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Effects of cattle manure compost application on crop growth and soil-to-crop transfer of cesium in a physically radionuclide-decontaminated field

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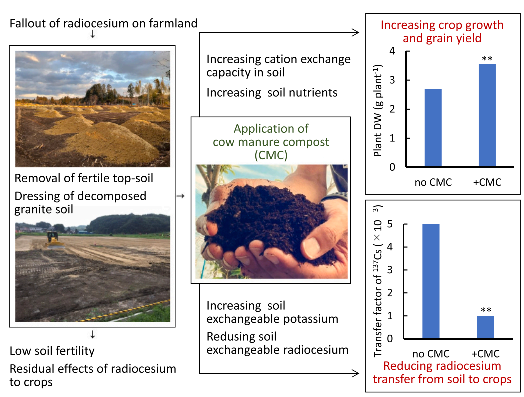
HIGHLIGHTS

- The decontaminated fields have problems with reduced soil fertility and radiocesium residues.
- Cattle manure compost application to the decontaminated fields increased the various soil nutrients.
- Cattle manure compost application enhanced crop growth and yield in the decontaminated fields.
- Cattle manure compost application reduced the transfer of radiocesium from soil to crop.
- The application of cattle manure compost promoted the fixation of radiocesium in the soil.

GRAPHICAL ABSTRACT



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ABSTRACT

Resuming crop production in physically decontaminated fields affected by radiocesium (^{134}Cs and ^{137}Cs) releases is crucial for restoring impacted areas. However, surface soil excavation to reduce radiocesium may lead to lower crop yield due to the loss of fertile topsoil. This study aimed to assess the effects of cattle manure compost (CMC) application on soil properties, crop growth, and ^{137}Cs soil-to-crop transfer in a physically decontaminated field and pot experiment. Field trials were conducted during 2018–2022, with CMC (1 and 2 kg m^{-2} year^{-1}) applied alongside conventional fertilization (CMC1 and CMC2 plots, respectively) in 2018–2019 and conventional

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fertilization alone in 2020–2022. Additionally, a pot experiment was used to evaluate the impact of CMC application in soil (1 kg m^{-2} $year^{-1}$ for 5 years) on ^{137}Cs transfer. In the field trial during 2018–2019, CMC1 and CMC2 plots exhibited higher soybean shoot dry weight (DW) compared with plots receiving conventional fertilization and additional K fertilizer (+K_2O). CMC application also improved soil nutrient content. The transfer factor of ^{137}Cs (TF-^{137}Cs: plant ^{137}Cs activity concentration/soil ^{137}Cs activity concentration) followed the order CMC2 < CMC1 ≈ +K_2O < conventional fertilization only (CF) and was negatively correlated with soil exchangeable K (Ex-K). During 2020–2022, when all plots received conventional fertilization alone, grain yields were higher in CMC1 and CMC2 plots than in the +K_2O plot, with the lowest TF-^{137}Cs in CMC2 plot followed by CMC1, +K_2O, and CF plots. The pot experiment confirmed that CMC soil had a lower TF-^{137}Cs and higher plant DW compared with CF soil with the same Ex-K level. Additionally, the soil exchangeable ^{137}Cs (Ex-^{137}Cs) level was significantly lower in CMC soil than CF soil. These findings demonstrate the potential of CMC application to improve crop growth and reduce ^{137}Cs transfer in physically decontaminated fields.

1. Introduction

The Fukushima Daiichi Nuclear Power Plant (FDNPP[1]) accident in 2011 resulted in the release of radioactive materials, impacting a wide area in eastern Japan. Certain municipalities with an annual radiation dose of 1–20 mSv were designated as "intensive contamination survey areas" by the Ministry of the Environment (MOE[2]) (MOE, 2019). Surveys and decontamination efforts were conducted in these areas based on municipality plans (MOE, 2019). By February 2018, decontamination of approximately 31,060 ha of farmland/meadows in the "intensive contamination survey areas" in Fukushima Prefecture and approximately 1,588 ha outside the prefecture had almost been completed (MOE, 2018). The more heavily affected areas, encompassing 11 municipalities in Fukushima Prefecture within a 20 km radius of FDNPP and areas with an annual dose exceeding 20 mSv, were designated as "special decontamination areas" and underwent partial decontamination (MOE, 2018; Fukushima Prefecture, 2021). Farmland decontamination in the "special decontamination areas" was completed in all designated municipalities (excluding areas where return was too difficult) by the end of March 2017, covering a total area of approximately 8,400 ha (MOE, 2019).

The decontamination process for farmland involved the removal of surface soil contaminated with radionuclides (approximately 5 cm in depth), followed by the application of uncontaminated soil and plowing to mix it with the original soil (approximately 0.15 m in depth) to reduce the concentration of radiocesium (^{134}Cs and ^{137}Cs) [Yukumoto, 2012; Ministry of Agriculture, Forestry and Fisheries (MAFF), 2013[3]]. However, physically decontaminated fields in the "special decontamination areas" tended to exhibit lower fertility compared with pre-decontamination conditions due to the removal of fertile topsoil and the use of low-fertility mountainous granite soil (Yoshino et al., 2015; Kubo et al., 2019; Inoue et al., 2020; Fukunaga-Sato et al., 2021; Kubo, 2021; Evrard et al., 2023). Although the radiocesium concentration in soil is reduced through surface soil removal, a certain amount of radiocesium remains, and the radiocesium transfer factor (TF: ratio of plant radiocesium activity concentration to soil radiocesium activity concentration) does not decrease solely through soil dressing (Hirayama et al., 2022). Therefore, the reduction of radiocesium transfer from soil to crops through potassium (K) application [reviewed by Fujii et al., 2021, among others] remains important in decontaminated fields within "intensive contamination survey areas".

Studies have shown that the application of cattle manure compost (CMC[4]) is beneficial for paddy rice (*Oryza sativa* L.) in decontaminated fields (Nishiwaki et al., 2017). Kubo et al. (2019) also reported that CMC application to decontaminated fields cultivated with buckwheat (*Fagopyrum esculentum* Moench) increased soil exchangeable K content (Ex-K[5]) and reduced RC transfer from soil to buckwheat. The high K content in CMC is considered a major factor influencing soil Ex-K and radiocesium transfer. CMC is rich in organic matter and enhances soil fertility by providing nutrient elements and improving physical properties through aggregate formation. However, the addition of organic polymers or humic substances to clay minerals can inhibit the strong adsorption of cesium, potentially increasing the mobility of radiocesium in soils with organic amendments (Dumat et al., 2000; Dumat and Staunton, 1999; Rigol et al., 2002). It is also important to consider the physical interactions and immobilization of ^{137}Cs as organic matter and minerals physically bond and form aggregates (Kögel-Knabner et al., 2008; Wagai et al., 2009). In soils with high organic matter content, ^{137}Cs preserved in aggregates may still be mobile and bioavailable (Koarashi et al., 2018).

To date, there is no literature available on the effects of applying CMC to decontaminated fields on (1) crop growth, (2) the transferability of ^{137}Cs from soil to crops, or (3) the dynamics of ^{137}Cs in the soil over multiple years. This research aims to clarify these issues and develop a sustainable method for restarting farming after decontamination. To achieve these objectives, field trials were performed to investigate the effects of two years of CMC application on the growth of crops and the dynamics of radiocesium, in addition to its sustainability over the subsequent three years. Additionally, we conducted a pot cultivation experiment using soil from the decontaminated field to provide a detailed analysis of the role of CMC in crop growth and radiocesium transfer, focusing on both ^{137}Cs and stable isotope cesium (^{133}Cs), which could support the analysis because of its similar kinetics in plants and soil to that of ^{137}Cs (Ogasawara et al., 2019). We believe that these efforts will support the resumption of farming and environmental recovery in areas affected by radiocesium. Owing to the lower concentration of ^{134}Cs because of relatively short half-life (2.1 years), only ^{137}Cs (the half-life is 30.1 years) was evaluated for radiocesium behavior in this study.

2. Material and methods

2.1. Field experiment

A field experiment was conducted during 2018–2022 in a local farmer's field in the coastal (Hamadori) area of Fukushima Prefecture, which underwent decontamination of radioactive materials through surface soil removal and a dressing of mountainous granite soil. The experiment involved the cultivation of soybean (*Glycine* max (L.) Merr.) in 2018–2020 and buckwheat in 2021–2022. Soybean was assessed as an important shifting crop, similar to buckwheat, with potentially higher radiocesium concentrations in grains compared with rice [International Atomic Energy Agency (IAEA), 2020[6]] and peanut (*Arachis*

[1] FDNPP: Fukushima Dai-ichi Nuclear Power Plant
[2] MOE: Ministry of the Environment
[3] MAFF: Ministry of Agriculture, Forestry and Fisheries
[4] CMC: cattle manure compost
[5] Ex-K: exchangeable potassium content in soil
[6] IAEA: International Atomic Energy Agency

hypogaea L.) (Kubo et al., 2021). The area was declared safe for farming after the lifting of the evacuation order in July 2016. In 2016–2017, prior to the experiment, the field was used to assess the transfer of ^{137}Cs from soil to various crops, including buckwheat, peanut, and upland rice, without K fertilization, although nitrogen (N) and phosphorus pentoxide (P_2O_5) were applied at 5 and 10 g m^{-2} through ammonium sulfate and superphosphate of lime, respectively. The soil in the field was classified as silandic Andosol, with a silty clay loam texture. The initial ^{137}Cs activity concentration in the soil was 3,100 ± 306 Bq kg^{-1} (mean ± standard error; n = 12) before the start of the experiment on May 22, 2018. Table 1 presents the soil chemical properties.

