil to plants is widely designated as TF, which is the ratio of the concentration of radionuclides in plants (any distinctive part of plants) to that in the soil. However, TF itself cannot be applied directly to the management of soil; the relationship between potassium availability in the soil and TF is highly regulated, and information on potassium availability (e.g., ExK) is essential. Furthermore, it should be mentioned that after fields were decontaminated, ExK tended to be decreased because of the low fertile non-contaminated soil addition (Kurokawa et al. 2019), so the importance of potassium fertilization becomes high especially under these decontaminated areas. As different levels of potassium are critical for regulating radiocesium uptake, field and pot experiments were conducted to demonstrate the effect of ExK levels at different sites and in different years. Finally, we demonstrate that a critical level of potassium in the soil regulates the radioactivity of several vegetables.

## 2. Materials and methods

### 2.1. Condition of fields before experiment

In 2015, the field was decontaminated by topsoil stripping and a non-contaminated soil dressing. Before 2011, the field was used as a paddy field, and after the FDNPP accident, the field was not used until 2019. Soil samples collected from five different points were analyzed separately after air-drying and passing through a 2-mm sieve. ExK was extracted by shaking for 1 h with 1 M ammonium acetate at a soil:solution ratio of 1:20. Soil chemical properties, including ExK, were determined using Eurofin Japan (Kanagawa, Japan). The data are presented in Table S1 in the Supplementary Material. The <sup>137</sup>Cs concentration was approximately 2.9 kBq kg<sup>-1</sup> dry soil, and no significant difference was observed between different crop fields. However, the heterogeneity of the soil <sup>137</sup>Cs is well known even after the initiation of agriculture after the accident (e.g., Editorial Boards of Methods for Soil Environment Analysis 1997b), and it could not be ignored in this study too. The difference was obvious in the ExK concentration, indicating that the decontamination process was not always constant.

### 2.2. Experiment 1 (Field experiment)

In 2020, squash (*Cucurbita moschata* Duch.), sweet potato (*Ipomoea batatas* L.), and turnip (*Brassica campestris* L. var. *glabra*) were planted in a field located in the northern

Hamadori area of Fukushima Prefecture, designated as Field A. The area of the field was 50 m × 20 m. The field was divided equally into five plots for different K fertilizer application (10 m × 20 m). Five hills were set in each plot, then each hill was divided into three sections as replications. Turnip was grown in one hill. Squash and sweet potato were grown separately in two hills. The soil taken from this field was designated Soil A and the type was originally lowland paddy soil. Planting and harvesting dates are listed in Table 1.

Nitrogen, phosphorus, and potassium fertilizers were applied before cultivation (Table 1). Five different levels of potassium were used to alter the ExK levels in the field. K<sub>2</sub>SO<sub>4</sub> fertilizer was applied at the rates of 0, 150 (conventional level), 300, 500, and 1000 kg K<sub>2</sub>O ha<sup>-1</sup>, designated as K0, K150, K300, K500, and K1000, respectively, for all vegetable crops, except for sweet potato. For sweet potato, K<sub>2</sub>SO<sub>4</sub> fertilizer was applied at rates of 0, 100, 200, 500, and 1000 K<sub>2</sub>O ha<sup>-1</sup>, designated as K0, K100, K200, K500, and K1000, respectively. Plants were cultivated using conventional farming protocols and four to eight plants were sampled per replication at harvest, with three replications. Tubers, leaves, and fruits were collected from sweet potato, komatsuna, and squash, respectively. In the case of turnip, leaves and roots were harvested (the leaves of turnip are used as leafy vegetables). After measuring the fresh weight, the samples were dried at 80°C for 48–72 h in an oven (VTRL-1000-2 T, Isuzu Seisakusyo, Niigata, Japan). The dry sample was ground for further analysis after weighing.

Soil samples were collected with three replicates after the cultivation of each soil with different K treatments. For the measurement of soil radiocesium concentration and ExK, about 100 g of soil sample was taken from beneath each sampled plant (4–8 samples in 2020 and 2–12 samples in 2021 depending on the size of the plant) to 0–15 cm depth and pooled for each replication. Exchangeable K was extracted by 1 M ammonium acetate and analyzed using atomic absorption spectrometry (Shimadzu SPCA-6210).

In 2021, a field in the middle part of the Hamadori area was used for squash, sweet potato, komatsuna (Japanese mustard spinach) (*Brassica rapa* L. var. *perviridis*), potato (*Solanum tuberosum* L.), and carrot (*Daucus carrot* subs. *sativus*) and the field was designated Field B, and was originally gray lowland soil and the soil taken from this field was designated Soil B. Soil B was also decontaminated in the same way in Soil A in 2015. The area of the field was 50 m × 22 m. The field was equally divided into five plots for different K fertilizer application (10 m × 22 m). Fifteen hills were set in each plot, and three hills were used for one species. Each line was used for the sampling of one replication.

Nitrogen, phosphorus, and potassium fertilizers were applied before cultivation, as shown in Table 1. Four different levels of potassium fertilizers were used to alter the ExK level in the field.  $K_2SO_4$  fertilizer was applied at rates of 0, 150, 300, and 500 kg  $K_2O$  ha<sup>-1</sup>, designated as K0, K150, K300, and K500, respectively, for all vegetables except for sweet potato. For sweet potato,  $K_2SO_4$  fertilizer was applied at rates of 0, 100, 200, and 400  $K_2O$  ha<sup>-1</sup>, designated as K0, K100, K200, and K400, respectively. Plants were cultivated using conventional farming protocols and four to eight plants were sampled per replication at harvest, with three replications. Tubers, leaves, and fruits were collected from sweet potato, and squash, respectively. In the case of turnip, leaves and roots were harvested (the leaves of turnip are used as leafy vegetables). Plant and soil sampling and subsequent procedures are same as in 2020.

All crop varieties and their growth conditions are shown in Table 1.

### 2.3. Experiment 2 (Pot experiment)

Plants were grown until harvest. Planting and harvesting dates are listed in Table 2. A pot experiment was conducted using soybean (*Glycine max* (L) Merr.) as edamame (immature soybeans with pods), spinach (*Spinacia oleracea* L.), and turnip for Soil C (obtained from a decontaminated field in the middle of the Hamadori area; gray lowland soil) in a greenhouse at the Fukushima Agricultural Technology Centre (FATC) (Koriyama, Fukushima). The pot size was 1/2000 a using Wagner pot (AS ONE, Osaka, Japan), containing 12 kg of fresh soil in 2020 and 10.3 kg of fresh soil in 2021, and three levels of K treatments were used. In pot experiments, nitrogen was applied at 0.22 g urea kg<sup>-1</sup> soil for all crops, except for soybean; in the case of soybean, 0.075 g urea kg<sup>-1</sup> soil was applied. Phosphorus was applied at 0.47 g superphosphate kg<sup>-1</sup> soil except for turnip; in the case of turnip, 0.67 g superphosphate kg<sup>-1</sup> soil was applied. For potassium treatment, potassium sulfate was applied at 0, 0.1, and 0.2 g  $K_2O$  kg<sup>-1</sup> soil. These application rates were equivalent to 0, 75, and 150 kg  $K_2O$  ha<sup>-1</sup> in 2020, designated as K0, K75, and K150, respectively. The treatment was conducted in triplicate.

Three experiments were conducted in 2021 by using edamame, spinach, and turnip. One was to observe the effects of repeated cultivation using the same soil used in 2020. K0, K150, and K300 pots were prepared using K0, K75, and K150 soils, respectively, after the experiment in 2020. K500 pots were prepared in 2021 using K150 soils in 2020. The K level was determined after the additional application of  $K_2SO_4$  at concentrations of 0, 150, 300, and 500 kg  $K_2O$  ha<sup>-1</sup>. The initial soil

<sup>137</sup>Cs radioactivity (Bq kg<sup>-1</sup> DW ± standard deviation) in Soil C was 2854 ± 135 in 2020, and 2665 ± 203 in 2021. Soil radioactivity was determined in each pot prepared for the experiment with three replicates.

