



# Can we use the addition of stable $^{133}\text{Cs}$ to evaluate the uptake risk of the radioequivalent $^{137}\text{Cs}$ by crops?

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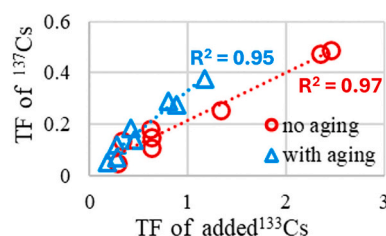
## HIGHLIGHTS

- Tracing stable  $^{133}\text{Cs}$  addition helps evaluate potential  $^{137}\text{Cs}$  uptake risk by crops
- Pot experiments with  $^{133}\text{Cs}$  addition accurately assess  $^{137}\text{Cs}$  uptake risk by crops
- Indices from batch  $^{133}\text{Cs}$  addition identified soils with high  $^{137}\text{Cs}$  uptake risk
- Exchangeable  $^{133}\text{Cs}$  and  $^{137}\text{Cs}$  exhibit similar  $K_d$  values

## GRAPHICAL ABSTRACT

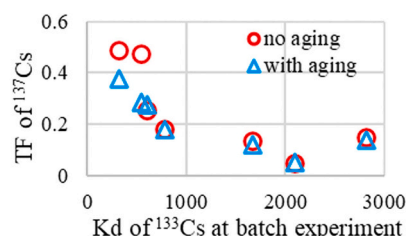
### Precise risk evaluation

=> Cultivation with  $^{133}\text{Cs}$  addition



### Screening of risky soil

=> Batch experiment alone



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## ABSTRACT

Assessing the risk of  $^{137}\text{Cs}$  root uptake by crops is crucial for nuclear emergency preparedness. However, this risk remains underexplored in low-latitude regions and the Southern Hemisphere compared to mid-latitude areas in the Northern Hemisphere due to lower  $^{137}\text{Cs}$  contamination rates, making  $^{137}\text{Cs}$  measurements impractically time-consuming. To evaluate  $^{137}\text{Cs}$  uptake risk using stable  $^{133}\text{Cs}$ , we conducted a batch experiment to determine the optimal amount of trace  $^{133}\text{Cs}$  addition one that minimally altered its solid-liquid distribution. Additionally, we performed a pot experiment with paddy rice (*Oryza sativa*, cv. Koshihikari) using eight soils with diverse clay mineralogy, collected from Fukushima and surrounding areas (2015–2020). The experiment included treatments with and without  $^{133}\text{Cs}$  addition and with and without an aging treatment (30 dry-wet cycles). The transfer factor of added  $^{133}\text{Cs}$  exhibited a strong linear correlation with that of  $^{137}\text{Cs}$  from the Fukushima accident ( $R^2 > 0.95$ ), demonstrating that combining batch experiments with pot cultivation and trace  $^{133}\text{Cs}$  addition effectively assessed  $^{137}\text{Cs}$  uptake risk in crops. The  $^{137}\text{Cs}$  transfer factor was five times lower than that of added  $^{133}\text{Cs}$  without wetting/drying cycles, aligning with the expected  $^{137}\text{Cs}$  aging post-2011. Indices from the batch experiment, including the solid-liquid distribution coefficient and exchangeable ratio of sorbed  $^{133}\text{Cs}$ , identified

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high-risk soils, suggesting batch experiments alone may suffice for screening. Furthermore, the  $^{137}\text{Cs}/^{133}\text{Cs}$  ratio in the soil exchangeable fraction and rice shoots indicated similar solid-liquid distribution coefficients, allowing estimation of  $^{137}\text{Cs}$  concentrations in soil solutions—otherwise impractically time-consuming—using  $^{133}\text{Cs}$  concentrations and the soil exchangeable  $^{137}/^{133}\text{Cs}$  ratio.

## 1. Introduction

The environmental behavior of radioactive cesium, particularly  $^{137}\text{Cs}$ , is a public health concern due to its long half-life of approximately 30 years and its ability to easily enter the food chain. The primary pathways for its entry are direct deposition onto plant surfaces and soil-to-plant transfer through root uptake. The risk associated with the latter persists longer than the former because the mobility of  $^{137}\text{Cs}$  in soil is low, owing to its strong affinity for certain clay minerals (Sawhney, 1964). Therefore, assessing the risk of  $^{137}\text{Cs}$  root uptake by crops is crucial, particularly for nuclear emergency preparedness in countries where nuclear power plants are located, under construction, or planned nearby. The risk of  $^{137}\text{Cs}$  root uptake is typically expressed as the transfer factor (TF), calculated as follows:

$$\text{TF} = (^{137}\text{Cs concentration of crop}) / (^{137}\text{Cs concentration of soil}) \quad (1)$$

The TF of  $^{137}\text{Cs}$  varies greatly depending on plant species (Tagami et al., 2020), soil potassium availability (Kato et al., 2015; Eguchi et al., 2015; Kurokawa et al., 2020), degree of aging (Absalom et al., 1995; Roig et al., 2007; Tagami et al., 2018; Takeda et al., 2013), and soil clay mineralogy (Kato et al., 2015; Vanheukelom et al., 2024). Among these factors, clay mineralogy has a particularly strong influence on the soil-to-crop transfer of  $^{137}\text{Cs}$ . For example, perennial ryegrass grown in tropical soils lacking 2:1 type clay minerals, which selectively retain  $^{137}\text{Cs}$ , exhibited TF values up to 10,000 times higher than those grown in temperate soils rich in 2:1 clay minerals (Vanheukelom et al., 2024). Accumulating information on TF values for crops is crucial in an emergency to support decision-making on resource allocation, such as K fertilizers. However, TF values in low-latitude areas and the Southern Hemisphere—where many nuclear power plants are operating or planned—remain underexplored compared to those in mid-latitude areas of the Northern Hemisphere (Kozhakhhanov et al., 2014). For example, a technical report from the IAEA published in 2010 included TF values for cereals derived from 470 data points for temperate regions (IAEA, 2010). In contrast, the IAEA TECDOC published in 2021 included only 21 data points from tropical regions (Doering et al., 2021). One of the reasons for the limited exploration of low-latitude areas and the Southern Hemisphere is the low level of  $^{137}\text{Cs}$  contamination, which makes the determination of  $^{137}\text{Cs}$  concentrations in crops laborious and time-consuming (Tsukada et al., 2023). One approach to overcome this constraint is the addition of radioactive cesium ( $^{137}\text{Cs}$  or  $^{134}\text{Cs}$ ) to soils in experimental settings (Smolders et al., 1997; Delvaux et al., 2000; Sreenivasa Chari et al., 2011, 2012; Vanheukelom et al., 2024). The radiocesium interception potential (RIP) is one of the major indices used to evaluate  $^{137}\text{Cs}$  retention by soils (Cremers et al., 1988; Wauters et al., 1996; Nakao et al., 2014). However, few studies have reported the influence of RIP on  $^{137}\text{Cs}$  uptake by crops (Delvaux et al., 2000; Takeda et al., 2014; Ministry of Agriculture, Forestry and Fisheries, 2014; Vanheukelom et al., 2024), primarily due to the limited availability of radioactive research facilities. Another potential approach is the use of stable cesium ( $^{133}\text{Cs}$ ), which can be measured more easily using conventional inductively coupled plasma mass spectrometry (ICP-MS) (Tsukada et al., 2023). Establishing a method to screen soils with a high risk of  $^{137}\text{Cs}$  uptake by crops using  $^{133}\text{Cs}$  as a proxy would greatly enhance preparedness for nuclear emergencies in countries with low contamination levels.