Four plots were established with different K fertilizer and CMC applications (Table 2). At the beginning of the experiment, there were no significant differences in soil Ex-K among the plots. The plot receiving conventional fertilization only (CF) served as the control plot. The +K_2O plot received increased K through chemical fertilizer (potassium sulfate) application to reduce ^{137}Cs soil-to-crop transfer in the first two years (2018–2019). The CMC1 and CMC2 plots received CMC treatments (1 and 2 kg m^{-2} $year^{-1}$) along with conventional fertilization in 2018–2019. Starting in 2020, the +K_2O, CMC1, and CMC2 plots transitioned to CF only to evaluate the sustainability of CMC effects. CMC contained 1.5 %, 2.4 %, and 2.2 % N, P_2O_5, and K_2O content, respectively, and did not contain radiocesium. N was applied at rates of 5 and 2 g m^{-2} for soybean and buckwheat, respectively, in the form of ammonium sulfate. P_2O_5 was applied at rates of 10 and 8 g m^{-2} for soybean and buckwheat, respectively, in the form of superphosphate of lime. The fertilizer and CMC were applied annually, excluding 2022, after the application of 100 g m^{-2} of magnesium lime for pH correction.

The soybean cultivar "Tachinagaha" was sown in 2018–2020, whereas the buckwheat cultivar "Nijiyutaka" was sown in 2021–2022, as per the request of local farmers. In soybean cultivation, the spacing between plants and rows was 15 and 75 cm, respectively, with two seeds sown per position. Sowing dates were May 29, May 30, and May 18 in 2018, 2019, and 2020, respectively. Buckwheat sowing took place on August 3 and July 27 in 2021 and 2022, respectively, with a sowing rate of 5 g m^{-2}. The row spacing was 75 and 25 cm in 2021 and 2022, respectively. Each plot covered an area of 22.5 m^2, and the experimental design followed a randomized block design with three replications.

Regarding soybean, aboveground shoots were sampled at the flowering stage (July 31, 2018, and August 7, 2019) and the maturity stage (October 27, 2020). In 2018–2019, mature grain samples could not be obtained due to severe insect damage. The sampling area comprised 0.34 m^2 (approximately five plants) at the flowering stage and 0.68 m^2 (approximately ten plants) at the maturity stage. During the flowering stage, shoots were washed with running tap water and oven-dried at 80 °C for up to 48 h. After measuring the dry weight (DW[7]), they were ground using a laboratory mill with stainless steel blades (WB-1; Osaka Chemical Co. Ltd.) for ^{137}Cs analysis. At the maturity stage, grain samples were obtained by threshing and passing through a 7.3 mm sieve for measuring grain weight and ^{137}Cs activity concentration. Regarding buckwheat, grain samples were taken from 4.5 and 1.5 m^2 areas of each plot in 2021 and 2022, respectively. Threshed samples were passed through a 4.5 mm sieve for measurement of grain weight and ^{137}Cs activity concentration, following the procedure described by Kubo et al. (2016).

Soil samples were collected from a 15 cm depth around the roots of the sampled plants using a worm scoop (Fujiwara Scientific Co., Ltd.) for each plot and sampling time. The samples were prepared according to the method described by Kubo et al. (2020) for determining ^{137}Cs activity concentration and chemical properties, including Ex-K. Chemical analyses of the soil were performed by Vegetech Co., Ltd. (Kanagawa, Japan). Available phosphorus (P) was measured using the Truog method (Truog, 1930). Available iron (Fe), manganese (Mn), zinc (Zn), and copper (Cu) were extracted using the diethylenetriaminepentaacetic acid method (Lindsay and Norvell, 1978) and measured via atomic absorption spectrophotometry. The ^{137}Cs in crops and soils were analyzed following methods similar to those described by Kubo et al. (2018). ^{137}Cs activity concentrations were decay-corrected for each sampling day.

Table 1
Soil chemical properties prior to the start of the field experiment.

		Mean	±	SE[a]
pH		5.6	±	0.0
EC[b]	(mS cm^{-1})	0.06	±	0.00
NH_4-N[c]	(mg kg^{-1})	10.5	±	0.8
NO_3-N[d]	(mg kg^{-1})	3.32	±	0.18
Available P[e]	(mg kg^{-1})	107	±	3
Ex-K[f]	(mg kg^{-1})	274	±	12
Ex-Ca[g]	(g kg^{-1})	1.24	±	0.01
Ex-Mg[h]	(mg kg^{-1})	422	±	16
CEC[i]	(cmolc kg^{-1})	19.1	±	0.1
Base saturation	(%)	54.2	±	0.5
Humus	(%)	3.78	±	0.07
Available Fe[j]	(mg kg^{-1})	108	±	5
Available Mn[k]	(mg kg^{-1})	25.5	±	0.9
Available Zn[l]	(mg kg^{-1})	0.79	±	0.03
Available Cu[m]	(mg kg^{-1})	1.33	±	0.05

[a] Standard error (n = 12).
[b] Electric conductivity.
[c] Ammonium nitrogen.
[d] Nitrate nitrogen.
[e] Available phosphate.
[f] Exchangeable potassium.
[g] Exchangeable calcium.
[h] Exchangeable magnesium.
[i] Cation exchange capacity.
[j] Available iron.
[k] Available manganese.
[l] Available zinc.
[m] Available copper.

2.2. Pot experiment

To evaluate the effect of CMC on ^{137}Cs uptake in more detail, a pot experiment was conducted using Komatsuna (*Brassica rapa* L. var. *perviridis*) cultivar "Rakuten" as a model plant, which has been shown to exhibit a high ^{137}Cs acquisition capability (Shinano et al., 2023) and rapid growth characteristics. Soil from the field experiment conducted in 2021 was collected for use in the pot experiment. In total, we collected 20 kg of soil from both the CF plot and the CMC plot receiving CMC application for five years (2016–2021) at 1 kg m^{-2} $year^{-1}$ with conventional fertilization from a 15 cm depth, and mixed the soil for the experiment. Supplementary Table 1 presents the chemical properties of the soil before the experiment.

Wagner pots (1 × 10^{-4} a) were used for cultivation, with each pot containing 1.6 kg of air-dried soil. The experiment was conducted in a controlled greenhouse at Hokkaido University in 2022, maintaining an air temperature of 25 °C. For fertilizer application, K was applied at rates of 0, 100, and 300 mg K_2O kg^{-1} (abbreviated as K0, K1, K2[8]) to both CF and CMC soils. The study design was completely randomized with three replicates. Ten seeds were sown on May 19, and after two weeks, only three plants were left per pot. The cultivation period lasted 7 weeks, from May 19 to July 7, 2022. Tap water (50–150 mL) was added to each pot daily based on plant growth.

After cultivation, plant shoots and soil samples were collected from each pot. Approximately 200 g of soil was taken from the middle of each pot using a scoop. Plant samples were washed, and their fresh weights were measured before drying at 80 °C for two days. The dried plant

[7] DW: dry weight
[8] K0, K1 and K2: pot applied K at rates of 0, 100, and 300 mg K_2O kg^{-1}, respectively.

Table 2
Amount of K_2O and cattle manure compost (CMC) applied to each plot in the field experiment.

Plot	2018 soybean		2019 soybean		2020 soybean		2021 buckwheat		2022 buckwheat	
	K_2O	CMC	K_2O	CMC	K_2O	CMC	K_2O	CMC	K_2O	CMC
	(g m^{-2})	(kg m^{-2})	(g m^{-2})	(kg m^{-2})	(g m^{-2})	(kg m^{-2})	(g m^{-2})	(kg m^{-2})	(g m^{-2})	(kg m^{-2})
CF	10	0	10	0	10	0	6	0	6	0
+K_2O	30	0	30	0	10	0	6	0	6	0
CMC1	10	1	10	1	10	0	6	0	6	0
CMC2	10	2	10	2	10	0	6	0	6	0

samples were then measured for DW and ground to a powder using a mortar and pestle. Soil samples were air-dried for seven days, crushed, and passed through a 2 mm sieve to remove plant roots or gravel.