The other experiment was to see the effect of repeated cultivation of Komatsuna in a year by using soil obtained from a field located in the northern part of the Nakadori area (Field D) as Soil D. The soil was classified as Andosol. No decontamination was conducted on Soil D before the experiment. Komatsuna was planted between 6 May and 7 June as Komatsuna 1st, and second cultivation was carried out between 2 July and 28 July as Komatsuna 2nd. After the first cultivation, soil was collected in each treatment and mixed thoroughly then prepared for the second cultivation. As the uptake of nutrient by Komatsuna cultivation was not expected to be large, no fertilization was performed before second cultivation. The soil type was humus gray soil. The initial <sup>137</sup>Cs radioactivity (Bq kg<sup>-1</sup> DW ± standard deviation) in Soil D was 1573 ± 124.

Using the same Soil D, a third experiment was carried out to compare the difference between Soil C and Soil D. For this purpose, turnip was cultivated in Soil D. Soil radioactivity was determined in each pot prepared for the experiment with three replicates.

Soil chemical properties including ExK were determined by FATC. The data are presented in Table S2 in the Supplementary Material. Soil ExK was extracted using 1 M ammonium acetate, and K concentration was determined by atomic absorption spectrometry (280FS AA, Agilent Technology, Santa Clara, CA, USA).

### 2.4. Gamma-ray spectrometry

The radiocesium concentration was determined by FATC, and only <sup>137</sup>Cs concentration was used in this study because the <sup>134</sup>Cs concentration of the plant sample was too low to determine the precise value. The <sup>137</sup>Cs concentration was decay-corrected to each sampling time. The <sup>137</sup>Cs concentration of the soil in the field experiments was determined using a Ge semiconductor detector (GCD-40190, BSI Instruments), and efficiency calibration was performed using a mixed nuclide solution for <sup>241</sup>Am and <sup>152</sup>Eu (Nuclear Technology Service, Inc., NIST (ANSI 42.22-195)) and a separate solution for <sup>60</sup>Co (Japan Radioisotope Association (JRIA), CO401). The <sup>137</sup>Cs concentration of the soil in the pot experiments was determined using an NaI detector (FNF-401, Ohyo Koken Kogyo Co. Ltd., Saitama, Japan), and efficiency calibration was performed using a separate solution for <sup>137</sup>Cs (JRIA, CS031). The <sup>137</sup>Cs

Table 2. Growth condition of pot experiments (2020 and 2021)

| Year | Soil   | Species       | Variety       | Sowing date | Harvesting date | Nitrogen (kg N ha <sup>-1</sup> ) | Phosphorus (kg P ha <sup>-1</sup> ) | Plants pot <sup>-1</sup> |
|------|--------|---------------|---------------|-------------|-----------------|-----------------------------------|-------------------------------------|--------------------------|
| 2020 | Soil C | Edamame       | Yuagarimusume | 04 Jun      | 11 Aug          | 50                                | 65                                  | 2                        |
|      |        | Spinach       | Mirage        | 08 Sep      | 05 Nov          | 150                               | 65                                  | 5                        |
|      |        | Turnip        | CR Mochibana  | 08 Sep      | 05 Nov          | 150                               | 65                                  | 3                        |
| 2021 | Soil C | Edamame       | Yuagarimusume | 24 May      | 28 Jul          | 50                                | 65                                  | 1                        |
|      |        | Spinach       | Mirage        | 30 Aug      | 08 Nov          | 150                               | 65                                  | 5                        |
|      |        | Turnip        | CR Mochibana  | 30 Aug      | 28 Oct          | 150                               | 65                                  | 3                        |
|      | Soil D | Komatsuna 1st | Rakuten       | 06 May      | 07 Jun          | 150                               | 65                                  | 4                        |
|      |        | Komatsuna 2nd | Rakuten       | 02 Jul      | 28 Jul          | 150                               | 65                                  | 4                        |
|      |        | Turnip        | CR Mochibana  | 30 Aug      | 27 Oct          | 150                               | 65                                  | 3                        |

concentrations of the plants in both the field and pot experiments were determined using a Ge semiconductor detectors (GC4020-7500SL-2002CSL, GC3520-7500SL-2002CSL, GC4020-7500SL-iPA-SL, GC3020-7500SL-2002CSL (Canberra Ltd., Boston, MA, USA)). Efficiency calibration was performed using a mixed nuclide solution (JRIA, MX033U8PP) and a separate solution for  $^{60}\text{Co}$  (JRIA, CO401).

The counting time was set to ensure that the counting error for each sample was below 10% for soils and 15% for plant samples. Each sample was placed in a cylindrical polypropylene container (U-8 container, 65 mm in height and 50 mm in diameter, RIG, Japan).

## 2.5. Statistical analysis

All statistical analyses were conducted using a statistical software (JMP Pro 16.1.0; USA). Multiple comparisons were performed using the Tukey–Kramer method. TF was converted to reciprocal values, and the relationship between ExK was calculated. Analysis of covariance (ANCOVA) was conducted to determine the difference in years, repetitions, and soil types to evaluate the difference in the relationship between ExK and TF on pot experiments.

## 3. Results

A summary of the experimental design used in this research is shown in Table 3.

### 3.1. Field experiment

For the field experiment, two different sites located in the Hamadori area of Fukushima were selected. One was located in the northern part of Hamadori (Field A) and the other was in the middle part of Hamadori (Field B). Both sites were decontaminated by topsoil stripping, and non-contaminated mountainous soil was dressed in 2015 and 2016, respectively. Weeds were collected, and after the FDNPP accident, no agricultural activities were conducted until the experiment. The soil mineral content was generally higher in Soil A than in Soil B, except for potassium (Table S1). The carbon and nitrogen contents were higher in Soil A, and it was speculated that the origin of the

dressed soil and the original soil characteristics caused these changes, although the details are not known.

Harvest weight, soil ExK, and  $^{137}\text{Cs}$  in the soil and plants after harvest are shown in Table 4. The growth of plant was better in Soil B than in Soil A, though soil physicochemical properties did not simply support the yield data (Supplementary Table 1); however, as the hill spacing and climate condition were not same, it is not concluded that the productivity between these soils is different. In 2020, a field experiment was conducted in the northern part of the Hamadori area (Field A, Soil A). Although there was no significant difference in the productivity of the examined crop species according to K levels. There was a clear tendency for ExK to increase with the application level of K even at harvest time, that is 213, 205, and 213 mg K kg<sup>-1</sup> at K0 treatment, and increased to 542, 340, and 520 mg K kg<sup>-1</sup> at K1000 treatment in squash, sweet potato, and turnip respectively. At the harvest,  $^{137}\text{Cs}$  content conversely decreased with increasing ExK (in case of sweet potato, there was no significant difference), that is 104, 83, 94, and 91 Bq kg<sup>-1</sup> dry at K0 treatment, and decreased to 13, 24, 24, and 47 Bq kg<sup>-1</sup> in squash, sweet potato, turnip root, and turnip leaf, respectively (Table 4). Although a significant difference of TF among the K treatments was not observed, there was a decreasing tendency with the higher K level (Table 4).

In 2021, although the field changed from Field A to Field B, the effect of K application level on the yield was not significant, except for potato where the fresh weight increased with the increase of K application, that is 2863 g m<sup>-2</sup> at K0 treatment to 4733 g m<sup>-2</sup> at K500 treatment (Table 4). ExK increased significantly with increasing K application level even at harvest time, that is 84, 59, 95, 56, and 89 mg K kg<sup>-1</sup> at K0 treatment and increased to 196, 132, 316, 208, and 124 mg K kg<sup>-1</sup> at K500 treatment in squash, sweet potato, komatsuna, potato, and carrot, respectively. And the harvest  $^{137}\text{Cs}$  content conversely decreased with increasing ExK, that is 165, 101, 788, 255, and 203 Bq kg<sup>-1</sup> dry at K0 treatment, and decreased to 29, 19, 51, 28, and 19 at K500 treatment (Table 4). TF was high in K0 treatment except for carrot, in carrot the highest average TF was observed in K10 treatment (Table 4). The lowest value of TF was observed in the highest K treatment regardless of crop species.