Several studies have reported linear correlations between the TF values of soil total  $^{137}\text{Cs}$  and total  $^{133}\text{Cs}$  (Tsukada and Nakamura, 1999; Tsukada et al., 2002; Ogasawara et al., 2018). The slopes of these

relationships were 4.5 for potato (Tsukada and Nakamura, 1999), 2.6 for polished rice (Tsukada et al., 2002), and 2.7 for paddy rice shoots (Ogasawara et al., 2018), indicating higher mobility of  $^{137}\text{Cs}$  compared to  $^{133}\text{Cs}$ . However, these results were obtained in limited geographical areas; hence, the slope values may not be applicable to soils in different regions, particularly where soil clay mineralogy differs substantially. In addition, no correlation between the TF values of grass for  $^{133}\text{Cs}$  and  $^{137}\text{Cs}$  was reported, even within specific areas of limited surface extent, such as near the Kaiga nuclear power station in India (Karunakara et al., 2013). Even though the TF of total  $^{133}\text{Cs}$  may not accurately reflect that of total  $^{137}\text{Cs}$  in certain areas,  $^{133}\text{Cs}$  in the labile fraction could serve as a useful proxy for  $^{137}\text{Cs}$ . Several studies have indicated that the  $^{137}\text{Cs}/^{133}\text{Cs}$  ratio is nearly equal in the soil exchangeable fraction and plant tissue (Ogasawara et al., 2018; Wakabayashi et al., 2020; Tsukada et al., 2023). Tsukada et al. (2023) proposed applying the specific activity ratio of  $^{137}\text{Cs}/^{133}\text{Cs}$  at a regional scale, which would significantly reduce the time and labor required to evaluate  $^{137}\text{Cs}$  transfer risk by replacing the time-consuming determination of  $^{137}\text{Cs}$  in crops with a rapid  $^{133}\text{Cs}$  analysis using ICP-MS. This approach is particularly beneficial in areas with low  $^{137}\text{Cs}$  contamination levels (Tsukada et al., 2023).

The evaluation of potential  $^{137}\text{Cs}$  uptake risk by crops during possible nuclear emergencies in mid-latitude areas and the southern hemisphere is also affected by the aging effect. The plant availability of  $^{137}\text{Cs}$  decreases over time (Absalom et al., 1995; Roig et al., 2007; Tagami et al., 2018; Takeda et al., 2013). Soil contamination by  $^{137}\text{Cs}$  in low-latitude areas and the southern hemisphere is generally derived from nuclear weapons testing in the 1960s, due to the relatively low impact of the Chernobyl and Fukushima accidents in these regions (Garcia Agudo, 1998; Steinhäuser et al., 2014). Therefore, it is difficult to evaluate the current  $^{137}\text{Cs}$  uptake risk during emergencies, as the aging process has reduced its bioavailability over the past 50 years. To emulate the aging effect and the consequent changes in the TF using  $^{133}\text{Cs}$ , Takeda et al. (2013) added trace amounts of  $^{133}\text{Cs}$ , which did not alter the solid-liquid distribution coefficient ( $K_d$ ) of  $^{137}\text{Cs}$ , and monitored changes in the extractability and plant availability of the added  $^{133}\text{Cs}$  over time. This trace  $^{133}\text{Cs}$  addition approach appears promising for evaluating  $^{137}\text{Cs}$  uptake risk by crops. However, to the best of our knowledge, the soil-to-plant transfer of trace  $^{133}\text{Cs}$  added to soils has not been directly compared to that of  $^{137}\text{Cs}$  contamination in agricultural fields.

The objective of this study was to establish a method for evaluating the  $^{137}\text{Cs}$  uptake risk by crops using  $^{133}\text{Cs}$  as a proxy for  $^{137}\text{Cs}$ . Trace amounts of  $^{133}\text{Cs}$  were added to eight soils that varied in clay mineralogy and  $^{137}\text{Cs}$  uptake risk. Both batch experiments and pot experiments with paddy rice were conducted. The distribution of the added  $^{133}\text{Cs}$  in the soil exchangeable fraction and its subsequent transfer to paddy rice shoots in the pot experiments were compared with those of  $^{137}\text{Cs}$  derived from the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident.

## 2. Materials and methods

### 2.1. Soils

Eight soils from agricultural fields and grassland in Fukushima and the surrounding areas, characterized by diverse clay mineralogy, were used in this study (Table 1). The soil types were classified as Fluvisols, except for soil 2, classified as Cambisol, and soil 8, classified as Andosol (Table S1), according to the Japanese digital soil map (National Institute for Agro-Environmental Sciences, 2019). Soils 1–7 contained significant

**Table 1**

Particle size distribution and crystalline clay mineralogy of soils.

No.	Clay %	Silt %	Sand %	Crystalline clay mineralogy <sup>†</sup>	Reference
1	11	16	73	Vr > HIV > > Mi	Soil 1 in Eguchi et al. (2023)
2	13	14	73	Vr > Sm, HIV/HIS, Mi	Soil 9 in Eguchi et al. (2023)
3	20	23	57	Vr, Mi, HIV	
4	37	19	44	Sm, Kt, HIS	
5	13	12	74	Vr, Mi/Vr, Mi	Field B in Eguchi et al. (2021)
6	14	15	70	Vr, Sm > HIV/HIS, Mi/Vr	Field C in Eguchi et al. (2021)
7	10	14	76	HIV > Mi, Mi/Vr, Ch	Pot soil in Eguchi et al. (2021)
8	16	12	72	not significant	Soil 11 in Eguchi et al. (2023)

Clay, &lt; 0.002 mm; Silt 0.002–0.02 mm; Sand 0.02–2 mm.

<sup>†</sup> Estimated from XRD charts (Fig. S1); Vr, vermiculite; HIV/HIS, hydroxy-interlayered vermiculite/smectite; Mi, mica; Mi/Vr, mica-vermiculite regular inter-stratified minerals.

amounts of 2:1 type clay minerals (Table 1). However, soils 6 and 7 exhibited a high risk of <sup>137</sup>Cs transfer due to their low selectivity for <sup>137</sup>Cs and K, resulting in increased <sup>137</sup>Cs mobility and a reduced capacity to retain K applied as a countermeasure to decrease <sup>137</sup>Cs absorption by plant roots (Eguchi et al., 2021; Eguchi et al., 2023). In contrast, soil 1 demonstrated a high K-fixing ability, attributed to its high vermiculite content and the low degree of Al-interlayering in the vermiculite (Kubo et al., 2018; Eguchi et al., 2023). Soil 8 was a grassland soil that did not contain a significant amount of 2:1 type clay minerals. Therefore, the soil-to-plant transfer risk of <sup>137</sup>Cs in the grass was high due to the soil's low retention capacity for both K and Cs (Eguchi et al., 2023).

Soil samples were collected from at least five points of each field. The sampling depth was approximately 10 cm for soil 6 with a shallow top soil, or 15 cm for other soils. The sampled soils were air-dried or oven-dried at 40 °C, passed through a 2 mm mesh, and then thoroughly mixed. The soils were collected between 2015 and 2020; thus, the rapid aging phase of <sup>137</sup>Cs was assumed to have passed (Roig et al., 2007; Tagami et al., 2018). Information on the sampling sites and general physicochemical properties is presented in Tables S1 and S2.

## 2.2. Batch experiment

Kd of <sup>133</sup>Cs was determined by shaking soil samples with a 0.01 M CaCl<sub>2</sub> solution containing trace amounts of <sup>133</sup>Cs (Staunton, 1994). In addition to Kd, the distribution ratio of sorbed <sup>133</sup>Cs in the exchangeable fraction was determined by extraction with 1 M NH<sub>4</sub>OAc. A 2.5 g portion of soil was placed in pre-weighed 50 mL centrifuge tubes in triplicate, followed by the addition of 25 mL of 0.01 M CaCl<sub>2</sub> solution containing CsCl at concentrations of 0, 25, 50, 100, 150, or 200 µg <sup>133</sup>Cs L<sup>-1</sup>. The tubes were shaken for 5 days at 20 °C and then centrifuged at 3000 rpm for 5 min. A portion of the clear supernatant was collected using a syringe for <sup>133</sup>Cs determination, and the remaining solution was discarded by decanting. The tubes were then weighed to calculate the amount of retained solution, after which 25 mL of 1 M NH<sub>4</sub>OAc was added. After shaking for 1 h at 20 °C, the tubes were centrifuged at 3000 rpm for 5 min, and the supernatants were collected. The solutions were passed through a 0.2 µm polypropylene membrane filter, and the <sup>133</sup>Cs concentration was determined by ICP-MS (NexION 350S, PerkinElmer, Waltham). The amount of <sup>133</sup>Cs sorbed by or released from the solid phase was plotted against the final <sup>133</sup>Cs concentration in the solution phase. The Kd was calculated as the slope of the line connecting the plot for the initial <sup>133</sup>Cs concentration (0 µg L<sup>-1</sup>) and each plot corresponding to the initial <sup>133</sup>Cs addition concentrations (25, 50, 100, 150, and 200 µg <sup>133</sup>Cs L<sup>-1</sup>). Although repeated ultracentrifugation is recommended for precise determination of the Kd of <sup>137</sup>Cs (Guivarch et al., 1999),

ultracentrifugation was omitted in this study to ensure wider applicability in laboratories without specialized equipment.