The ^{137}Cs activity concentration and ^{133}Cs mass concentration were determined in the plant samples, with the ^{137}Cs activity concentration determined as described previously. For the measurement of the ^{133}Cs mass concentration, 0.05 g of plant samples was digested in 2.0 mL of 61 % nitric acid (HNO_3; electronic grade; KANTO CHEMICAL, Tokyo, Japan) using a DigiPREP apparatus (SCP SCIENCE, Quebec, Canada) at approximately 107.5 °C for around 2 h. When the solution was almost evaporated, 0.5 mL of hydrogen peroxide (H_2O_2; semiconductor grade; SANTOKU CHEMICAL INDUSTRIES, Tokyo, Japan) was added, and the samples were further heated at 107.5 °C for 20 min (Watanabe et al., 2022). The ^{133}Cs mass concentration was determined using inductively coupled plasma mass spectrometry (ICP–MS; ELAN DRC-e; PerkinElmer, Waltham, MA, USA).

Regarding soil Ex-K and water soluble K (Ws-K[9]), 2 g of soil was extracted using 20 mL of 1 M ammonium acetate (FUJIFILM WAKO JUNYAKU, Osaka, Japan) and ultra-pure water, respectively. The extractions were performed by shaking the samples for 1 h at 150 rpm and filtering them through ADVANTEC No.6 filter paper (ADVANTEC TOYO KAISHA, Tokyo, Japan). Ex-K and Ws-K were determined using atomic absorption spectrometry (AA-6200, SHIMADZU, Kyoto, Japan). To ensure analysis accuracy, external calibration standards containing K were measured after every 5 samples. Humus content was determined using the Kumata method (Hokkaido Research Organization Agricultural Department, 2012). Soil Cs (both ^{137}Cs and ^{133}Cs) was sequentially fractionated into three fractions: an exchangeable fraction extracted in 1 M ammonium acetate solution (Ex-Cs[10]), an exchangeable fraction after organic matter decomposition (Org-Cs[11]), and a residue fraction following the method of Tsukada et al. (2008). The total ^{133}Cs was decomposed using a mixture of 2.5 mL of hydrofluoric acid (Kanto Kagaku, Tokyo, Japan), 7 mL of HNO_3, and 0.5 mL of H_2O_2 with the Advanced Microwave Digestion Labstation (ETOHS1, MILESTONE GENERAL, Kanagawa, Japan) and determined using ICP–MS. Radiocesium interception potential (RIP[12]) was determined for both CF and CMC soils before fertilization using the method of Wauters et al. (1996). Six replications were used for measurements in both soils.

2.3. Calculation of ^{137}Cs and ^{133}Cs transferability and statistical analyses

The transfer factor (TF[13])s for ^{137}Cs and ^{133}Cs were calculated using the following formulas:

$$TF-{}^{137}Cs = \frac{{}^{137}Cs\ activity\ concentration\ in\ plant\ \left(Bq\ kg^{-1}\ DW\right)}{Total\ {}^{137}Cs\ activity\ concentration\ in\ soil\ \left(Bq\ kg^{-1}\ DW\right)}$$

$$TF-{}^{133}Cs = \frac{{}^{133}Cs\ mass\ concentration\ in\ plant\ \left(ng\ g^{-1}\ DW\right)}{Total\ {}^{133}Cs\ mass\ concentration\ in\ soil\ \left(ng\ g^{-1}\ DW\right)}$$

The data obtained were subjected to analysis of variance (ANOVA[14]) to evaluate the differences among treatments. Multiple comparisons after ANOVA were conducted using the Ryan–Einot–Gabriel–Welsch procedure. Additionally, multiple regression analyses using the stepwise method and Pearson correlation analyses were used to examine the relationships between traits. These statistical analyses were performed using IBM SPSS software for Windows (ver. 29; IBM Japan Ltd.). To assess differences between groups, analysis of covariance (ANCOVA[15]) was performed using R version 4.1.3 (R core team, 2022).

3. Results

3.1. Field experiment

3.1.1. Effects of CMC application on crop growth and grain yield

During the CMC application period (2018–2019), significant differences in shoot DW of soybean at the flowering stage were observed among plots (Table 3). The CMC2 plot, receiving conventional fertilizer and 2 kg m^{-2} of CMC, consistently exhibited the highest shoot DW in both years. The shoot DW of the CMC2 plot was approximately 1.2-fold larger than that of plots without CMC application (average of the CF and +K_2O plots). No significant differences were found between years, and no interaction effect between plot and year was observed.

After transitioning all plots to CF (2020–2022), grain yield was compared among plots for soybean (2020) and buckwheat

Table 3
Shoot dry weight (DW) and ^{137}Cs transfer factor (TF-^{137}Cs) from soil to shoot in soybean in the field experiment during the cattle manure compost (CMC) application period (2018–2019).

Plot	Shoot DW (g m^{-2})			TF-^{137}Cs		
	2018	2019	Mean	2018	2019	Mean
CF	301	323	312^{ab} [a]	0.052	0.060	0.056^{a}
+K_2O	264	275	269^{b}	0.041	0.038	0.039^{ab}
CMC1	334	320	327^{a}	0.045	0.040	0.042^{ab}
CMC2	365	328	347^{a}	0.039	0.023	0.032^{b}
Mean	316	312	314	0.044	0.040	0.042
ANOVA						
Plot (P)		* [b]			*	
Year (Y)		ns			ns	
P x Y		ns			ns	

The experimental design followed a randomized block design with three replications.
[a] Different lowercase letters indicate significant differences (Ryan–Einot–Gabriel–Welsch test; $P < 0.05$).
[b] * indicates a significant difference at $P < 0.05$; ns indicates "not significant."

[9] Ws-K: water soluble potassium content in soil
[10] Ex-Cs: exchangeable Cs (^{137}Cs and/or ^{133}Cs) concentration in soil extracted in 1 M
[11] Org-Cs: exchangeable Cs (^{137}Cs and/or ^{133}Cs) concentration after organic decomposition
[12] RIP: radiocesium interception potential
[13] TF: transfer factor
[14] ANOVA: analysis of variance
[15] ANCOVA: analysis of covariance

(2021–2022). In 2020, no significant differences in grain yield were observed among plots, although the highest values were recorded in the CMC2 plot (Table 4). There were no significant differences in 100-grain weight and the number of grains per area among plots. Notably, grain yield and shoot DW at maturity showed a significant positive correlation ($r = 0.643$, $P < 0.05$, $n = 12$; data not shown).

In 2021 and 2022, the grain yield of buckwheat significantly differed among plots, with the CMC2 plot exhibiting a higher yield compared with the $+K_2O$ plot (Table 5). The difference in grain yield between years was also significant. Although there were no significant differences in 100-grain weight among plots, significant differences were observed in the number of grains per area, both among plots and between years.

3.1.2. Effect of CMC application on ^{137}Cs transfer

During the CMC application period (2018–2019), the TF-^{137}Cs, calculated as shoot ^{137}Cs activity concentration divided by soil ^{137}Cs activity concentration, exhibited a significant difference among plots (Table 3). The CMC2 plot had the lowest TF-^{137}Cs, followed by the $+K_2O$ and CMC1 plots, with the CF plot exhibiting the highest value. Plots CMC2 and CMC1 showed approximately 43 % and 25 % reductions in ^{137}Cs soil to shoot transfer, respectively, compared with the CF plot (control). No significant differences were observed between years, and there was no interaction effect between plot and year.

After transitioning all plots to CF (2020–2022), the TF-^{137}Cs for soybean in 2020 was significantly different among plots (Table 4). The CF plot exhibited the highest value, and the $+K_2O$ and CMC2 plots showed significantly lower values compared with the CF plot. In 2021–2022, the TF-^{137}Cs from soil to buckwheat grain differed significantly among plots (Table 5). The $+K_2O$, CMC1, and CMC2 plots had significantly lower TF-^{137}Cs values compared with the CF plot, and the $+K_2O$ plot had a significantly higher value compared with the CMC2 plot. The interaction effect between plots and years was significant for TF, indicating that the effect of the relatively high TF-^{137}Cs in the CF plot in 2022 compared with 2021.

3.1.3. Relationship of soil chemical properties with crop growth and grain yield

The relationships between soil chemical properties and soybean shoot DW (2018–2019), soybean grain yield (2020), and buckwheat grain yield (2021–2022) are shown in Table 6. Regarding soybean, no significant relationships were found between soil chemical properties and shoot growth or grain yield, although CMC applications, particularly in the CMC2 plot, tended to result in higher growth and grain yield compared with plots without CMC application (CF and $+K_2O$ plots; Tables 3 and 4). Multiple regression analyses using the stepwise method did not reveal relationships between any soil properties and shoot DW or grain yield (data not shown). Similarly, no definitive relationships between soil properties and buckwheat grain yield were observed in common for both years, although grain yield showed significant relationships with soil exchangeable calcium (Ex-Ca[16]), cation exchange capacity (CEC[17]), and available Zn in 2021 and with base saturation in 2022. In 2021, Ca:Mg ratio also had significant positive correlation ($r = 0.639$, $P < 0.05$; data not shown). Significant differences among plots were observed for pH, available P, Ex-K, Ex-Ca, exchangeable magnesium (Ex-Mg[18]), CEC, base saturation, humus, available Fe, available Zn, and available Cu (Supplementary Fig. 1). All properties, except for available Fe, were highest in the CMC2 plot and tended to be high in the CMC1 plot compared with the CF and $+K_2O$ plots.