Although  $^{137}\text{Cs}$  in the soil was not same within a field of soil A or B, it is considered that the contamination by the  $^{137}\text{Cs}$  by the nuclear accident did not equally occur to the field. Furthermore,

Table 3. Experimental designs used in this study.

| Table 1. Experimental designs used in this study. |                 |                 |        |      |        |
|---|-----------------|-----------------|--------|------|--------|
| Species   | Culture         |                 |        |      |        |
|   | Field           |                 | Pot    |      |        |
|   | Field A<br>2020 | Field B<br>2021 | Soil C |      | Soil D |
|   |                 |                 | 2020   | 2021 | 2021   |
| Squash  | ⊙               | ⊙               |        |      |        |
| Sweet potato                                      | ⊙               | ⊙               |        |      |        |
| Turnip root                                       | ⊙               |                 | ○      | ○    | ○      |
| Turnip leaf                                       | ⊙               |                 |        | ○    | ○      |
| Komatsuna   |                 | ⊙               |        |      | ○○     |
| Potato  |                 | ⊙               |        |      |        |
| Carrot  |                 | ⊙               |        |      |        |
| Edamame   |                 |                 | ○      | ⊙    |        |
| Spinach   |                 |                 | ○      | ⊙    |        |

Double circle indicates that the obtained relationship was used to estimate the required K level for the estimation of radioactivity of harvest in Table 5.



**Table 4.** Growth of harvest and soil characteristics after the experiment (field experiment).

| Year | Field  | Vegetables    | Treatment | Number of samples | Yield (g m <sup>-2</sup> ) |              | ExK<br>mg K kg <sup>-1</sup> | Soil <sup>137</sup> Cs<br>kBq kg <sup>-1</sup> dry | Plant <sup>137</sup> Cs   |                         |
|------|--------|---------------|-----------|-------------------|----------------------------|--------------|------------------------------|--|---------------------------|-------------------------|
|      |        |               |           |                   | Fresh                      | Dry          |                              |  | Bq kg <sup>-1</sup> fresh | Bq kg <sup>-1</sup> dry |
| 2020 | Soil A | Squash        | K0        | 3                 | 136 ± 25 a                 | 32 ± 4 a     | 213 ± 13 b                   | 2.56 ± 0.42 a                                      | 24.1 ± 6.6 a              | 104 ± 36 a              |
|      |        |               | K150      | 3                 | 134 ± 27 a                 | 28 ± 6 a     | 241 ± 30 b                   | 2.48 ± 0.54 a                                      | 17.5 ± 12 ab              | 84 ± 59 a               |
|      |        |               | K300      | 3                 | 134 ± 15 a                 | 29 ± 6 a     | 271 ± 83 b                   | 2.44 ± 0.51 a                                      | 6.9 ± 3.2 ab              | 34 ± 20 a               |
|      |        |               | K500      | 3                 | 142 ± 34 a                 | 33 ± 14 a    | 205 ± 21 b                   | 2.20 ± 0.23 a                                      | 20.7 ± 11 ab              | 88 ± 33 a               |
|      |        |               | K1000     | 3                 | 149 ± 52 a                 | 35 ± 9 a     | 542 ± 158 a                  | 3.02 ± 1.5 a                                       | 2.8 ± 1.1 b               | 13 ± 6 a                |
|      |        | Sweet potato  | K0        | 3                 | 414 ± 131 a                | 120 ± 42 a   | 205 ± 67 a                   | 2.28 ± 1.1 a                                       | 23.9 ± 5.4 a              | 83 ± 16 a               |
|      |        |               | K100      | 3                 | 972 ± 440 a                | 275 ± 115 a  | 208 ± 46 a                   | 2.29 ± 0.20 a                                      | 28.6 ± 12.4 a             | 100 ± 46 a              |
|      |        |               | K200      | 3                 | 1056 ± 1096 a              | 281 ± 292 a  | 191 ± 54 a                   | 2.85 ± 0.28 a                                      | 14.6 ± 13.6 a             | 92 ± 49 a               |
|      |        |               | K500      | 3                 | 2722 ± 858 a               | 776 ± 265 a  | 205 ± 34 a                   | 1.85 ± 0.34 a                                      | 14.1 ± 4.5 a              | 49 ± 15 a               |
|      |        |               | K1000     | 3                 | 1989 ± 1549 a              | 522 ± 414 a  | 340 ± 174 a                  | 2.34 ± 0.35 a                                      | 6.4 ± 4.6 a               | 24 ± 17 a               |
|      |        | Turnip (Root) | K0        | 3                 | 307 ± 131 a                | 16 ± 7.2 a   | 213 ± 99 b                   | 1.80 ± 0.56 a                                      | 4.5 ± 4.0 a               | 94 ± 93 a               |
|      |        |               | K150      | 3                 | 157 ± 61 a                 | 7.2 ± 2.0 a  | 161 ± 31 b                   | 2.25 ± 1.1 a                                       | 3.9 ± 1.1 a               | 81 ± 22 a               |
|      |        |               | K300      | 3                 | 163 ± 102 a                | 7.4 ± 5.4 a  | 202 ± 75 b                   | 1.90 ± 0.59 a                                      | 2.3 ± 1.3 a               | 53 ± 36 a               |
|      |        |               | K500      | 2                 | 183 ± 76 a                 | 10 ± 4.7 a   | 315 ± 82 ab                  | 2.89 ± 0.54 a                                      | 1.4 ± 0.3 a               | 26 ± 7 a                |
|      |        |               | K1000     | 2                 | 262 ± 105 a                | 8.1 ± 1.6 a  | 520 ± 85 a                   | 2.69 ± 0.72 a                                      | 0.8 ± 0.4 a               | 24 ± 3 a                |
|      |        | Turnip (Leaf) | K0        | 3                 | 71 ± 65 a                  | 7.1 ± 5.7 a  | 213 ± 99 b                   | 1.80 ± 0.56 a                                      | 10.5 ± 7.7 a              | 91 ± 49 a               |
|      |        |               | K150      | 3                 | 18 ± 23 a                  | 2.1 ± 2.5 a  | 161 ± 31 b                   | 2.25 ± 1.1 a                                       | 21.6 ± 22 a               | 114 ± 26 a              |
|      |        |               | K300      | 3                 | 14 ± 11 a                  | 1.4 ± 1.6 a  | 202 ± 75 b                   | 1.90 ± 0.59 a                                      | 8.2 ± 7.4 a               | 91 ± 77 a               |
|      |        |               | K500      | 2                 | 36 ± 26 a                  | 4.1 ± 2.9 a  | 315 ± 82 ab                  | 2.89 ± 0.54 a                                      | 6.1 ± 3.7 a               | 53 ± 31 a               |
|      |        |               | K1000     | 2                 | 61 ± 78 a                  | 6.1 ± 4.2 a  | 520 ± 85 a                   | 2.69 ± 0.72 a                                      | 12.0 ± 11 a               | 47 ± 18 a               |
|      |        | Squash        | K0        | 5                 | 1583 ± 1083 a              | 369 ± 231 a  | 84 ± 1.5 c                   | 1.96 ± 1.2 a                                       | 39.9 ± 14.8 a             | 165 ± 55 a              |
|      |        |               | K150      | 3                 | 1725 ± 936 a               | 409 ± 230 a  | 104 ± 25 bc                  | 2.68 ± 0.99 a                                      | 44.1 ± 14 a               | 188 ± 64 a              |
|      |        |               | K300      | 3                 | 1327 ± 339 a               | 262 ± 139 a  | 139 ± 36 ab                  | 3.11 ± 1.1 a                                       | 27.4 ± 5.9 ab             | 149 ± 41 ab             |
|      |        |               | K500      | 3                 | 1264 ± 80 a                | 276 ± 34 a   | 196 ± 31 a                   | 0.77 ± 0.04 a                                      | 6.2 ± 0.2 b               | 29 ± 4 b                |
|      |        |               | K100      | 5                 | 1720 ± 1319 a              | 635 ± 492 a  | 59 ± 10 c                    | 2.64 ± 0.63 a                                      | 37.0 ± 6.0 a              | 101 ± 18 a              |
|      |        | Sweet potato  | K100      | 3                 | 2257 ± 618 a               | 804 ± 184 a  | 75 ± 13 bc                   | 2.97 ± 0.76 a                                      | 29.3 ± 8.3 ab             | 81 ± 23 ab              |
|      |        |               | K200      | 3                 | 3474 ± 853 a               | 1196 ± 282 a | 96 ± 28 ab                   | 2.47 ± 0.43 a                                      | 40.3 ± 22 a               | 118 ± 67 a              |
|      |        |               | K400      | 3                 | 1905 ± 963 a               | 651 ± 313 a  | 132 ± 8 a                    | 0.90 ± 0.20 b                                      | 6.7 ± 0.6 b               | 19 ± 2 b                |
|      |        | Komatsuna     | K0        | 5                 | 2063 ± 401 a               | 107 ± 16 a   | 95 ± 37 c                    | 2.76 ± 0.81 a                                      | 40.8 ± 6.9 a              | 788 ± 182 a             |
|      |        |               | K150      | 3                 | 2019 ± 455 a               | 97 ± 19 a    | 161 ± 32 bc                  | 3.03 ± 1.0 a                                       | 12.8 ± 5.0 b              | 262 ± 94 b              |
|      |        |               | K300      | 3                 | 1972 ± 159 a               | 97 ± 10 a    | 233 ± 85 ab                  | 2.90 ± 0.36 a                                      | 9.9 ± 1.7 b               | 202 ± 33 b              |
|      |        |               | K500      | 3                 | 1827 ± 272 a               | 103 ± 15 a   | 316 ± 22 a                   | 0.85 ± 0.12 b                                      | 2.8 ± 0.4 b               | 51 ± 8 b                |
|      |        | Potato        | K0        | 5                 | 2863 ± 574 b               | 719 ± 145 a  | 56 ± 12 c                    | 2.07 ± 0.36 ab                                     | 64.1 ± 9.7 a              | 255 ± 39 a              |
|      |        |               | K150      | 3                 | 3234 ± 985 ab              | 783 ± 234 a  | 103 ± 15 b                   | 2.52 ± 0.69 ab                                     | 31.8 ± 6.2 b              | 131 ± 27 b              |
|      |        |               | K300      | 3                 | 3371 ± 700 ab              | 774 ± 139 a  | 144 ± 23 b                   | 3.13 ± 0.95 a                                      | 24.8 ± 10 bc              | 107 ± 41 bc             |
|      |        |               | K500      | 3                 | 4733 ± 551 a               | 1089 ± 118 a | 208 ± 26 a                   | 1.19 ± 0.77 b                                      | 6.4 ± 0.9 c               | 28 ± 4 c                |
|      |        | Carrot        | K0        | 3                 | 1111 ± 446 a               | 107 ± 43 a   | 89 ± 26 a                    | 3.22 ± 0.62 a                                      | 19.3 ± 5.1 a              | 203 ± 35 a              |
|      |        |               | K150      | 3                 | 1410 ± 176 a               | 122 ± 19 a   | 101 ± 42 a                   | 2.96 ± 0.97 ab                                     | 15.7 ± 4.7 a              | 182 ± 58 a              |
|      |        |               | K300      | 3                 | 789 ± 272 a                | 76 ± 23 a    | 139 ± 23 a                   | 2.78 ± 1.3 ab                                      | 12.0 ± 4.0 ab             | 127 ± 53 ab             |
|      |        |               | K500      | 3                 | 835 ± 102 a                | 85 ± 10 a    | 124 ± 22 a                   | 0.81 ± 0.21 b                                      | 1.7 ± 0.6 b               | 19 ± 4 b                |