## 2.3. Pot cultivation

Paddy rice (*Oryza sativa* Cv. *Koshihikari*) was cultivated in 250 mL plastic cups for 28 days using soils with or without <sup>133</sup>Cs addition and with or without aging treatment. The amount of <sup>133</sup>Cs addition was set at 0.5 mg Cs kg<sup>-1</sup> air-dried soil and applied as a CsCl solution. A 20 g portion of soil was weighed into a plastic container, and the CsCl solution was applied as evenly as possible to achieve a <sup>133</sup>Cs application at 0.5 mg kg<sup>-1</sup> and a soil water content of 80 % of the water holding capacity (WHC). Another 20 g of soil was then added to the container, and the CsCl solution was applied again. This cycle was repeated until 200 g of soil and a total of 0.1 mg of added <sup>133</sup>Cs were obtained in each container. Six containers with <sup>133</sup>Cs-amended soil were prepared for each soil type. Of these, three containers were subjected to 30 wetting (80 % of WHC) and drying (overnight at 40 °C) cycles (30 W/D) to promote aging (Yamaguchi et al., 2019). The remaining three containers were oven-dried at 40 °C overnight (1 W/D). In addition, six containers of soil without <sup>133</sup>Cs addition were prepared, consisting of three 30 W/D samples and three 1 W/D samples.

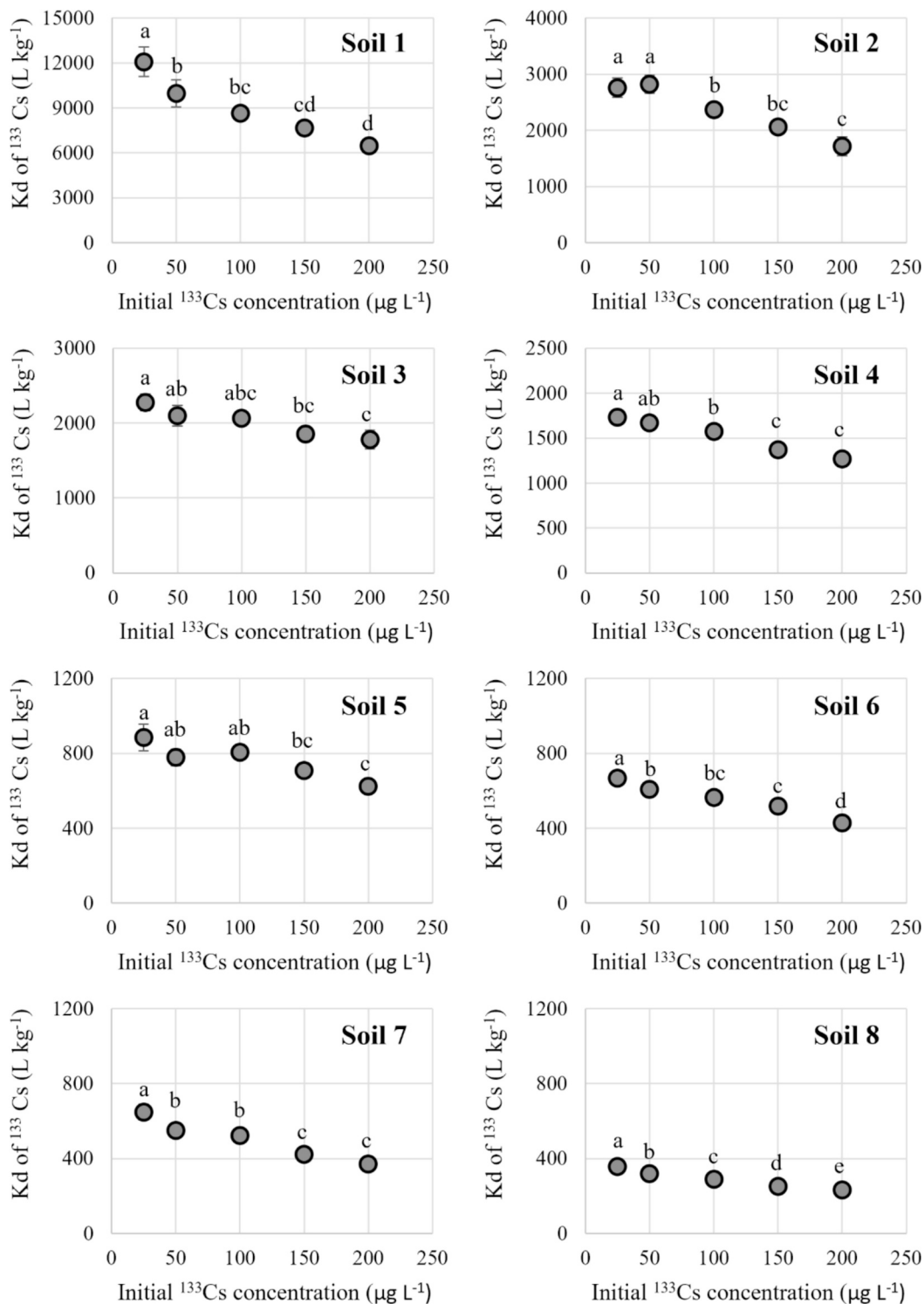
A 100 g portion of soil was weighed into a plastic container with a volume of 250 mL. Then, 10 mL of a solution containing NH<sub>4</sub>NO<sub>3</sub>, KH<sub>2</sub>PO<sub>4</sub>, and KCl—corresponding to N:P<sub>2</sub>O<sub>5</sub>:K<sub>2</sub>O = 50:50:50 mg—was added. Germinated paddy rice seeds were sown onto the soil at a rate of 100 grains per pot. An additional 50 g portion of soil was then added to cover the seeds. Cultivation was conducted in a growth chamber at 30 °C during a 12 h daylight period and 25 °C during a 12 h nighttime period. Additional fertilization with a solution containing N:P<sub>2</sub>O<sub>5</sub>:K<sub>2</sub>O = 50:50:50 mg was applied 15 days after sowing. Pots were irrigated with pure water once daily to maintain 60 % of WHC at the start of cultivation, and twice daily to reach 160 % of WHC by the end of the cultivation period. No signs of soil reduction—such as the development of a grayish color or the emission of a rotten egg smell—were observed throughout the experiment. At 28 days after sowing, rice shoots were harvested, dried at 80 °C for 48 h, weighed, and cut into segments of approximately 5 mm in length. The soil was air-dried and passed through a 2 mm mesh.

## 2.4. Analytical methods

Soils collected after harvest without <sup>133</sup>Cs addition were decomposed by lithium metaborate fusion and dissolved in 4 % HNO<sub>3</sub> for total <sup>133</sup>Cs analysis (Suhr and Ingamells, 1966). The total <sup>133</sup>Cs content in rice shoots was extracted by boiling in HNO<sub>3</sub>. Exchangeable <sup>133</sup>Cs and <sup>137</sup>Cs

were extracted by shaking the soil samples for 1 h with 1 M  $\text{NH}_4\text{OAc}$  at a soil-to-solution ratio of 1:10 at 20 °C. Due to the limited amount of soil sample and the low  $^{137}\text{Cs}$  concentration, exchangeable  $^{137}\text{Cs}$  in soils before planting was not determined. The  $^{133}\text{Cs}$  concentrations in extracts were determined using ICP-MS (NexION 350S, PerkinElmer, Waltham, MA, USA). The activity of  $^{137}\text{Cs}$  in soils, rice shoots, and extracts was measured by high purity Ge spectrometers (GC2520 or GC4520,