Table 4
Grain yield, 100-grain weight, number of grains, and ^{137}Cs transfer factor (TF-^{137}Cs) from soil to grain in soybean during the first year after the cessation of cattle manure compost (CMC) application (2020) in the field experiment.

Plot	Grain yield ($g\ m^{-2}$)	100 grains weight (g)	Number of grains (m^{-2})	TF-^{137}Cs
CF	296	30.9	958	0.021 ^{a} [a]
$+K_2O$	310	31.6	981	0.012 ^{bc}
CMC1	335	33.8	991	0.017 ^{ab}
CMC2	362	37.6	963	0.007 ^{c}
	ns [b]	ns	ns	**

The experimental design followed a randomized block design with three replications.

[a] Different lowercase letters indicate significant differences (Ryan–Einot–Gabriel–Welsch test; $P < 0.05$).

[b] ** indicate a significant difference at $P < 0.01$; ns indicates "not significant."

3.1.4. Relationship between soil chemical properties and ^{137}Cs transfer

TF-^{137}Cs exhibited a significant negative correlation with soil Ex-K in all years (Table 6; Fig. 1). Soil Ex-K was highest in the CMC2 plot, followed by the $+K_2O$ and CMC1 plots at similar levels, whereas the CF plot exhibited the lowest Ex-K (Table 7). The K_2O input during 2018–2019 was 10 g m^{-2} $year^{-1}$ (potassium sulfate) for CF, 30 g m^{-2} $year^{-1}$ (potassium sulfate) for $+K_2O$, 10 g m^{-2} $year^{-1}$ (potassium sulfate) + 22 g m^{-2} $year^{-1}$ (CMC) for CMC1, and 10 g m^{-2} $year^{-1}$ (potassium sulfate) + 44 g m^{-2} $year^{-1}$ (CMC) for CMC2. Soil Ex-K was higher in plots with higher K_2O inputs (Table 7), and TF-^{137}Cs was lower in plots with higher soil Ex-K (Tables 3–5). Although the difference was not significant, ratio of soil Ex-^{137}Cs and soil Ex-K (soil Ex-^{137}Cs/soil Ex-K) tended to be low in CMC1 plot (0.58 ± 0.06 Bq mg^{-1}) compared with $+K_2O$ plot (0.69 ± 0.08 Bq mg^{-1}), which was applied almost the same amount of K, in the field experiments (data not shown).

3.2. Pot experiment

3.2.1. Plant growth and Cs uptake by plants

The plant DW, TF-^{137}Cs, and TF-^{133}Cs in the pot experiment are presented in Table 8. There were no significant differences in DW among K levels. However, the DW in CMC soil was significantly greater than that in CF soil, consistent with the findings for soybean and buckwheat in the field experiment. TF-^{137}Cs and TF-^{133}Cs were lower in CMC soil than in CF soil, and they decreased in the order of K2 (+ 300 mg K_2O kg), K1 (+ 100 mg K_2O kg), and K0 (+ 0 mg K_2O kg), irrespective of CF or CMC soils. TF-^{137}Cs and TF-^{133}Cs were significantly positively correlated, with TF-^{137}Cs being higher than TF-^{133}Cs (Fig. 2). The Ex-^{137}Cs ratio (Ex-^{137}Cs/total ^{137}Cs) and the Ex-^{133}Cs ratio (Ex-^{133}Cs/total ^{133}Cs) also had a similar relationship with TF-^{137}Cs and TF-^{133}Cs. The mineral concentrations of plants are provided in Supplementary Table 2. CMC treatment affected various mineral concentrations in plants, including aluminum (Al), boron, calcium (Ca), cadmium, cobalt, Cu, K, Mn, molybdenum, sodium (Na), rubidium, sulfur (S), and strontium (Sr), in addition to ^{133}Cs. Concentrations of barium (Ba), Cs, P, S, Sr, and Zn in plants were significantly influenced by K levels.

3.2.2. Soil chemical properties after harvest

At harvest, total ^{137}Cs activity concentration and ^{133}Cs mass concentration in soil did not differ significantly, irrespective of CF or CMC soil and K levels (Table 9). However, Ex-^{137}Cs and Ex-^{133}Cs were significantly lower in CMC soil compared with CF soil and showed a decreasing trend with increasing K levels. Org-^{137}Cs showed no significant difference between CMC and CF soil, whereas Org-^{133}Cs was significantly lower in CMC soil compared with CF soil. Ex-K and Ws-K were higher in CMC soil compared with CF soil. The concentration of Ex-K was approximately fivefold higher than that of Ws-K. Other soil chemical properties are shown in Supplementary Table 3. The humus content in CMC soil was higher than that in CF soil.

[16] Ex-Ca: exchangeable calcium content in soil

[17] CEC: cation exchange capacity

[18] Ex-Mg: exchangeable magnesium content in soil

Table 5
Grain yield, 100-grain weight, number of grains, and ^{137}Cs transfer factor (TF-^{137}Cs) from soil to grain in buckwheat during the second and third years after the cessation of cattle manure compost (CMC) application (2021–2022) in the field experiment.

Plot	Grain yield (g m^{-2})			100 grains weight (g)			Number of grains (m^{-2})			TF-^{137}Cs		
	2021	2022	Mean	2021	2022	Mean	2021	2022	Mean	2021	2022	Mean
CF	119	244	182 ^{ab} [a]	3.64	3.69	3.67	3281	6614	4948 ^{ab}	0.0041	0.0061	0.0051 ^{a}
+K_2O	90	231	160 ^{b}	3.71	3.71	3.71	2434	6215	4324 ^{b}	0.0038	0.0032	0.0035 ^{b}
CMC1	120	246	183 ^{ab}	3.65	3.72	3.69	3282	6612	4947 ^{ab}	0.0034	0.0034	0.0034 ^{bc}
CMC2	145	269	207 ^{a}	3.65	3.72	3.68	3980	7227	5604 ^{a}	0.0024	0.0026	0.0025 ^{c}
Mean	119	248	183	3.66	3.71	3.69	3244	6737	4956	0.0034	0.0038	0.0036
ANOVA												
Plot (P)		* [b]			ns			*			**	
Year (Y)		**			ns			**			ns	
P x Y		ns			ns			ns			*	

The experimental design followed a randomized block design with three replications.
[a] Different lowercase letters indicate significant differences (Ryan–Einot–Gabriel–Welsch test; $P < 0.05$).
[b] * and ** indicate significant differences at $P < 0.05$ and $P < 0.01$, respectively; ns indicate "not significant."

Table 6
Correlation coefficients of the relationships between soil chemical properties and shoot dry weight (DW), grain yield, and ^{137}Cs transfer factor (TF-^{137}Cs) from soil to shoot/grain in the field experiment.

	Shoot DW		Grain yield			TF-^{137}Cs				
	2018	2019	2020	2021	2022	2018	2019	2020	2021	2022
pH	0.073	−0.014	0.202	0.411	0.484	0.077	−0.543	−0.698 * [a]	−0.691 *	−0.090
EC	−0.288	−0.295	−0.078	0.066	−0.387	0.309	−0.513	−0.530	−0.349	−0.230
NH_4-N	−0.076	0.388	−0.039	0.288	0.133	0.119	−0.157	−0.381	−0.177	0.096
NO_3-N	−0.069	0.118	−0.024	0.224	0.093	−0.055	−0.232	0.388	−0.578 *	−0.377
Available P	−0.408	0.251	0.167	0.539	0.395	0.303	−0.355	−0.723 **	−0.659 *	−0.502
Ex-K	0.369	−0.126	0.113	0.456	0.031	−0.605 *	−0.705 *	−0.748 **	−0.731 **	−0.590 *
Ex-Ca	0.283	0.031	0.091	0.589 *	0.377	0.276	−0.413	−0.631 *	−0.642 *	−0.218
Ex-Mg	0.397	0.376	0.096	0.309	0.199	0.222	−0.398	−0.355	−0.489	0.300
CEC	0.300	−0.131	0.099	0.651 *	−0.213	−0.278	−0.285	−0.522	−0.389	0.080
Base saturation	0.222	0.203	0.095	0.243	0.664 *	0.275	−0.501	−0.609 *	−0.624 *	−0.324
Humus	−0.261	−0.041	0.107	0.163	−0.093	−0.378	−0.225	−0.529	−0.146	−0.339
Available Fe	−0.352	−0.067	0.305	−0.059	−0.289	−0.556	−0.055	−0.617 *	−0.014	0.174
Available Mn	0.319	0.326	−0.151	−0.573	−0.055	−0.153	−0.135	0.601 *	0.209	−0.144
Available Zn	0.346	0.275	−0.047	0.633 *	0.396	−0.132	−0.624 *	−0.580 *	−0.745 **	−0.643 *
Available Cu	−0.386	0.300	0.177	0.391	0.082	−0.398	−0.739 **	−0.724 **	−0.629 *	−0.639 *

[a] * and ** indicate significant differences at $P < 0.05$ and $P < 0.01$, respectively (N = 12).