Numbers after ± denote the standard deviation. Different letters represent significant differences according to the Tukey's HSD test ( $p < 0.05$ ). Experimental procedures are same as shown in Table S1.

decontamination process was also considered that it was difficult to remove the contaminated soil equally from a field. Thus, the evaluation of <sup>137</sup>Cs uptake is not simply carried out by the plant <sup>137</sup>Cs, the concept of TF is important and widely used to explain the difference of <sup>137</sup>Cs uptake from the soil to plant.

### 3.2. Pot experiment

#### 3.2.1. Repeated experiment using Soil C in 2020 and 2021

Edamame, turnip root, and spinach were cultivated on the same soil with different range of K application level for 2 years. There was no significant difference in the harvest (fresh weight) in all the crop species regardless of years (Table 5). On the other hand, the harvest weight is lower in 2021 than in 2020 in every crop species. This may be due to the lower irradiation and temperature in 2021, but the detailed information of the greenhouse was not collected. ExK increased significantly with increasing K application level, that is even at harvest time, that is 177, 181, and 94 mg K kg<sup>-1</sup> at K0 treatment and increased to 249, 243, and 171 mg K kg<sup>-1</sup> at K150 treatment in edamame, spinach, and turnip root, respectively, in 2020 (Table 5). And, 112, and 174, and 96 mg K kg<sup>-1</sup> at K0

treatment and increased to 354, 318, and 211 mg K kg<sup>-1</sup> at K500 treatment in edamame, spinach, and turnip root, respectively, in 2021. When the applied K level increased the <sup>137</sup>Cs concentration of plant decreased. In 2020, the level was 122, 91, and 1262 Bq kg<sup>-1</sup> dry at K0 treatment and decreased to 53, 49, and 497 Bq kg<sup>-1</sup> dry at K150 treatment in edamame, spinach, and turnip root, respectively (Table 5). In 2021, it was 376, 60, and 1123 at K0 treatment and decreased to 58, 30, and 84 Bq kg<sup>-1</sup> dry at K500 treatment in edamame, spinach, and turnip root, respectively. Although <sup>137</sup>Cs concentration on dry weight basis was high in turnip root compared to other crops, the difference of the concentration on fresh weight basis was rather small. High concentration of water of turnip root dilutes the radioactivity of plant. A decrease in <sup>137</sup>Cs in spinach was observed but not significantly confirmed.

#### 3.2.2. Repeated experiment in a year, using komatsuna in Soil D

In 2021, komatsuna was cultivated twice in a year by using the same soil (without additional fertilization at the second trial). The yield was not changed by the different K level in the first trial, while the yield of K0 treatment was significantly lower