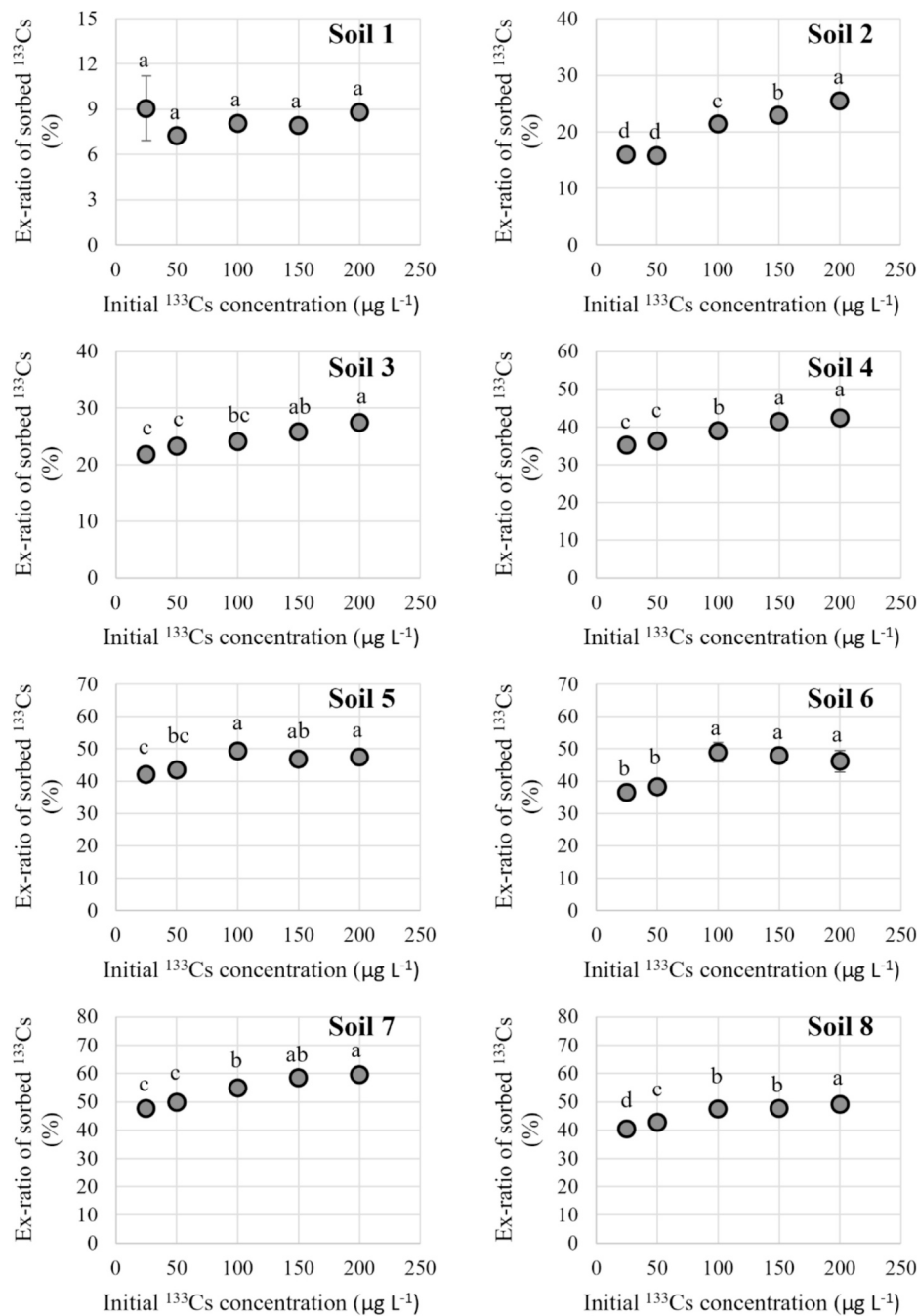
Canberra, Meriden, CT, USA), of which the detection efficiency was calibrated daily using a  $^{60}\text{Co}$  solution (CO401, Japan Radioisotope Association, Tokyo, Japan) and biennially using a mix nuclide solution (MX-033U8PP, Japan Radioisotope Association, Tokyo, Japan). The counting time was set for each sample to ensure the counting error of 10 % or less. Transfer factors (TF) of  $^{133}\text{Cs}$  or  $^{137}\text{Cs}$  to rice shoots were calculated based on soil total Cs (TF- $^{133}\text{Cs}$  or TF- $^{137}\text{Cs}$ ), added  $^{133}\text{Cs}$



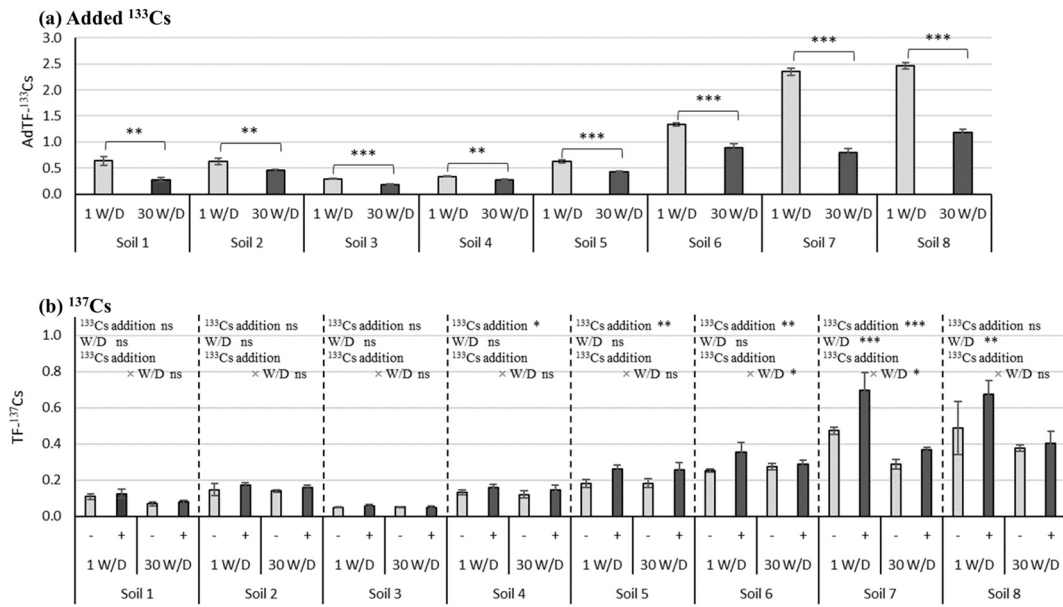
**Fig. 1.** Changes in the solid/liquid distribution coefficient of  $^{133}\text{Cs}$  with increasing initial  $^{133}\text{Cs}$  concentrations.

Kd, solid/liquid distribution coefficient.

Same alphabet indicates no significant difference in Tukey's test ( $p < 0.05$ ).



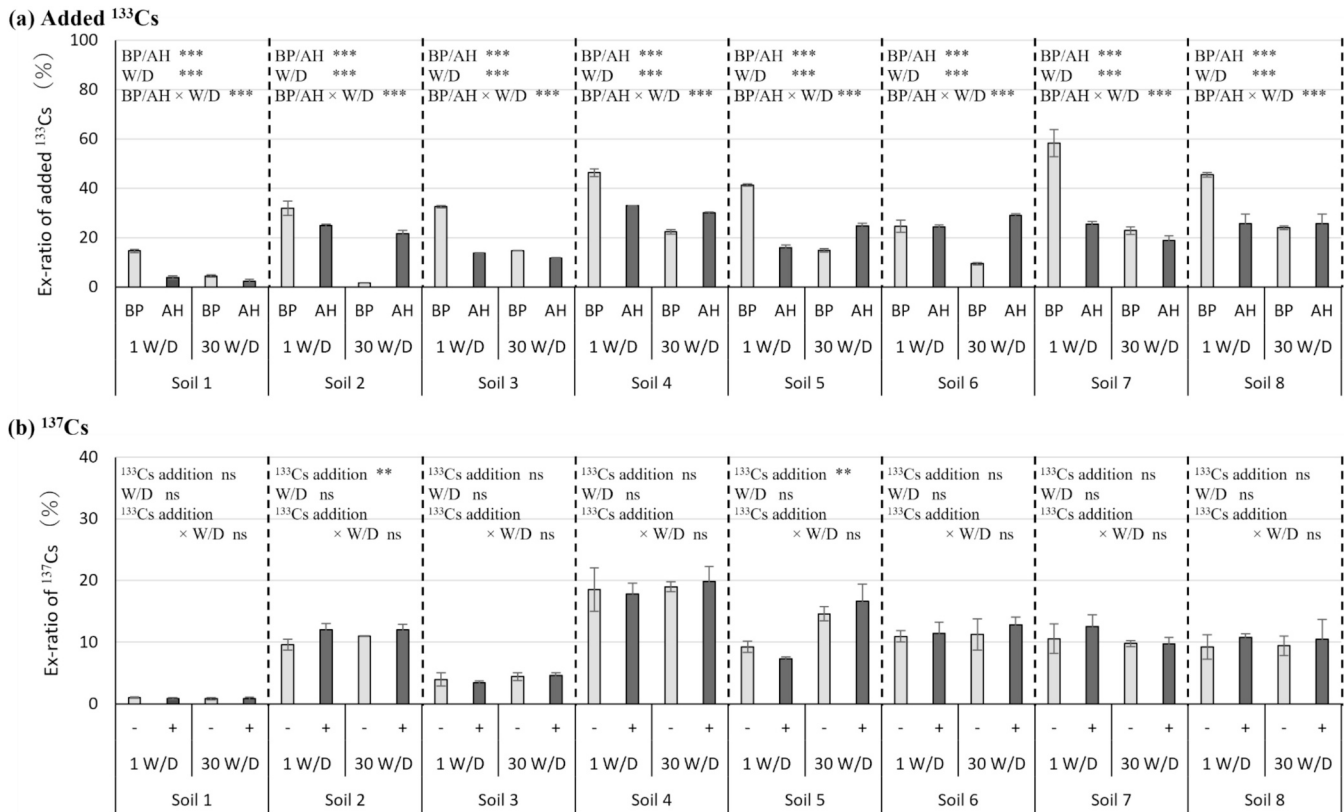
**Fig. 2.** Changes in the exchangeable ratio of sorbed  $^{133}\text{Cs}$  with increasing initial  $^{133}\text{Cs}$  addition concentrations.  
Ex-, exchangeable.  
Same alphabet indicates no significant difference in Tukey's test ( $p < 0.05$ ).