3.2.3. Effect of CMC on Cs transferability from soil to plant

The negative relationships of Ex-K and Ws-K with TF-^{137}Cs and TF-^{133}Cs (Fig. 3), were consistent with the field experiments. Additionally, the relationship between Ex-K and TF-^{137}Cs or TF-^{133}Cs differed significantly between CF- and CMC-treated soils. Such a relationship was also observed between Ws-K and TFs. ANCOVA was performed to analyze the difference between CMC and CF, after log-transforming TF-^{137}Cs and TF-^{133}Cs (data not shown), and significant differences were found between the regression lines of CF and CMC in terms of the relationships between Ex-K and TF-^{137}Cs, Ws-K and TF-^{137}Cs, and Ws-K and TF-^{133}Cs ($P < 0.001$), as well as the relationship between Ex-K and TF-^{133}Cs ($P < 0.01$).

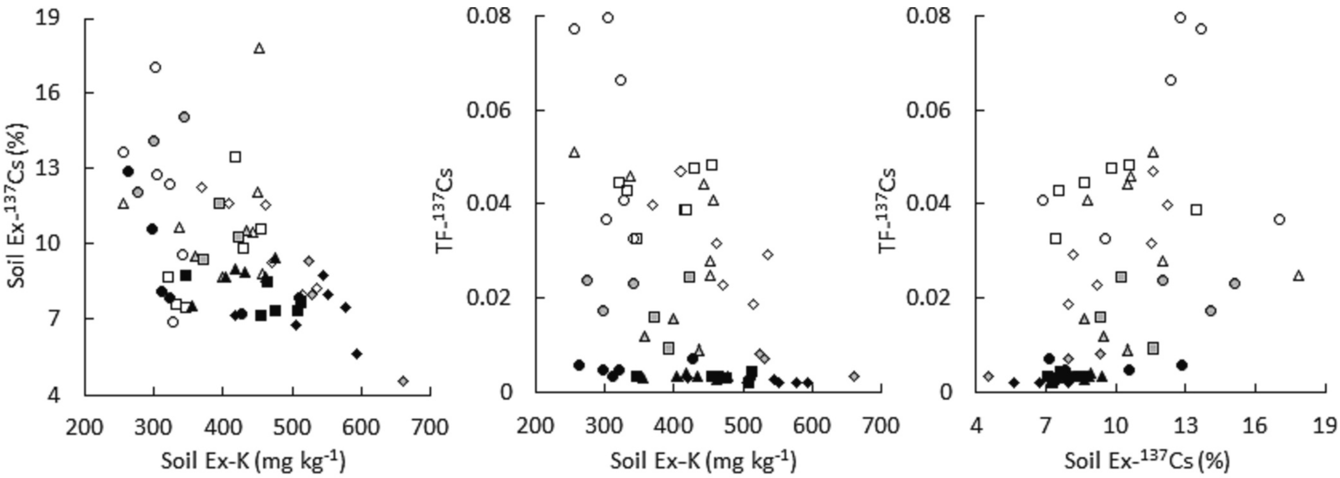
The relationships between Ex-K and the presence ratio of different Cs forms are depicted in Fig. 4. Increasing soil Ex-K led to decreased Ex-^{137}Cs, Ex-^{133}Cs, Org-^{137}Cs, and Org-^{133}Cs levels with increased residual ^{137}Cs and residual ^{133}Cs. Based on ANCOVA, the relationship between Ex-K and the Ex-Cs ratio differed significantly between CF and CMC, regardless of ^{137}Cs or ^{133}Cs ($P < 0.05$ and $P < 0.01$, respectively; data not shown). No significant difference was observed between Ex-K and the Org-^{137}Cs ratio between CMC and CF according to ANCOVA. Regarding the relationship between Ex-K and the residual-^{137}Cs ratio, there was no significant difference between CMC and CF based on ANCOVA, but a significant difference was observed for the relationship between Ex-K and the residual ^{133}Cs ratio ($P < 0.01$).

4. Discussion

This study used field and pot experiments to test the hypothesis that the application of CMC to decontaminated fields can improve crop productivity as well as reduce the soil-to-crop transfer of ^{137}Cs. The hypothesis was verified by these experiments and the application of CMC was considered to be effective when farming is resumed in a decontaminated field under the conditions implemented. This section discusses the effects of CMC application on crop growth, crop yield, and the dynamics of ^{137}Cs in soil and its transfer to crops.

4.1. Crop growth and grain yield

In the 2018–2019 field experiment, the shoot DW of soybean significantly increased with CMC application compared with no CMC application (Table 3). However, grain yield was not obtained owing to severe insect damage in both years. Previous studies have shown that increased total dry matter production generally leads to increased grain yield (Yoshida, 1972). Regarding soybean, Kakiuchi (2013) reported a significant correlation between the DWs of stems and leaves and grain weight during the maturity stage. Similarly, Saito and Goto (1991) found a positive correlation between shoot DW at the flowering stage and grain yield in the soybean cultivar "Tachiyutaka" up to a shoot DW of 345 g m^{-2}. Beyond this threshold, grain yield slightly declined. In our experiments, mean (± standard error) shoot DW at the flowering stage was 316 ± 15 and 312 ± 11 g m^{-2} in 2018 and 2019, respectively. Although the grain yield in each plot was not evaluated, the larger shoot



	Soybean shoot 2018-2019	Soybean grain 2020	Buckwhaet grain 2021-2022
Soil Ex-K - Soil Ex-^{137}Cs	-0.188	-0.820 (P<0.01)	-0.600 (P<0.01)
Soil Ex-K - TF-^{137}Cs	-0.651 (P<0.01)	-0.748 (P<0.01)	-0.629 (P<0.01)
Soil Ex-^{137}Cs - TF-^{137}Cs	0.201	0.671 (P<0.05)	0.474 (P<0.05)

Fig. 1. Relationships among soil exchangeable (Ex)-K, soil Ex-^{137}Cs, and ^{137}Cs transfer factor (TF-^{137}Cs) from soil to shoot/grain.
White, gray, and black plots indicate soybean shoot (N = 24), soybean grain (N = 12), and buckwheat grain (N = 24), respectively.
Circles, triangles, squares, and diamonds indicate the CF, +K_2O, CMC1, and CMC2 plots, respectively.
The table below the figures show the correlation coefficient for each relationship.

Table 7
Soil exchangeable (Ex)-K and Ex-^{137}Cs ratio of each plot during the field experiment.

Plot	Soil Ex-K (mg kg^{-1})						Soil Ex-^{137}Cs ratio (%)					
	2018	2019	2020	2021	2022	Mean	2018	2019	2020	2021	2022	Mean
CF	296	323	306	373	337	327^{c} [a]	12.52	11.57	13.75	8.84	9.29	11.20^{a}
+K_2O	349	450	398	442	407	409^{b}	13.37	10.43	9.56	9.08	8.28	10.15^{ab}
CMC1	369	398	396	494	425	417^{b}	8.94	10.26	10.41	7.79	7.71	9.02^{bc}
CMC2	413	507	572	572	492	511^{a}	11.81	8.47	7.27	7.27	7.26	8.42^{c}
Mean	357^{B} [b]	420^{A}	418^{AB}	471^{A}	415^{AB}		11.66^{A}	10.18^{A}	10.25^{A}	8.25^{B}	8.14^{B}	
ANOVA												
Plot (P)			** [c]						**			
Year (Y)			**						**			
P x Y			ns						ns			

The experimental design followed a randomized block design with three replications.
[a] Different lowercase letters indicate significant differences among plots (Ryan–Einot–Gabriel–Welsch test; P < 0.05).
[b] Different uppercase letters indicate significant differences among years (Ryan–Einot–Gabriel–Welsch test; P < 0.05).
[c] ** indicates a significant difference at P < 0.01; ns indicates "not significant."

Table 8
Plant dry weight (DW) and ^{137}Cs and ^{133}Cs transfer factors (TF-^{137}Cs and TF-^{133}Cs, respectively) in the pot experiment.