**Table 5.** Growth of harvest and soil characteristics after the experiment (Pot experiment).

| Year | Field  | Vegetables    | Treatment | Number of samples | Yield (g pot <sup>-1</sup> ) |             | ExK<br>mgK kg <sup>-1</sup> | Soil <sup>137</sup> Cs<br>kBq kg <sup>-1</sup> dry | Plant <sup>137</sup> Cs   |                         |
|------|--------|---------------|-----------|-------------------|------------------------------|-------------|-----------------------------|--|---------------------------|-------------------------|
|      |        |               |           |                   | Fresh                        | Dry         |                             |  | Bq kg <sup>-1</sup> fresh | Bq kg <sup>-1</sup> dry |
| 2020 | Soil C | Edamame       | K0        | 3                 | 122 ± 7 a                    | 38 ± 1 a    | 177 ± 23 a                  | 2.81 ± 0.10 a                                      | 38 ± 7 a                  | 122 ± 19 a              |
|      |        |               | K75       | 3                 | 123 ± 10 a                   | 38 ± 2 a    | 198 ± 33 a                  | 2.62 ± 0.05 a                                      | 24 ± 3 b                  | 77 ± 11 b               |
|      |        |               | K150      | 3                 | 130 ± 10 a                   | 39 ± 3 a    | 249 ± 35 a                  | 2.85 ± 0.15 a                                      | 16 ± 4 b                  | 53 ± 13 b               |
|      |        | Spinach       | K0        | 3                 | 91 ± 25 a                    | 10 ± 3 a    | 181 ± 13 a                  | 2.84 ± 0.26 a                                      | 11 ± 1.7 a                | 91 ± 17 a               |
|      |        |               | K75       | 3                 | 107 ± 5 a                    | 12 ± 1 a    | 196 ± 18 a                  | 2.53 ± 0.15 a                                      | 7.5 ± 0.4 b               | 66 ± 8 ab               |
|      |        |               | K150      | 3                 | 112 ± 6 a                    | 13 ± 1 a    | 243 ± 39 a                  | 2.72 ± 0.19 a                                      | 5.6 ± 0.4 b               | 49 ± 2 b                |
|      |        | Turnip (root) | K0        | 3                 | 512 ± 6 a                    | 31 ± 1 b    | 94 ± 7 c                    | 2.82 ± 0.05 a                                      | 77 ± 13 a                 | 1262 ± 221 a            |
|      |        |               | K75       | 3                 | 522 ± 29 a                   | 34 ± 1 a    | 122 ± 16 b                  | 2.70 ± 0.23 a                                      | 67 ± 15 a                 | 1023 ± 250 ab           |
|      |        |               | K150      | 3                 | 528 ± 28 a                   | 34 ± 1 a    | 171 ± 6 a                   | 3.05 ± 0.07 a                                      | 32 ± 9 b                  | 497 ± 150 b             |
| 2021 | Soil C | Edamame       | K0        | 3                 | 90 ± 5 a                     | 20 ± 1 a    | 112 ± 10 c                  | 2.06 ± 0.18 b                                      | 85 ± 8 a                  | 376 ± 36 a              |
|      |        |               | K150      | 3                 | 87 ± 11 a                    | 20 ± 3 a    | 179 ± 22 bc                 | 2.36 ± 0.07 a                                      | 39 ± 3 b                  | 173 ± 13 b              |
|      |        |               | K300      | 3                 | 78 ± 3 a                     | 18 ± 0 a    | 283 ± 16 ab                 | 2.35 ± 0.05 a                                      | 20 ± 2 c                  | 87 ± 9 c                |
|      |        |               | K500      | 3                 | 81 ± 9 a                     | 19 ± 2 a    | 354 ± 115 a                 | 2.34 ± 0.09 a                                      | 13 ± 2 c                  | 58 ± 6 c                |
|      |        | Spinach       | K0        | 3                 | 65 ± 9 a                     | 7.8 ± 0.9 a | 174 ± 25 b                  | 2.40 ± 0.05 a                                      | 7.2 ± 3.8 a               | 60 ± 32 a               |
|      |        |               | K150      | 3                 | 103 ± 43 a                   | 11 ± 3.7 a  | 216 ± 8 b                   | 2.43 ± 0.02 a                                      | 8.7 ± 2.7 a               | 79 ± 26 a               |
|      |        |               | K300      | 3                 | 60 ± 5 a                     | 7.1 ± 0.8 a | 277 ± 21 a                  | 2.25 ± 0.12 a                                      | 5.0 ± 3.6 a               | 42 ± 30 a               |
|      |        |               | K500      | 3                 | 78 ± 14 a                    | 8.8 ± 1.3 a | 318 ± 13 a                  | 2.44 ± 0.13 a                                      | 3.3 ± 1.2 a               | 30 ± 12 a               |
|      |        | Turnip (root) | K0        | 3                 | 180 ± 48 a                   | 12 ± 4 a    | 96 ± 18 c                   | 2.53 ± 0.01 a                                      | 75 ± 29 a                 | 1123 ± 375 a            |
|      |        |               | K150      | 3                 | 232 ± 12 a                   | 13 ± 1 a    | 130 ± 15 bc                 | 2.57 ± 0.09 a                                      | 30 ± 3.0 b                | 524 ± 67 b              |
|      |        |               | K300      | 3                 | 238 ± 14 a                   | 14 ± 2 a    | 185 ± 34 ab                 | 2.54 ± 0.02 a                                      | 11 ± 2.9 b                | 178 ± 41 b              |
|      |        |               | K500      | 3                 | 241 ± 11 a                   | 15 ± 2 a    | 211 ± 17 a                  | 2.65 ± 0.16 a                                      | 5.3 ± 2.5 b               | 84 ± 35 b               |
|      | Soil D | Komatsuna 1st | K0        | 4                 | 79 ± 6 a                     | 5.4 ± 0.5 a | 25 ± 6 c                    | 1.61 ± 0.07 a                                      | 109 ± 23 a                | 1013 ± 150 a            |
|      |        |               | K150      | 4                 | 83 ± 8 a                     | 6.3 ± 0.8 a | 91 ± 34 b                   | 1.61 ± 0.14 a                                      | 85 ± 35 ab                | 793 ± 233 ab            |
|      |        |               | K300      | 4                 | 81 ± 5 a                     | 7.1 ± 0.6 a | 129 ± 34 b                  | 1.63 ± 0.11 a                                      | 62 ± 10 bc                | 600 ± 84 bc             |
|      |        |               | K500      | 4                 | 77 ± 7 a                     | 6.2 ± 1.2 a | 268 ± 4 a                   | 1.56 ± 0.12 a                                      | 27 ± 6 c                  | 300 ± 39 c              |
|      |        | Komatsuna 2nd | K0        | 4                 | 60 ± 10 c                    | 4.7 ± 0.6 b | 29 ± 2 d                    | 1.59 ± 0.02 a                                      | 128 ± 15 a                | 1613 ± 95 a             |
|      |        |               | K150      | 4                 | 120 ± 20 ab                  | 9.0 ± 1.8 a | 93 ± 12 c                   | 1.49 ± 0.04 b                                      | 37 ± 4 b                  | 497 ± 49 b              |
|      |        |               | K300      | 4                 | 139 ± 10 ab                  | 11 ± 1.1 a  | 164 ± 28 b                  | 1.54 ± 0.04 ab                                     | 17 ± 1 c                  | 225 ± 12 c              |
|      |        |               | K500      | 4                 | 110 ± 9 a                    | 8.3 ± 0.5 a | 318 ± 30 a                  | 1.63 ± 0.05 a                                      | 13 ± 1 c                  | 169 ± 14 c              |
|      |        | Turnip (root) | K0        | 4                 | 149 ± 16 a                   | 9.6 ± 1.4 b | 42 ± 2 b                    | 1.73 ± 0.05 a                                      | 51 ± 13 a                 | 781 ± 163 a             |
|      |        |               | K150      | 4                 | 160 ± 16 a                   | 12 ± 1.0 a  | 71 ± 8 b                    | 1.72 ± 0.07 a                                      | 33 ± 4 b                  | 458 ± 54 b              |
|      |        |               | K300      | 4                 | 171 ± 4 a                    | 12 ± 0.3 a  | 101 ± 18 b                  | 1.73 ± 0.09 a                                      | 17 ± 2 c                  | 257 ± 34 c              |
|      |        |               | K500      | 4                 | 153 ± 4 a                    | 10 ± 0.3 ab | 184 ± 65 a                  | 1.84 ± 0.09 a                                      | 11 ± 4 c                  | 168 ± 56 c              |
|      |        | Turnip (leaf) | K0        | 4                 | 88 ± 12 a                    | 7.9 ± 1.4 a | 42 ± 2 b                    | 1.73 ± 0.05 a                                      | 184 ± 46 a                | 2053 ± 454 a            |
|      |        |               | K150      | 4                 | 86 ± 11 a                    | 8.6 ± 1.0 a | 71 ± 8 b                    | 1.72 ± 0.07 a                                      | 140 ± 15 a                | 1390 ± 96 b             |
|      |        |               | K300      | 4                 | 83 ± 5 a                     | 7.7 ± 0.3 a | 101 ± 18 b                  | 1.73 ± 0.09 a                                      | 74 ± 13 b                 | 792 ± 129 c             |
|      |        |               | K500      | 4                 | 76 ± 7 a                     | 7.0 ± 0.4 a | 184 ± 65 a                  | 1.84 ± 0.09 a                                      | 53 ± 14 b                 | 570 ± 164 c             |

Numbers after ± denote the standard deviation. Different letters represent significant differences according to the Tukey's HSD test ( $p < 0.05$ ). Exchangeable K was extracted by 1 M ammonium acetate and analyzed by atomic absorption spectrometry (Agilent, 280FS AA).

than other treatments regardless of fresh weight or dry weight basis in second trial (Table 5). ExK level at the harvest of each trial at K0 treatment was 25 and 29 mgK kg<sup>-1</sup>, in the first and second trials, respectively, and these low values may affect the productivity of komatsuna, and increased to 268 and 318 mgK kg<sup>-1</sup> in the first and second trials, respectively. However, as the difference of ExK level in the first and second trials was not significant (data not shown), thus the other factor except for ExK may have an effect on the productivity. <sup>137</sup>Cs concentration decreased with increasing the K application in both trials. At K0 treatment, it was 1013 and 1613 Bq kg<sup>-1</sup> in the first and second trials, respectively, and decreased to 300 and 169 Bq kg<sup>-1</sup> in the first and second trials, respectively, at K500 treatment. The decrease of radioactivity can be explained by the increase of ExK level by K application, however the difference of radioactivity at K0 level could not be explained simply by ExK level between trials.