**Fig. 3.** Transfer factor of (a) added  $^{133}\text{Cs}$  and (b)  $^{137}\text{Cs}$ .

AdTF- $^{133}\text{Cs}$ , transfer factor of added  $^{133}\text{Cs}$ ; TF- $^{137}\text{Cs}$ , transfer factor of  $^{137}\text{Cs}$ ; W/D, wetting/drying cycle; −, without  $^{133}\text{Cs}$  addition; +, with  $^{133}\text{Cs}$  addition.

\*,  $p < 0.05$ ; \*\*,  $p < 0.01$ ; \*\*\*,  $p < 0.001$ ; in one-way or two-way ANOVA for each soil.



**Fig. 4.** Distribution ratios of (a) added  $^{133}\text{Cs}$  and (b)  $^{137}\text{Cs}$  in the soil exchangeable fraction.

Ex-, exchangeable; W/D, wetting/drying cycle; BP, before planting; AH, after harvest cycle; −, without  $^{133}\text{Cs}$  addition; +, with  $^{133}\text{Cs}$  addition.

\*,  $p < 0.05$ ; \*\*,  $p < 0.01$ ; \*\*\*,  $p < 0.001$ ; ns, not significant; in two-way ANOVA for each soils.

(AdTF- $^{133}\text{Cs}$ ) and exchangeable Cs (ExTF- $^{133}\text{Cs}$  or ExTF- $^{137}\text{Cs}$ ; Suzuki et al., 2022) as following:

$$\text{TF-}^{133}\text{Cs or TF-}^{137}\text{Cs} = (\text{rice shoot}^{133\text{ or }137}\text{Cs}) / (\text{soil total}^{133\text{ or }137}\text{Cs}) \quad (2)$$



$$\text{AdTF-}^{133}\text{Cs} = \{ (\text{rice shoot } ^{133}\text{Cs with } ^{133}\text{Cs addition}) - (\text{rice shoot } ^{133}\text{Cs without } ^{133}\text{Cs}) \} / (\text{amount of } ^{133}\text{Cs addition}) \quad (3)$$

$$\text{ExTF-}^{133}\text{Cs or Ex-TF}^{137}\text{Cs} = (\text{rice shoot } ^{133}\text{ or } ^{137}\text{Cs}) / (\text{soil exchangeable } ^{133}\text{ or } ^{137}\text{Cs}) \quad (4)$$

Soil exchangeable K was extracted by shaking the soil samples for 1 h with 1 M  $\text{NH}_4\text{OAc}$  at a soil-to-solution ratio of 1:10 at 20 °C and was determined by atomic absorption spectroscopy (AAS; ZA-3000, Hitachi High-Technologies Corporation, Tokyo, Japan).

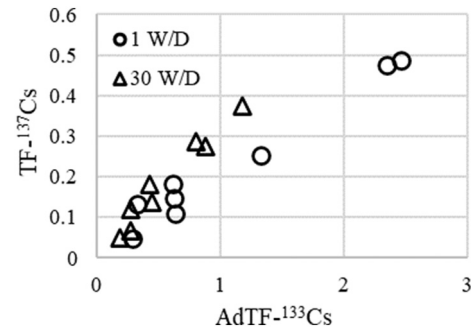
### 3. Results

#### 3.1. Batch experiment

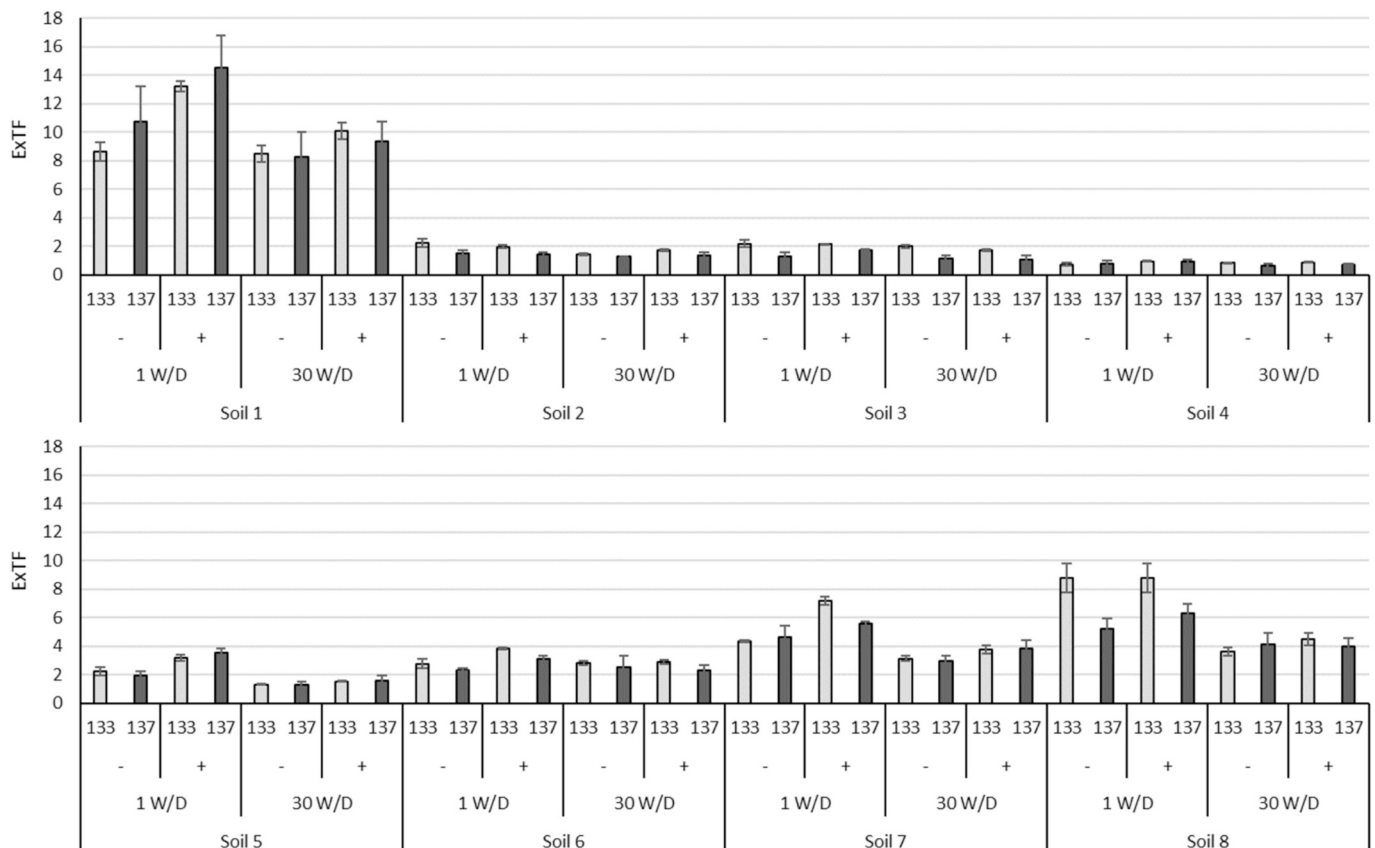
The batch experiment data on the distribution of added  $^{133}\text{Cs}$  are presented in Table S3. Figs. 1 and 2 illustrate the results for the solid-liquid distribution coefficient ( $K_d$ ) and the distribution ratio of sorbed  $^{133}\text{Cs}$  to the soil exchangeable fraction, respectively. At an initial  $^{133}\text{Cs}$  concentration of  $50 \mu\text{g L}^{-1}$ ,  $K_d$  values did not greatly decrease compared to those at  $25 \mu\text{g L}^{-1}$ . However,  $K_d$  values considerably decreased at initial  $^{133}\text{Cs}$  concentrations of  $100 \mu\text{g L}^{-1}$  or higher (Fig. 1). Similarly, the distribution ratio of sorbed  $^{133}\text{Cs}$  to the exchangeable fraction was comparable between 25 and  $50 \mu\text{g L}^{-1}$  but significantly increased at  $100 \mu\text{g L}^{-1}$  or higher, except for Soil 1 (Fig. 2). Since an initial concentration of  $50 \mu\text{g L}^{-1}$  corresponds to  $0.5 \text{ mg kg}^{-1}$  of air-dried soil—based on the soil-to-solution ratio of 1:10 in the batch experiment—the amount of  $^{133}\text{Cs}$  addition was set at  $0.5 \text{ mg kg}^{-1}$  air-dried soil.