	Plant DW (g plant^{-1})			TF-^{137}Cs			TF-^{133}Cs		
	CF	CMC	Mean	CF	CMC	Mean	CF	CMC	Mean
K0 [a]	2.63	3.69	3.16	0.162	0.038	0.100	0.064	0.015	0.040
K1	2.85	3.80	3.33	0.107	0.024	0.066	0.042	0.009	0.026
K2	2.61	3.20	2.91	0.062	0.018	0.040	0.022	0.008	0.015
Mean	2.70	3.56		0.110	0.027		0.043	0.011	
ANOVA									
CF/CMC (T)		** [b]			**			**	
K		ns			ns (P = 0.071)			ns (P = 0.095)	
T x K		ns			ns			ns	

The experimental design was complete randomization with three replications.
[a] K0, K1 and K2 indicate the application of potassium fertilizer at 0, 100, and 300 mg K_2O kg^{-1}, respectively.
[b] * and ** indicate significant differences at P < 0.05 and P < 0.01, respectively; ns indicate "not significant."

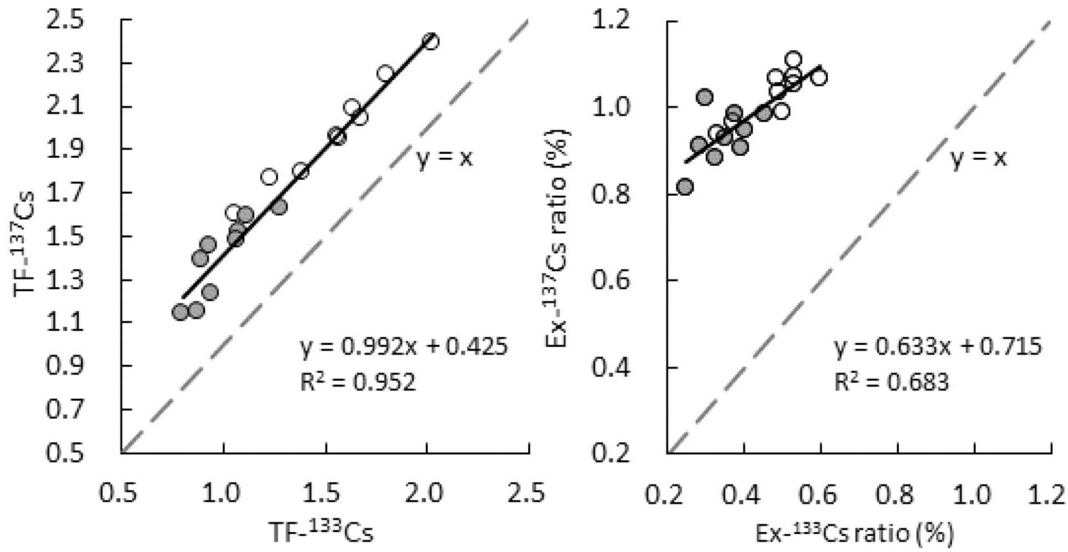


Fig. 2. Relationship between ^{137}Cs and ^{133}Cs transfer factors (TF-^{137}Cs and TF-^{133}Cs, respectively) from soil to plant (left) and between Ex-^{137}Cs ratio to total ^{137}Cs and Ex-^{133}Cs ratio to total ^{133}Cs in soil (right) in the pot experiment.
Open and closed circles indicate CF and CMC, respectively ($N = 9$).
Values on each axe was logarithmically converted from TF-^{137}Cs ($\times 10^3$), TF-^{133}Cs ($\times 10^3$), Ex-^{137}Cs ratio and Ex-^{133}Cs ratio, respectively.

biomass observed in plots with CMC application suggests a potential contribution to higher grain yield.

After switching all plots to CF, the grain yield of soybean in 2020 tended to be higher in plots CMC1 and CMC2, although not significantly higher (Table 4). The shoot DW of soybean was largest in the CMC2 plot (data not shown) and significantly positively correlated with grain yield. The grain yield of buckwheat in 2021–2022 was higher in the CMC2 plot than in the $+K_2O$ plot (Table 5). The difference in grain yield among buckwheat plots was similar to the difference in the number of grains per area (Table 5), indicating that the latter contributed to grain yield. The variation in buckwheat grain yield between years (Table 5) may be partly attributed to the difference in row spacing between years (75 and 25 cm in 2021 and 2022, respectively).

Soil analysis in each plot revealed that CMC application increased the concentrations of various chemical properties (Supplementary Fig. 1). The grain yield of buckwheat was significantly positively correlated with soil Ex-Ca, CEC, and available Zn in 2021 and with base saturation in 2022 (Table 6). This suggests that CMC application supplied nutrients and increased CEC, contributing to improved soil nutrient content. Similar effects of CMC on soil fertility have been reported in soybean (Moreira et al., 2015) and red chili (Situmeang et al., 2021) cultivation. In the present study, CMC application also increased humus content in the soil (Supplementary Fig. 1; Supplementary Tables 1 and 3). Humus has a negative charge (Ellerbrock and Gerke, 2013), allowing it to attract and adsorb positively charged nutrients, thereby protecting them from leaching. These effects may be associated with the higher soybean and buckwheat grain yields observed in our study. Miura et al. (2017) found that the relative deficiency of soil Ex-Mg caused by high soil Ex-K levels leads to reduced soybean yield, with variations according to year, in nonallophanic Andosol fields in the Tohoku region in Japan. Takamoto et al. (2021) showed that a low Ca:Mg ratio could potentially limit soybean growth. These results suggest that the content and balance of Ex-K, Ex-Ca, and Ex-Mg may have an influence on crop growth. In the present, the buckwheat grain yield showed relatively large variation in 2021, with the lowest yield in the $+K_2O$ plot (Table 5). The grain yield in 2021 was significantly correlated with Ex-Ca (Table 6) and the Ca:Mg ratio, indicating that CMC application may prevent deficiencies in soil Ex-Ca and Ex-Mg compared with the use of chemical potassium fertilizer alone. Additionally, the increased shoot DW and grain yield observed with CMC application may be influenced by other factors, such as soil mechanical improvement, although this was not investigated in our study.

In the pot experiment, the DW of Komatsuna in CMC soil was significantly larger than that in CF soil (Table 8), consistent with the results for soybean and buckwheat in the field experiments. CMC soil showed higher concentrations of Ex-K, Ex-Ca, and Ex-Mg before the experiment (Supplementary Table 1). Although we could not investigate the chemical properties of topsoil before stripping and dressing soil, Yoshino et al. (2015), Inoue et al. (2020), and Fujii et al. (2021) have reported that the post-decontamination fields have lower fertility because (1) the fertile topsoil was removed and (2) the mountainous soil around the area, which was primarily weathered granite and/or soil containing a large amount of sand and therefore had poor fertility, was used as the dressing soil. In surveys at multiple locations conducted before and after decontamination, it was reported that the total carbon and total nitrogen contents in the soil after decontamination in the fields were 64 % and 62 % of the pre-decontamination levels, respectively (NARO, 2021), which indicated that the fertility of the soil had largely decreased. These results confirm that CMC application to physically decontaminated fields contribute in increasing soil fertility and promoting the growth of various crops. As described above, this study revealed an improvement in crop growth and yield, and determined the sustainability of the improvement resulting from CMC application to fields after the decontamination of ^{137}Cs.

4.2. Cs behavior in the soil and Cs soil-to-crop transfer

4.2.1. TF-^{137}Cs and TF-^{133}Cs from soil to crops

The TF-^{137}Cs values in the field experiments were 0.007–0.021 (2020) for soybean and 0.0034–0.0061 (2021 and 2022) for buckwheat (Tables 4 and 5). Previous studies conducted in the central (Nakadori) area of Fukushima Prefecture reported higher TF values for soybean (0.033–0.049 in 2019; 0.022–0.033 in 2020; Kubo et al., 2021). The soil Ex-K, which was negatively associated with TF, was 167–227 mg kg^{-1} in soybean cultivation in 2019 and 2020. For buckwheat, TF values were 0.0050–0.0150 in 2014, 0.0025–0.0083 in 2015, and 0.0032–0.0134 in 2016, and soil Ex-K content was 111–389, 137–442, and 154–339 mg kg^{-1} in these years, respectively (Kubo et al., 2019). The soil Ex-K values in the present study were relatively higher than those reported in previous studies (Table 7), which may explain why the observed TF-^{137}Cs

Table 9
Soil chemical properties after cultivation under different K-, ^{137}Cs-, and ^{133}Cs-related conditions in the pot experiment.