### 3.3. Effect of soil on the relationship between ExK and TF in pot experiment

In 2021, turnip was grown in soil C and soil D at the same time in the same greenhouse. Turnip root was used to detect the soil

effect. The growth in soil D was smaller than in Soil C. In soil D, EC, exchangeable Ca, and exchangeable Mg were lower than that of soil C before the cultivation (Table S2). The lower fertility of soil D may reduce the productivity of turnip. ExK level at the harvest at K0 treatment was 96 and 42 mg K kg<sup>-1</sup> in soil C and D, respectively, and increased to 211 and 184 mg K kg<sup>-1</sup> at K500 treatment (Table 5). <sup>137</sup>Cs concentration was decreased with increasing the K application in both soils. It was 1123 and 781 Bq kg<sup>-1</sup> in soil C and soil D, respectively, at K0 treatment and decreased to 84 and 168 Bq kg<sup>-1</sup> in soil C and soil D, respectively, at K500 treatment. It seems that the decrease of <sup>137</sup>Cs concentration with K level was steeper in soil C than in soil D.

### 3.4. Relationship between ExK and TF

K is known to have similar chemical properties in the soil solution; therefore, K in the solution competes with Cs at the absorption site of the plant root (Zhu and Smolder 2000; White and Broadley 2000). Furthermore, it is known that the K transporter absorbs Cs due to its high Cs affinity, especially under low K conditions (Fujimura et al. 2014; Rai and Kawabata 2020). The relationships between ExK at harvest and the TF of each crop cultivated under field conditions in 2020 and 2021

are shown in Figure 1. In all crop species and years (locations), there was a clear trend that the TF increased with the decrease in ExK. This result was also confirmed in pot experiments (Figures 2 and 3). The fitting curve was produced by changing the TF into a reciprocal value because as K and Cs compete with each other, the higher K application level may decrease the Cs uptake (indicating a low TF) and vice versa.

## 4. Discussion

### 4.1. Relationship between ExK and TF in field experiment

The differences among crop species have been demonstrated in IAEA (International Atomic Energy Agency) (2010), but the simple comparison of TF among species fluctuated too much and could not be directly applied to agricultural countermeasures. It is

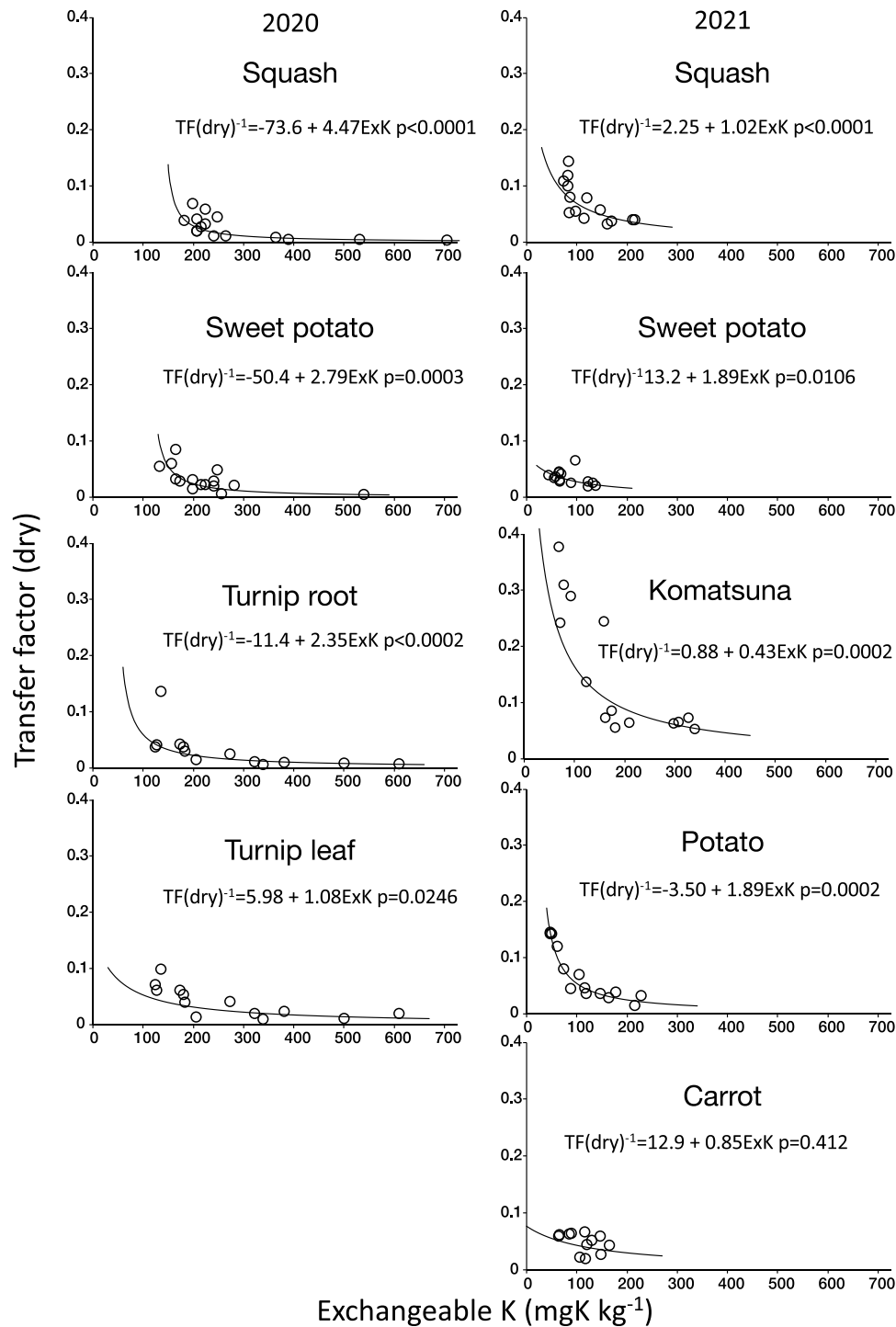


Figure 1. Relationship between ExK and TF of a field experiment in 2020 and 2021.