#### 3.2. Pot experiment

Data from the pot experiment on soil exchangeable  $^{133}\text{Cs}$  and inorganic N before planting, soil K and Cs status after harvest, and Cs uptake by harvested rice shoots are presented in Tables S4–S6, respectively. The



**Fig. 6.** Relationship between the transfer factor of  $^{137}\text{Cs}$  and added  $^{133}\text{Cs}$ .  $\text{TF-}^{137}\text{Cs}$ , transfer factor of  $^{137}\text{Cs}$ ;  $\text{AdTF-}^{133}\text{Cs}$ , transfer factor of added  $^{133}\text{Cs}$ . 1 W/D;  $y = 0.187x + 0.026$  ( $r = 0.983$  \*\*\*). 30 W/D;  $y = 0.319x + 0.009$  ( $r = 0.977$  \*\*\*). \*\*\*,  $p < 0.001$  in Pearson's correlation coefficient.



**Fig. 5.** Transfer factor of exchangeable Cs. ExTF, transfer factor of exchangeable Cs; 133,  $^{133}\text{Cs}$ ; 137,  $^{137}\text{Cs}$ ; –, without  $^{133}\text{Cs}$  addition; +, with  $^{133}\text{Cs}$  addition; W/D, wetting/drying cycle.

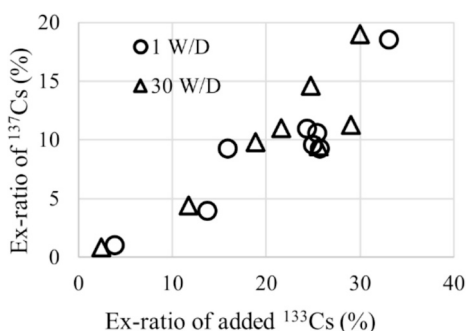


Fig. 7. Relationship between the distribution of  $^{137}\text{Cs}$  in the exchangeable fraction and that of added  $^{133}\text{Cs}$  after harvest in pot cultivation.

Ex-, exchangeable; W/D, wetting/drying cycle.

1 W/D;  $y = 0.519x - 0.017$  ( $r = 0.919^{**}$ ).

30 W/D;  $y = 0.535x - 0.009$  ( $r = 0.897^{**}$ ).

\*,  $p < 0.05$  in Pearson's correlation coefficient.

wetting/drying (W/D) cycle significantly decreased  $\text{AdTF}^{133}\text{Cs}$  in all soils (Fig. 3). In contrast, its effect on  $\text{TF}^{137}\text{Cs}$  was significant only in soils 7 and 8 (Fig. 3). The W/D cycles also reduced the distribution ratio of added  $^{133}\text{Cs}$  to the exchangeable fraction (Ex-ratio) (Fig. 4). Notably, the Ex-ratio of added  $^{133}\text{Cs}$  in 30 W/D increased during pot cultivation in soils 2, 4, and 6, whereas it tended to decrease in 1 W/D. The availability of inorganic N was higher in 30 W/D than in 1 W/D, except for soil 6. Similarly, the dry matter yield of harvested rice shoots was significantly higher in 30 W/D than in 1 W/D, except for soils 1, 6, and 7 (Table S6).

The  $\text{TF}^{137}\text{Cs}$  values were slightly higher in soils with added  $^{133}\text{Cs}$  than in non-added soils, although the degree of overestimation remained within an acceptable range for screening purposes (Fig. 3). Hereafter,  $\text{AdTF}^{133}\text{Cs}$  values are compared with  $\text{TF}^{137}\text{Cs}$  in soils without  $^{133}\text{Cs}$  addition. The  $\text{AdTF}^{133}\text{Cs}$  values were higher than  $\text{TF}^{137}\text{Cs}$  values, even when  $\text{AdTF}^{133}\text{Cs}$  in 30 W/D was compared to  $\text{TF}^{137}\text{Cs}$  in 1 W/D (Fig. 3). This reflects the greater degree of aging of  $^{137}\text{Cs}$ , primarily derived from the FDNPP accident, which occurred at least four years before soil sampling. In contrast, there was no consistent trend in whether  $\text{ExTF}^{133}\text{Cs}$  values were higher or lower than  $\text{ExTF}^{137}\text{Cs}$  values (Fig. 5). Overall,  $\text{ExTF}^{133}\text{Cs}$  and  $\text{ExTF}^{137}\text{Cs}$  showed similar values (Table S6).

## 4. Discussion

### 4.1. Risk evaluation by $^{133}\text{Cs}$ addition in pot experiment

The  $\text{AdTF}^{133}\text{Cs}$  values showed a strong linear correlation with  $\text{TF}^{137}\text{Cs}$  ( $R^2 > 0.95$ , Fig. 6). Soils 6–8, which posed a high  $^{137}\text{Cs}$  uptake risk, were effectively identified using the combination of  $^{133}\text{Cs}$  addition at  $0.5 \text{ mg kg}^{-1}$  air-dried soil and pot cultivation. The Ex-ratios of added  $^{133}\text{Cs}$  and  $^{137}\text{Cs}$  after harvest also exhibited a strong linear correlation (Fig. 7). The  $\text{AdTF}^{133}\text{Cs}$  values were five times higher than  $\text{TF}^{137}\text{Cs}$  in 1 W/D (Fig. 6). In both Fukushima and Ibaraki Prefectures (the latter located south of Fukushima),  $\text{TF}^{137}\text{Cs}$  in paddy rice decreased by a factor of four from 2011 (the year of the FDNPP accident) to 2014 (Tagami et al., 2018). Since the soils used in this study were collected between 2015 and 2020, the  $\text{AdTF}^{133}\text{Cs}$  values appear to realistically represent  $\text{TF}^{137}\text{Cs}$  levels in 2011. These results strongly suggest that trace  $^{133}\text{Cs}$  addition effectively mimicked  $^{137}\text{Cs}$  pollution, regardless of the diverse clay mineralogy of the soils. The TF values of added  $^{133}\text{Cs}$  were always significantly lower in 30 W/D than in 1 W/D (Fig. 3), indicating that W/D cycles promoted the aging of  $^{133}\text{Cs}$ . However, this decrease was less pronounced for both  $^{133}\text{Cs}$  and  $^{137}\text{Cs}$  after harvest. Along with the  $\text{AdTF}^{133}\text{Cs}$  values (Fig. 3), the slopes in Fig. 6 further support the promotion of aging by W/D cycles. The slope was closer to 1 in 30 W/D than in 1 W/D. Nevertheless,  $\text{AdTF}$  in 30 W/D remained approximately three