	Total ^{137}Cs (Bq kg^{-1})			Ex-^{137}Cs (Bq kg^{-1})			Org-^{137}Cs (Bq kg^{-1})			Total ^{133}Cs (ng g^{-1})			Ex-^{133}Cs (ng g^{-1})			Org-^{133}Cs (ng g^{-1})			Soil Ex-K (mg kg^{-1})			Soil Ws-K (mg kg^{-1})		
	CF	CMC	Mean	CF	CMC	Mean	CF	CMC	Mean	CF	CMC	Mean	CF	CMC	Mean	CF	CMC	Mean	CF	CMC	Mean	CF	CMC	Mean
K0 [a]	2321	2520	2421	272	222	247	185	176	181 [a] [b]	1174	1081	1128	37.6	28.3	33.0 [a]	23.1	14.1	18.6 [a]	173	287	230 [a]	24.6	47.3	36.0 [a]
K1	2353	2457	2405	272	234	253	183	169	176 [a]	1102	1067	1085	39.6	23.6	31.6 [a]	19.0	12.1	15.6 [b]	238	382	310 [b]	43.7	67.3	55.5 [b]
K2	2459	2727	2593	227	201	214	136	168	152 [b]	1199	1086	1143	30.1	21.1	25.6 [b]	12.5	12.9	12.7 [c]	445	521	483 [c]	108.3	116.3	112.3 [c]
Mean	2378	2568		257	219		168	171		1158	1078		35.8	24.3		18.2	13.0		285	397		58.9	77.0	
ANOVA																								
CF/CMC (T)		ns [c]			*			ns			ns			**			**			**			*	
K		ns			ns			**			ns			**			**			**			**	
T x K		ns			ns			*			ns			*			**			ns			ns	

The experimental design was complete randomization with three replications.
[a] K0, K1 and K2 indicate the application of potassium fertilizer at 0, 100, and 300 mg K_2O kg^{-1}, respectively.
[b] Different lowercase letters indicate significant differences among K levels (Ryan–Einot–Gabriel–Welsch test; $P < 0.05$).
[c] * and ** indicate significant differences at $P < 0.05$ and $P < 0.01$, respectively; ns indicate "not significant."

values were similar to or lower than those reported by Kubo et al. (2019, 2021).

In the pot experiment, TF-^{137}Cs was approximately 2.5-fold higher than TF-^{133}Cs (Fig. 2), indicating that plant availability of ^{137}Cs were higher than that of ^{133}Cs in the soil. Similar trends were observed in previous studies, although the specific soil and plant species varied. For example, Ogasawara et al. (2019) reported a TF-^{137}Cs to TF-^{133}Cs ratio of 2.3 in rice using eight soil types from Fukushima Prefecture. Tsukada et al. (2002) reported a TF-^{137}Cs to TF-^{133}Cs ratio of 2.6 in rice and soils from 20 paddy fields in Aomori, Japan, which was based on ^{137}Cs from the global fallout effect (Hirose et al., 1987). These differences in TF values between ^{137}Cs and ^{133}Cs may be attributed to differences in their forms in the soil. Ex-Cs and Org-Cs fractions are more readily absorbed by plants (Puhakainen et al., 2001; Hou et al., 2003), and the ratio of these fractions to the total fraction was higher for ^{137}Cs (15 %–20 %) than for ^{133}Cs (4 %–8 %) (Table 9, Fig. 2). The residual fraction accounted for the majority of both ^{137}Cs (80 %–85 %) and ^{133}Cs (92 %–96 %). The higher ratio of Ex-^{137}Cs and Org-^{137}Cs compared with Ex-^{133}Cs and Org-^{133}Cs likely contributes to the higher TF-^{137}Cs values. These results suggest that ^{133}Cs is incorporated into more structurally bound compounds in soil minerals compared with ^{137}Cs, and even after several decades, ^{137}Cs does not undergo incorporation into the structural fraction.

4.2.2. *Influence of soil Ex-K on Cs behavior in the soil and soil-to-crop transfer*

TF-^{137}Cs values differed significantly among plots in all years, and plots with higher soil Ex-K content exhibited lower TF-^{137}Cs values in the field experiments (Tables 3–5 and 7). Indeed, there was a significant negative correlation between soil Ex-K and TF-^{137}Cs (Table 6; Fig. 1). In the pot experiment, TF-^{137}Cs and TF-^{133}Cs were significantly lower at higher K levels (Table 8). The CMC used in our study contained 2.2 % K_2O, and TF-^{137}Cs and TF-^{133}Cs values were lower in CMC soils compared with CF soils in both field and pot experiments (Tables 3–5 and 8). Kato et al. (2015) also reported that the application of manure could effectively reduce the radiocesium uptake in rice as a result of the K content. Overall, these results suggest that CMC can supply K_2O to the soil and reduce soil-to-crop Cs transfer. This study also showed that two years of CMC application reduced the soil-to-crop transfer of ^{137}Cs, and the effect persisted for at least three years thereafter.

One factor contributing to soil-to-crop Cs migration with increasing soil Ex-K content is reduction of Ex-^{137}Cs in the soil (Kubo et al., 2015, 2017). The amount of Ex-^{137}Cs is an important factor to explain the transfer of radiocesium from soil to crops (Suzuki et al., 2023). Wakabayashi et al. (2022) showed that Ex-^{137}Cs decreased with an increase in non-exchangeable K content extracted by the mild tetraphenyl-boron, which reflects fertilizer K fixed by the minerals, and indicated that ^{137}Cs was fixed together with K. Kondo et al. (2015) demonstrated that rice Cs uptake decreased with higher soil Ex-K content and correlated linearly with the exchangeable Cs:K ratio in the soil. In the present study, Ex-^{137}Cs content in the soil tended to be lower in plots with higher K_2O inputs from CMC and potassium sulfate (Table 7). The soil Ex-^{137}Cs ratio was significantly negatively correlated with soil Ex-K and significantly positively correlated with TF-^{137}Cs from 2020 to 2022 (Fig. 1). Similar trends were observed in the pot experiment, where increasing soil Ex-K content resulted in decreased Ex-^{137}Cs, Ex–^{133}Cs, Org-^{137}Cs, and Org-^{133}Cs ratios, as well as increased residual^{137}Cs and ^{133}Cs ratios (Fig. 4). These results are consistent with previous studies indicating that decreased Ex-K content can lead to the release of Cs fixed at frayed edge sites of weathered mica (Gommers et al., 2005; Thiry et al., 2005). These results revealed that potassium derived from CMC could increase Ex-K, promoting the fixation of Cs to minerals as well as potassium sulfate. In the field experiment, the relationship between soil Ex-K and Ex-^{137}Cs was not significant from 2018 to 2019 (Fig. 1). Although the reason could not be determined in this study, it is assumed that this might be a time-consuming reaction or related to other properties of the

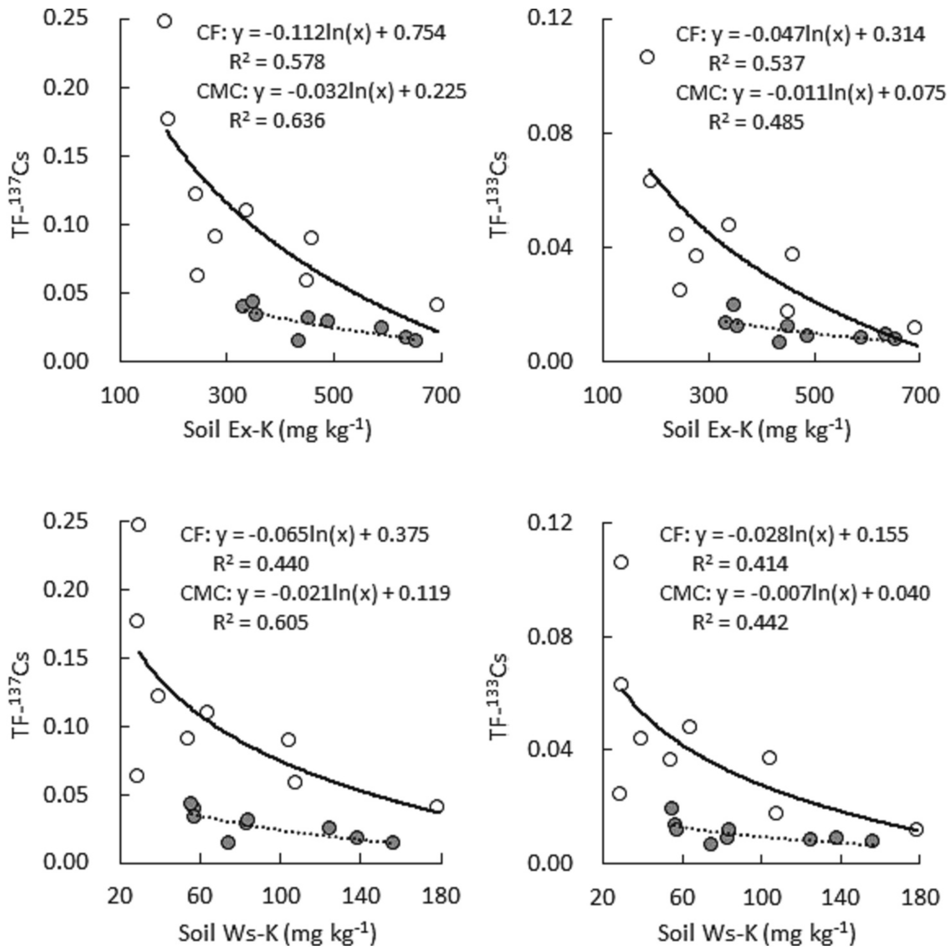


Fig. 3. Relationships between soil exchangeable (Ex)- and water soluble (Ws)-K and ^{137}Cs and ^{133}Cs transfer factors (TF-^{137}Cs and TF-^{133}Cs, respectively) from soil to Komatsuna in the pot experiment.
Open and closed circles indicate CF and CMC, respectively (N = 9).

soil.