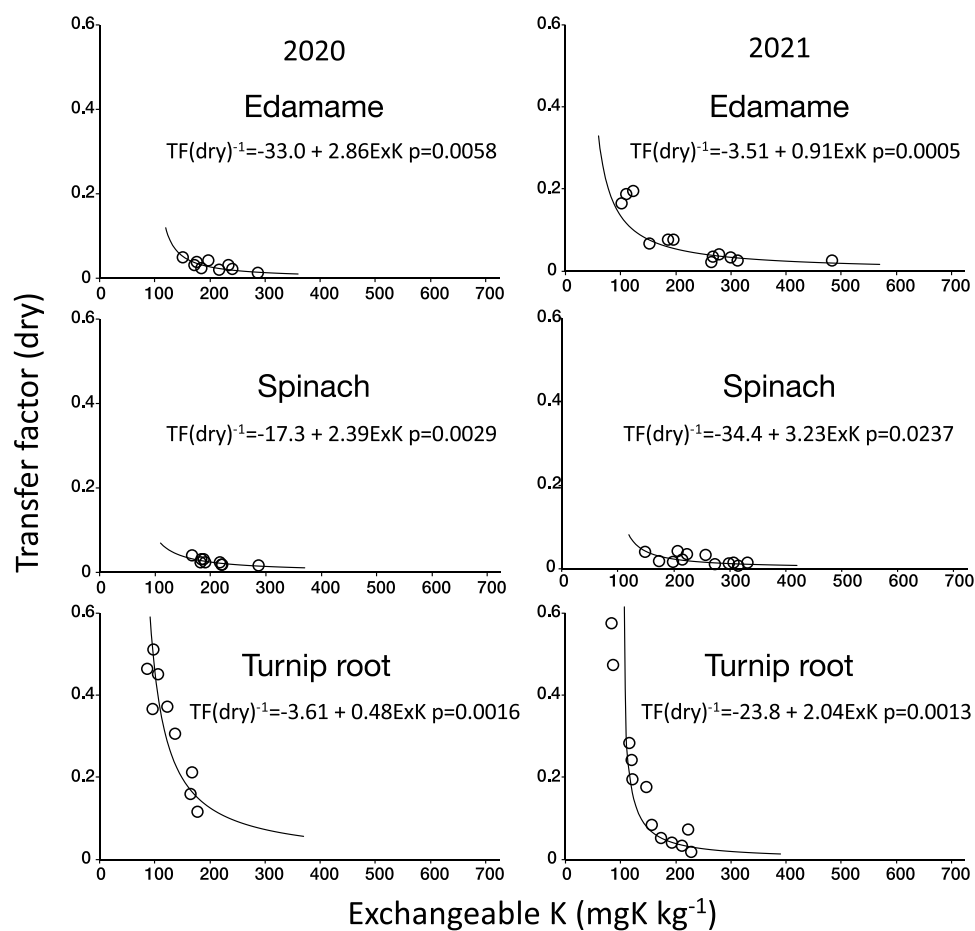


Figure 2. Relationship between ExK and TF of a pot experiment using Soil C in 2020 and 2021.

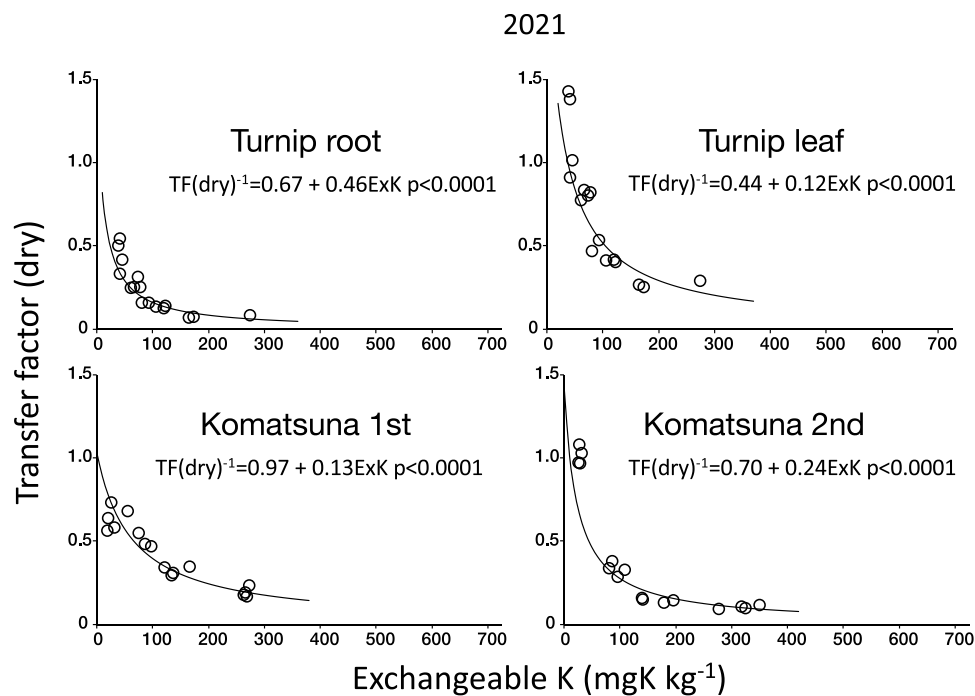


Figure 3. Relationship between ExK and TF of a pot experiment using Soil D in 2021.

important to determine the relationship between a wide range of ExK levels and TF values in the field, as demonstrated in rice, soybean, and buckwheat (IAEA (International Atomic Energy Agency) 2020), and this will also be required for vegetables. In the present experiment, we have tried to demonstrate the ExK-TF relationship of squash, sweet potato, turnip root, turnip leaf, komatsuna, potato, and carrot by using field condition, and turnip root, turnip leaf, spinach, and edamame in pot experiment. This relationship is the most important factor to decide the actual countermeasure under radioactive cesium contamination after the nuclear accident (e.g., Kato et al. 2015). In all the examined vegetables in field experiment, a close relationship between ExK and TF was observed (Figure 1). However, the relationship among species and between soil type was not equal (Figure 1). In the examined vegetables, komatsuna seems to have higher TF (dry weight basis) at the same ExK compared to other species, and sweet potato and carrot seem to have lower TF. It should be mentioned that as the water content is high in komatsuna leaf (more than 90%), the TF (fresh weight basis) will be low.

Squash and sweet potato were cultivated in both Soil A, 2020 and Soil B, 2021. The relationship between ExK-TF was not same in Soil A and Soil B. It seems that the TF was higher in Soil B in case of squash, but the difference in sweet potato was not clear (Figure 1). It is required to have similar and wider ExK variation and under similar environmental condition to evaluate the difference but the importance of ExK to regulate TF was consistent. Soil chemical properties were very different between Soil A and B. As CEC, exchangeable Ca, Mg, total carbon, and total nitrogen were higher in Soil A than in Soil B (Table S1). These differences are partly derived by the composition of dressed soil after the decontamination process and may also affect the relationship between ExK-TF. However, the information about the dressed soil after FDNPP accident is limited (Yoshino et al. 2015), it is required to have a large survey of how the dressed soil was obtained and applied to each field to have more precise evaluation of ExK-TF relationship.

#### 4.2. Effect of repeated cultivation on the relationship between ExK and TF in pot experiment

As the transfer of radiocesium from soil to plants is strongly regulated by the ratio of the amounts of Cs and K in the soil solution (Shaw et al. 1992; Smolder et al. 1996), other factors that may change this ratio and radiocesium transfer, such as soil type and ongoing fixation in the soil, have been reported (e.g., Yamaguchi et al. 2016). We chose two combinations to examine the effect of other factors on the relationship between ExK and TF by using ANCOVA (Table 6). (1) Turnip root, spinach, and edamame were used to evaluate the ongoing fixation by using the same soil in two consecutive years in pot cultivation and (2) komatsuna in a pot trial was used to determine the ongoing fixation by repeated cultivation using the same soil in a year.

In Table 3, 2 years of cultivation and repeated cultivation in a year were evaluated by conducting an ANCOVA, and the results are shown in Table 6. Radiocesium fixation to the soil clay has been reported to increase annually (Tsukada 2014), and fixation to the field soil was especially obvious in the early

Table 6. Results of ANCOVA.

|   |                       | Effect of interaction ( <i>p</i> value) |         |                    |
|---|-----------------------|---|---------|--------------------|
| Trial   | Species               | ExK                                     | Factor  | Interaction effect |
| Pot trial (repeated in 2 years using the same soil) |                       | Year                                    |         |                    |
|   | Turnip root           | 0.0025                                  | 0.057   | 0.042              |
|   | Spinach               | 0.015                                   | 0.60    | 0.69               |
|   | Edamame               | <0.0001                                 | <0.0001 | 0.0092             |
| Pot trial (repeated in a year)                      | Komatsuna 1st and 2nd | <0.0001                                 | <0.0001 | 0.0002             |
| Pot trial (different soils in the same year)        |                       | Soil                                    |         |                    |
|   | Turnip root           | <0.0001                                 | 0.78    | 0.0003             |

stages after the accident (Yamamura et al. 2018). Radiocesium fixation to soil is known as an ongoing fixation and has been considered that it may decrease the risk of radiocesium transfer from soil to plant (MAFF (Ministry of Agriculture, Forestry and Fisheries) 2021). From a 2-year experiment using the same soil by pot experiments, there was a significant interaction effect, except for spinach (Table 6). However, it is not able to confirm that there is an ongoing fixation in edamame, because even at the same ExK level, the TF was higher (Figure 2). In case of turnip root, it seems that the ExK-TF relationship decreased in 2021; however, as the range of ExK between 2 years is different, it is not suitable to conclude that ongoing fixation is confirmed. Other environmental factors should be considered. Besides the effect of repeated cultivation, the effect of soil is clearly demonstrated. It is important to consider the difference of soil to determine the precise prediction of required amount of ExK to regulate TF.