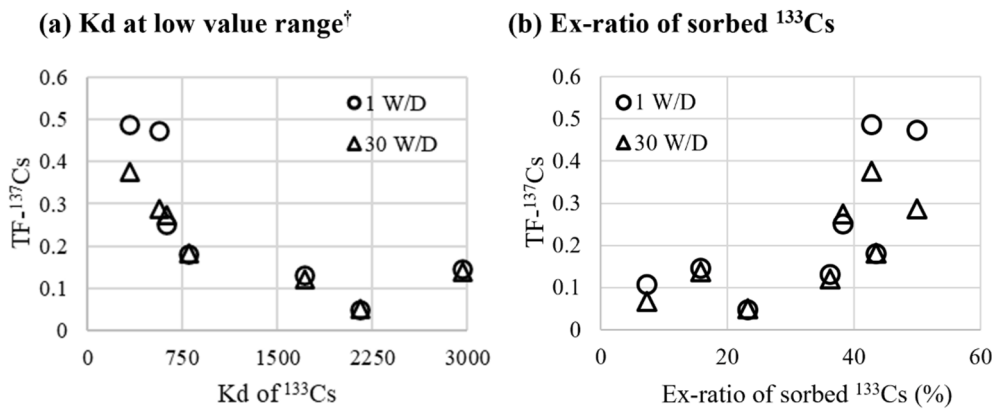
times higher than  $\text{TF}^{137}\text{Cs}$  (Fig. 6). To fully emulate the 4–9 years of aging in the soils used in this study, additional W/D cycles, prolonged incubation of  $^{133}\text{Cs}$ -amended soil (Takeda et al., 2013), or an extended pot cultivation experiment lasting multiple years would be required.

The  $K_d$  values of soils 6–8 at a  $^{133}\text{Cs}$  concentration of  $50 \mu\text{g L}^{-1}$  were slightly but significantly lower than those at  $25 \mu\text{g L}^{-1}$  in the batch experiment conducted at a soil-to-solution ratio of 1:10 (Fig. 1). Moreover, the  $K_d$  values at  $25 \mu\text{g L}^{-1}$  may have already been lower than those without  $^{133}\text{Cs}$  addition (Cremers et al., 1988). In this study, the amount of  $^{133}\text{Cs}$  addition was set at  $0.5 \text{ mg kg}^{-1}$  to account for a possible overestimation of  $^{137}\text{Cs}$  uptake risk in these soils, following a conservative approach to risk assessment. Compared to soils without  $^{133}\text{Cs}$  addition, the  $\text{TF}^{137}\text{Cs}$  values of  $^{133}\text{Cs}$ -amended soils increased, when significant, by a factor of  $<1.5$ , which is an acceptable level for screening purposes. Ideally, the amount of  $^{133}\text{Cs}$  addition should be determined for each soil based on batch experiment results. However, the  $^{133}\text{Cs}$  concentration in the batch experiment may be lower than the detection limit of ICP-MS when background  $^{133}\text{Cs}$  levels are high. For example, the  $^{133}\text{Cs}$  concentration in a  $0.01 \text{ M CaCl}_2$  solution without  $^{133}\text{Cs}$  addition was  $11 \text{ ng L}^{-1}$ , whereas the  $^{133}\text{Cs}$  content in rice shoots was sufficiently high to be detected even by ICP-MS with a high background  $^{133}\text{Cs}$  level (Table S6). If determining  $K_d$  values is challenging, the amount of  $^{133}\text{Cs}$  addition at  $0.5 \text{ mg kg}^{-1}$  soil should be adequate, at least for screening purposes.

Many studies have highlighted the importance of exchangeable potassium (Ex-K) in soil for the uptake of  $^{137}\text{Cs}$  by crops (Kato et al., 2015; Kondo et al., 2015; Kubo et al., 2015; Yamamura et al., 2018). In this study, both  $\text{TF}^{137}\text{Cs}$  and  $\text{AdTF}^{133}\text{Cs}$  exhibited a significant negative correlation with Ex-K. Due to the small pot size, soil Ex-K levels were severely depleted during cultivation (Ishikawa et al., 2017). Consequently, the  $\text{TF}^{137}\text{Cs}$  and  $\text{AdTF}^{133}\text{Cs}$  values represent the potential risk of  $^{137}\text{Cs}$  uptake in soils lacking adequate potassium fertilization, a common issue, particularly in tropical regions (Dobermann et al., 1996; Pathak et al., 2010). In the Fukushima area, increased potassium fertilization has been recommended to maintain an Ex-K content of at least  $250 \text{ mg K}_2\text{O kg}^{-1}$  during the paddy rice growing season (Fukushima Prefectural Government, 2014). The amount of potassium applied in the pot experiment corresponded to  $750 \text{ mg K}_2\text{O kg}^{-1}$ . However, post-harvest Ex-K levels were below  $100 \text{ mg K}_2\text{O kg}^{-1}$  in all but three soils in the 30 W/D treatment, except for soil 1 (Table S5). These results clearly indicate that larger pots or field cultivation are preferable for evaluating the effectiveness of K fertilization on  $^{137}\text{Cs}$  uptake by crops and for establishing target Ex-K levels to maintain  $^{137}\text{Cs}$  concentrations within acceptable limits. The Ex-K values after harvest were significantly lower in the 30 W/D treatment than in the 1 W/D treatment in soils 3, 5, 7, and 8 (Table S5), where inorganic N content had significantly increased during the 30 W/D period (Table S4). The increase in inorganic N likely promoted K uptake by rice roots, leading to lower Ex-K levels in the 30 W/D treatment compared to the 1 W/D treatment (Table S5). A decrease in Ex-K increases TF by reducing competition between K and Cs at root uptake sites and by facilitating the release of Cs once it has been fixed in the interlayers of 2:1-type clay minerals (Thiry et al., 2005; Kubo et al., 2015; Eguchi et al., 2015, 2021; Wakabayashi et al., 2020). In this study,  $\text{TF}^{137}\text{Cs}$  in both the 1 W/D and 30 W/D treatments, as well as  $\text{AdTF}^{133}\text{Cs}$  in the 30 W/D treatment, were significantly and negatively rank-correlated with Ex-K after harvest (Fig. S2). However,  $\text{AdTF}^{133}\text{Cs}$  in the 1 W/D treatment did not show a significant correlation with Ex-K (Fig. S2), suggesting that the availability of added  $^{133}\text{Cs}$  strongly influenced  $\text{AdTF}^{133}\text{Cs}$ . The TF values in the 30 W/D treatment may have been slightly overestimated due to an artifact effect on inorganic N content, which led to severe Ex-K depletion. However, the lower  $\text{AdTF}^{133}\text{Cs}$  in the 30 W/D treatment compared to the 1 W/D treatment suggests that the degree of overestimation is within an acceptable range, at least for screening risky soils.

In conclusion, the combination of trace  $^{133}\text{Cs}$  addition, W/D cycles,





**Fig. 8.** Relationship between the transfer factor of  $^{137}\text{Cs}$  in the pot experiment and (a) the Kd of  $^{133}\text{Cs}$  or (b) the exchangeable ratio of sorbed  $^{133}\text{Cs}$  in the batch experiment.

†Figure for the full Kd range is shown in the supplementary file (Fig. S3).

TF- $^{137}\text{Cs}$ , transfer factor of  $^{137}\text{Cs}$ ; Ex-, exchangeable; Kd, solid/liquid distribution coefficient; W/D, wetting/drying cycle.

(a) 1 W/D,  $\rho = -0.881^{**}$ ; 30 W/D,  $\rho = -0.881^{**}$ .

(b) 1 W/D,  $\rho = -0.762^{*}$ ; 30 W/D,  $\rho = -0.732^{*}$ .

\*,  $p < 0.05$ ; \*\*,  $p < 0.01$  in Spearman's rank correlation.

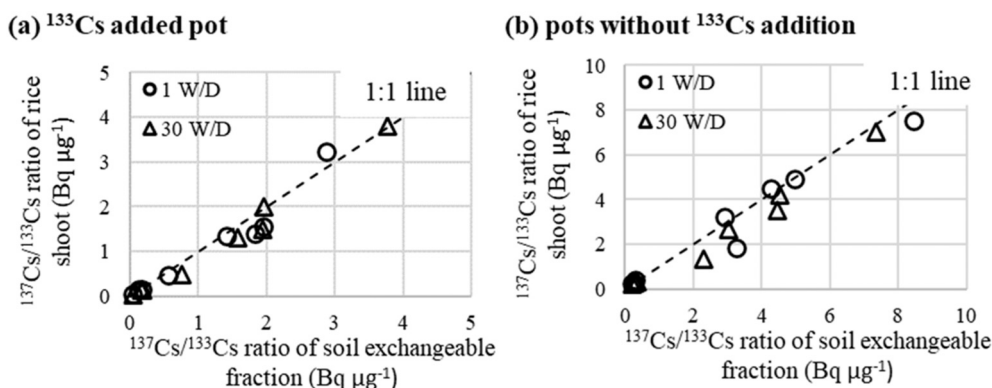
and pot cultivation is useful for evaluating the risk of  $^{137}\text{Cs}$  uptake by crops, particularly when  $^{137}\text{Cs}$  contamination levels are low.

#### 4.2. Use of indices from batch experiment for screening purpose

The Kd values of soils in the batch experiment were significantly and negatively rank-correlated with TF- $^{137}\text{Cs}$  in the pot experiment (Fig. 8). A Kd value of  $750 \text{ L kg}^{-1}$  appeared to be a threshold for increased TF. The Kd values of soils 6–8 were below this threshold, identifying them as risky soils based on field cultivation results for paddy rice or grass (Eguchi et al., 2021; Eguchi et al., 2023). Similarly, Takeda et al. (2014) reported a negative correlation between TF- $^{137}\text{Cs}$  values in soybeans and RIP values in Fukushima Prefecture. The Ministry of Agriculture, Forestry, and Fisheries (2014) classified soils with an RIP of  $5000 \text{ mmol kg}^{-1}$  or more as low-risk due to the low TF values observed in paddy rice grown in these soils. RIP determination is a type of Kd measurement that uses radioactive Cs to avoid underestimation caused by the saturation of Cs-selective sites, with strict control of the ionic conditions to exclude the influence of coexisting cations (Cremers et al., 1988; Wauters et al., 1996). In this study, Kd values were determined using  $^{133}\text{Cs}$ , acknowledging a slight underestimation of Kd, without strict ionic control, allowing the influence of coexisting cations to be reflected.

The Ex-ratio of sorbed  $^{133}\text{Cs}$  in the batch experiment was also significantly rank-correlated with TF (Fig. 8). Although the Ex-ratio of sorbed  $^{133}\text{Cs}$  in the batch experiment was significantly correlated with the Ex-ratio of added  $^{133}\text{Cs}$  before planting in the pot cultivation (Fig. S4), this correlation disappeared after harvest due to changes in the Ex-ratio during cultivation (Figs. 4 and S5). This result suggests that the pot experiment with trace  $^{133}\text{Cs}$  addition is preferable for a more precise evaluation of  $^{137}\text{Cs}$  uptake. Similar to the Ex-ratio of  $^{133}\text{Cs}$ , Kd values can also change during cultivation. Coexisting cations, particularly K, influence Kd (Wauters et al., 1996; Wakabayashi et al., 2020). Moreover, RIP values can also change during plant cultivation (Ogasawara et al., 2018). These studies suggest that Kd values fluctuate throughout crop cultivation. As discussed in the previous subsection, TF- $^{137}\text{Cs}$  likely indicates the potential risk of  $^{137}\text{Cs}$  uptake by crops, as Ex-K was severely depleted during cultivation due to the small pot size. If the pot size were larger and the initial Ex-K was adjusted to several levels through K fertilization, TF values in a scatter plot with Kd would align along the same Kd value.

In summary, the Kd value from the batch experiment may be useful for screening soils with a high potential risk of  $^{137}\text{Cs}$  uptake at low Ex-K levels. However, the combination of trace  $^{133}\text{Cs}$  addition and crop cultivation is preferable for a more precise risk evaluation.



**Fig. 9.** Relationship between the  $^{137}\text{Cs}/^{133}\text{Cs}$  ratio in rice shoots and the soil exchangeable fraction after harvest in pot cultivation: (a) with  $^{133}\text{Cs}$  addition and (b) without  $^{133}\text{Cs}$  addition.

TF, transfer factor; Ex-, exchangeable, W/D, wetting/drying cycle.

#### 4.3. Possible application of $^{137}\text{Cs}/^{133}\text{Cs}$ ratio

Plant roots absorb Cs from the soil solution (Zhu and Smolders, 2000), which mediates Cs uptake from the exchangeable fraction. Tsukada et al. (2023) reported that the  $^{137}\text{Cs}/^{133}\text{Cs}$  ratio did not differ significantly between the soil exchangeable fraction and field-grown potatoes in Fukushima, Aomori (northeastern Japan), and Chernobyl. Local studies have also suggested that the  $^{137}\text{Cs}/^{133}\text{Cs}$  ratio is similar between plant tissue and the soil exchangeable fraction, provided that  $^{133}\text{Cs}$  uptake from the subsoil is not significant (Ogasawara et al., 2018; Wakabayashi et al., 2020; Murakami et al., 2019). In this study,  $\text{ExTF-}^{133}\text{Cs}$  and  $\text{ExTF-}^{137}\text{Cs}$  exhibited similar values (Table S6), resulting in a comparable  $^{137}\text{Cs}/^{133}\text{Cs}$  ratio between rice shoots and the soil exchangeable fraction, regardless of  $^{133}\text{Cs}$  addition or W/D conditions (Fig. 9). This result, along with existing literature on specific activity ratios, strongly suggests that exchangeable  $^{137}\text{Cs}$  and exchangeable  $^{133}\text{Cs}$  were released into the soil solution at similar ratios and subsequently absorbed by plant roots, regardless of soil clay mineralogy and the degree of aging. This finding supports the conclusion that soil solution  $^{137}\text{Cs}$  concentration can be estimated by multiplying the soil solution  $^{133}\text{Cs}$  concentration by the  $^{137}\text{Cs}/^{133}\text{Cs}$  ratio in the exchangeable fraction. The primary factors determining  $^{137}\text{Cs}$  uptake are soil solution K concentration and  $^{137}\text{Cs}$  concentration (Zhu and Smolders, 2000). However, due to its low concentration, direct determination of soil solution  $^{137}\text{Cs}$  without  $^{137}\text{Cs}$  spiking is not practical for routine soil testing in risk evaluation. Compared to  $\text{TF-}^{137}\text{Cs}$ ,  $\text{ExTF-}^{137}\text{Cs}$  allows for a more precise assessment of  $^{137}\text{Cs}$  uptake risk in crops (Suzuki et al., 2022). Estimating soil solution  $^{137}\text{Cs}$  concentration based on soil solution  $^{133}\text{Cs}$  concentration and the exchangeable  $^{137}\text{Cs}/^{133}\text{Cs}$  ratio would significantly enhance the accuracy of risk evaluation.

#### 5. Conclusion

The combination of trace  $^{133}\text{Cs}$  addition, repeated wetting/drying cycles, and pot cultivation is useful for evaluating the  $^{137}\text{Cs}$  uptake risk in crops, particularly when  $^{137}\text{Cs}$  contamination levels are low.  $\text{AdTF-}^{133}\text{Cs}$  exhibited a strong positive correlation with  $\text{TF-}^{137}\text{Cs}$ . The Kd values and Ex-ratio of added  $^{133}\text{Cs}$  from the batch experiment successfully identified soils with a high  $^{137}\text{Cs}$  uptake risk in crops, suggesting that these indices could also be useful for evaluating  $^{137}\text{Cs}$  uptake risk, at least for screening high-risk soils. However, the repetition of 30 wetting/drying cycles was insufficient to replicate the 4–9 years of soil aging observed in this study. Further studies are needed to replicate long-term soil aging by implementing more wetting/drying cycles while allowing  $^{133}\text{Cs}$ -amended soil to age for several years or by conducting a long-term pot cultivation experiment. The similarity in the  $^{137}\text{Cs}/^{133}\text{Cs}$  ratio between the soil exchangeable fraction and rice shoots suggests the potential for estimating soil solution  $^{137}\text{Cs}$ , which would significantly enhance the evaluation of  $^{137}\text{Cs}$  uptake risk in crops.

#### CRediT authorship contribution statement

**Tetsuya Eguchi:** Writing – original draft, Visualization, Validation, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Hisaya Matsunami:** Investigation, Data curation. **Shigeto Fujimura:** Writing – review & editing, Methodology. **Adriana Nario:** Writing – review & editing, Validation, Supervision, Resources, Project administration, Methodology, Funding acquisition. **Poulette Blanc:** Validation, Methodology, Investigation. **Takuro Shinano:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization. **Gerd Dercon:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization.

#### Declaration of competing interest

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

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2025.179912>.

#### Data availability

Data will be made available on request.

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