From the experiment by a repeated cultivation trial within a year using komatsuna in the pot experiment, the interaction effect was positive, and it seems that the TF was lower in the second trial than in the first trial at a similar ExK level (demonstrated from the 100 to 200 mg K<sub>2</sub>O kg<sup>-1</sup> range) (Table 6, Figure 3). However, it is not simply concluded that there was a fixation progressed in this experiment. As the pot soil was thoroughly mixed before the second trial, and it is considered to have similar effect as plowing in the field condition. Plowing may accompany the mechanical increase in the contact of <sup>137</sup>Cs with the soil, and it is expected that this process promotes the fixation rate. On the other side, plant root uptake nutrient from the soil solution and available fraction as a source for nutrient. As observed in ExK level change, plant growth can reduce the available fraction of soil, and it is also expected the decrease in exchangeable <sup>137</sup>Cs. That is, there is a possibility that repeated cultivation especially during a short time repetition may decrease the exchangeable <sup>137</sup>Cs then decrease the TF. It is required to check how the exchangeable <sup>137</sup>Cs level changes in these repetition experiments. Furthermore, as the other environmental conditions (e.g., temperature) were not controlled, further precise evaluation is needed. Furthermore, fertilization is also known to increase fixation to the soil (Kubo et al. 2017; Wakabayashi et al. 2022), and it is also important to know how the availability of <sup>137</sup>Cs (exchangeable <sup>137</sup>Cs) changed through these processes. In addition, the growth of K0 treatment only significantly low

**Table 7.** Estimation of required ExK level for the harvest radioactivity.

| Species      | Soil condition | Coefficients of the regression equation for fresh harvest <sup>a</sup> |       | Required ExK (mg K kg <sup>-1</sup> ) to keep the harvest radioactivity less than |                              |
|--------------|----------------|--|-------|---|------------------------------|
|              |                | a  | b     | 100 Bq kg <sup>-1</sup> fresh   | 25 Bq kg <sup>-1</sup> fresh |
| Squash       | Field, Soil A  | 2.20   | -269  | 136   | 177                          |
| Squash       | Field, Soil B  | 0.74   | -8.66 | 53  | 175                          |
| Sweet potato | Field, Soil A  | 1.87   | -228  | 138   | 186                          |
| Sweet potato | Field, Soil B  | 0.67   | 33.8  | <0  | 129                          |
| Turnip root  | Field, Soil A  | 8.17   | -666  | 85  | 96                           |
| Turnip leaf  | Field, Soil A  | 0.69   | 163   | <0  | <0                           |
| Komatsuna    | Field, Soil B  | 0.98   | 25    | 5   | 97                           |
| Potato       | Field, Soil B  | 0.99   | -20.8 | 51  | 142                          |
| Carrot       | Field, Soil B  | 1.00   | 158   | <0  | <0                           |
| Edamame      | Pot, Soil C    | 0.47   | -13.3 | 92  | 284                          |
| Spinach      | Pot, Soil C    | 3.46   | -321  | 102   | 128                          |

<sup>a</sup>The equation is as follows:  $TF(\text{Fresh weight basis})^{-1} = b + a \cdot \text{ExK}$ .  
Soil radiocesium Cs concentration is estimated as 3,000 Bq kg<sup>-1</sup> dry.

in second trial, it is speculated that the contribution of other K source (such as non-exchangeable K) in addition to ExK may explain the difference between first and second trials, but further research is required.

#### 4.3. Effect of soil type on the relationship between ExK and TF in pot experiment

From the observation of more than 400 fields over 6 years in Fukushima Prefecture, to compare different soils, it was suggested that the relationship between ExK and TF was not similar in the Nakadori and Hamadori areas (Yamamura et al. 2018). Two experimental sites were used in this study in different years. An interaction effect was observed in turnip root in the pot experiments (Table 6). Soil D seems to have lower fertility from the data of EC, ExK, Ex Mg, and Ex Ca compared to Soil C (Supplementary Table 2). Although we do not have the information about the original soil before the decontamination, it is suggested that contribution of additional soil by decontamination procedure is larger in soil D. It is speculated that the soil characteristics may change the relationship between ExK and TF through the availability of K and <sup>137</sup>Cs in the soil. Further investigation is required how the soil characteristics change the relationship between ExK and TF. The usage of non-contaminated soil after decontamination could change the original soil characteristics, and as the detail information of each decontamination process is not open, the effect of decontamination on ExK-TF behavior is required. It is suggested that exchangeable <sup>137</sup>Cs level is another important factor to explain the difference in the behavior of ExK-TF (Yagasaki et al. 2019a, 2019b), we are trying to develop another equation to cancel the different soil characteristics now (Suzuki et al. 2023). However, regardless of these interaction effects, ExK was clearly demonstrated to be the most powerful factor regulating TF.

#### 4.4. Countermeasure for the safe production of vegetables

The application of a sufficient amount of K was confirmed to be a feasible method for mitigating <sup>137</sup>Cs transfer from soil to plants. However, the effects of fixation and/or cultivation, soil type, and crop species must also be considered for precise

estimation of the required level of ExK during plant growth. From the results, the TF was calculated based on the fresh weight of the plant harvest, and the required level of ExK for each site and crop species was estimated. For this calculation, the data for komatsuna, squash, sweet potato, and turnip were obtained from the field, and the data for edamame and spinach were obtained using a pot (Table 3) because we did not have the data for these crops under field conditions. The TF values obtained from the pot experiments were higher than those of the field (Figures 1,2,3); therefore, the data based on pot experiments may be estimated on the safe side.

Based on the standard value of radioactivity in food, we calculated the required level of ExK using the following equation:

$$TF(\text{wet})^{-1} = b + a \cdot \text{ExK}$$

where TF (wet) was the ratio of <sup>137</sup>Cs concentration of the wet harvest to that of the dry soil, and a and b are coefficients. The estimated coefficients are summarized in Table 7. For this calculation, we set the soil <sup>137</sup>Cs concentration as 3,000 Bq kg (dry weight)<sup>-1</sup>, though the soil radioactivity is not constant even in a small area even after the decontamination this value is generally observed in the contaminated fields especially in the Hamadori area where the decontamination is still in progress (data not shown) and furthermore the value is close to the <sup>137</sup>Cs concentration we have collected for the experiment in this experiment. To evaluate the required amount of ExK, not only the standard value of 100 (Bq kg<sup>-1</sup> fresh) but also a quarter of that (25 Bq kg<sup>-1</sup> fresh) was set as the target value. Considering for the usage in the actual practice for agriculture in the contaminated fields, it is required to treat the product in fresh weight basis, we used the equation for TF estimation based on fresh weight of each data.

The value of 25 Bq kg<sup>-1</sup> fresh was close to the detection limit in most food monitoring locations, and this value was sometimes used to indicate exhaustive food safety for consumers. Table 7 shows that the required ExK level needed to produce all the examined vegetables under the standard limit (100 Bq kg<sup>-1</sup> fresh) using the same soil. In some cases (sweet potato and carrot in Soil B and turnip leaf in Soil A), no further countermeasures were needed. Additionally, to reduce the value to the detection limit (25 Bq kg<sup>-1</sup> fresh) as closely as possible, the ExK level higher than 200 mgK<sub>2</sub>O kg<sup>-1</sup> was maintained in most

cases, and for edamame, it was maintained at more than 300 mgK<sub>2</sub>O kg<sup>-1</sup>. These results indicate that even in the case of vegetables, a sufficient amount of potassium application is required to maintain high ExK levels during growth.

## 5. Conclusion

Several vegetables (komatsuna, squash, sweet potato, turnip, potato, carrot, spinach, and edamame) were investigated to determine the relationship between ExK and TF. Regardless of the crop species or other factors, such as soil type and/or year, TF was strongly negatively regulated by the amount of ExK. The ongoing fixation was not clearly observed especially under field condition, it should also be evaluated for other soil chemical properties. The differences among the soil types were demonstrated and must be investigated in the future. Taking into account all the factors, it is strongly supported that maintaining a sufficient amount of ExK is the most reliable method to decrease the uptake of <sup>137</sup>Cs from the soil.

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