The soil Ex-^{137}Cs ratio differed between soybean cultivation (2018–2020) and buckwheat cultivation (2021–2022) in the field experiments, with higher values observed for soybean, which had higher TF-^{137}Cs compared with buckwheat (Table 7). Kubo et al. (2021) found that the soil Ex-^{137}Cs ratio was lower after cultivating peanut, which has lower ^{137}Cs accumulation ability in grains compared with soybean. They discussed that the differences between soybean and peanut in root biomass and the ability to solubilize K and Cs fixed in soil minerals may contribute to the variation in the soil Ex-^{137}Cs ratio after cultivation. Immediately following the nuclear power plant accident in 2011, radiocesium fixation in the soil accelerated while the TF decreased, whereas radiocesium fixation in the soil is unlikely to have changed between 2020 and 2021 (IAEA, 2020). This analysis revealed that cultivation has different effects on the dynamics of ^{137}Cs in soil between soybean and buckwheat, which have different ^{137}Cs transferability from the soil to the grain.

Increasing soil Ex-K content also influenced the concentrations of various minerals (Ba, Cs, P, S, Sr, and Zn) in plants (Supplementary Table 2). Sr concentrations in plants were lower with higher soil K levels. Syaifudin et al. (2023) reported that K application reduced the translocation of Sr in soybean plants, which is consistent with the present results. Although soil Ex-K levels affect the concentrations of various elements in crops is a novel finding of this study, further research is required to elucidate mechanisms related to it.

4.3. Influence of CMC on Cs behavior in the soil and soil-to-crop transfer

In the pot experiment, Ex-^{137}Cs and Ex-^{133}Cs levels were significantly lower in CMC soil compared with CF soil (Table 9). The ratios of Ex-^{137}Cs and Ex-^{133}Cs in CMC soil (Fig. 4) as well TF-^{137}Cs (Fig. 3) were significantly lower than those in CF soil with the same soil Ex-K level. In the field experiment, the ratio of soil Ex-^{137}Cs to soil Ex-K tended to be lower (although not significantly) in the CMC1 plot compared with the $+K_2O$ plot, which received a similar amount of K. These results indicate that CMC treatment directly affects the behavior of Cs in the soil.

The application of CMC to the soil has two opposing effects: increasing the frayed edge sites (FES[19]) by supplying K to the soil (Nakao et al., 2014), and masking the FES through organic matter, reducing the ability to fix Cs into the mineral layer (Maguire et al., 1992; Nakao et al., 2015; Tashiro et al., 2018). Given that CMC soil exhibited lower Ex-^{137}Cs, Ex-^{133}Cs, and TF-Cs values compared with CF soil with the same soil Ex-K level, the increased fixation capacity due to K supply from CMC apparently outweighed the masking effect of organic matter. Another role of organic matter is to enhance RIP by binding with Al (organo–Al complexation), which reduces the formation of interlayering Al(*OH*)x, thereby maintaining the availability of FES for radiocesium adsorption (Maes et al., 1998; Rigol et al., 2002; Tashiro et al., 2018). The presence of organic acids from plant roots contributes to the solubilization of Al in the soil (Ma et al., 2001), as organic acids lead to

[19] FES: frayed edge sites

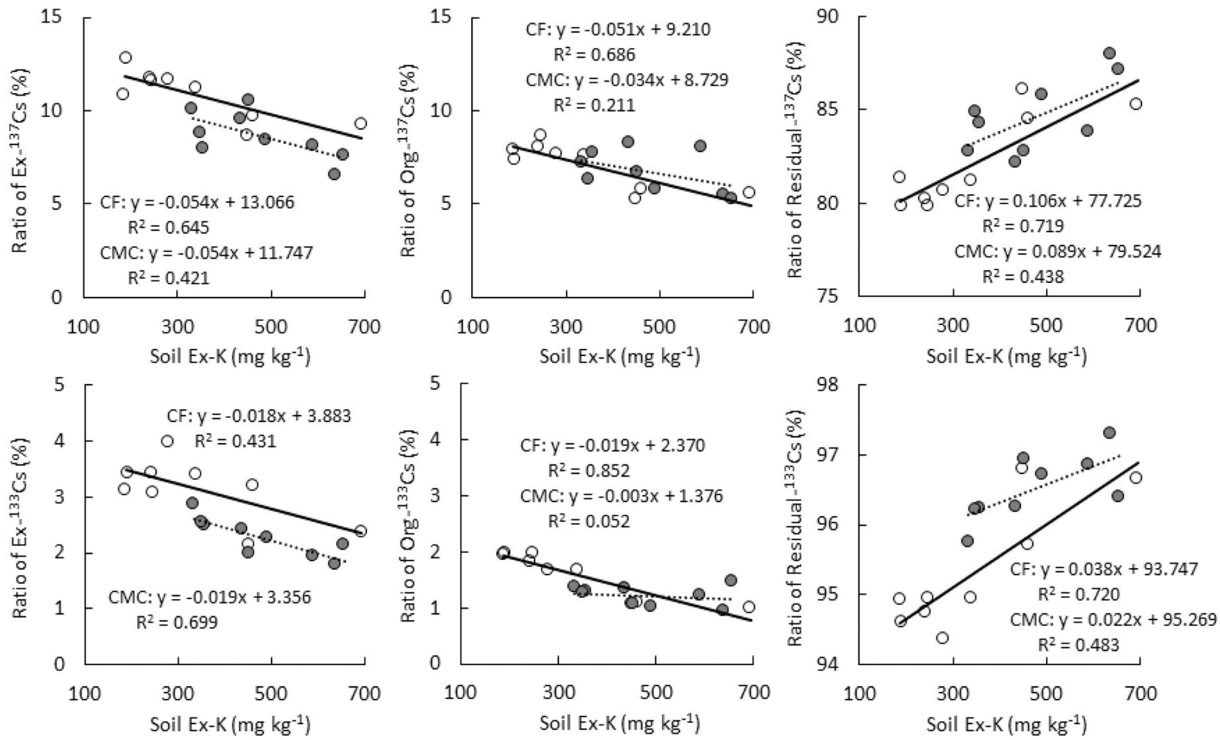


Fig. 4. Relationships between soil exchangeable (Ex)-K and Ex-Cs, organic-bound (Org)-Cs, and residual Cs ratios for ^{137}Cs and ^{133}Cs in the pot experiment. Open and closed circles indicate CF and CMC, respectively (N = 9).

rhizosphere acidification (Panchal et al., 2021). The humus content tended to increase with CMC application in both field and pot experiments (Supplementary Fig. 1; Supplementary Table 1). The formation of humus–Al complexes and the preservation of fixation sites for Cs adsorption by the reduction of intercalated Al(*OH*)x through CMC application may contribute to the decrease in Ex-^{137}Cs, Ex-^{133}Cs, and TF-Cs levels observed in our study. The concentration of Al in plants was significantly higher in CMC soil compared with CF soil (Supplementary Table 2), indicating that Al bound to humus is more readily absorbed by plants than Al(*OH*)x formed in the interlayers. RIP, which indicates the amount of fixation sites available in the soil, slightly increased with CMC application but showed no significant difference compared with CF soil (Supplementary Table 1). As described above, this study included a comprehensive analysis of the effects of supplying organic matter to the soil on the ^{137}Cs dynamics in the soil and of soil-to-crop transfer by field and pot experiments. Takeda et al. (2020) showed that, in contrast to the present study, the application of compost increases the TF-^{137}Cs in orchard grass (*Dactylis glomerata* L.) by using humus-rich Andosols with high Ex-K. This result may indicate that the application of CMC does not reduce the transfer of ^{137}Cs from the soil to the plant if the soil has a naturally high Ex-K content. Moreover, additional organic matter may block ^{137}Cs sorption by the soil, resulting in increased Ex-^{137}Cs in the soil. We considered that whether organic matter or ^{137}Cs is added first may also affect the dynamics of ^{137}Cs in the soil and the soil-to-crop transfer. Further investigation is needed to elucidate the mechanisms underlying the decrease in Ex-^{137}Cs, Ex-^{133}Cs, and TF-Cs levels due to CMC application.

In both field and pot experiments, plant DW and grain yield were higher in plots and soil with CMC application compared to those without CMC application (Tables 3, 5, and 8). It is possible that the dilution effect of Cs in plants occurred owing to differences in plant growth when the absorption amount of ^{137}Cs is same between CMC application and no CMC application. However, the concentrations of K and other nonessential elements in plants did not differ between CMC and CF treatments (Supplementary Table 2), suggesting that the lower ^{137}Cs and ^{133}Cs levels observed with CMC application cannot be solely explained by dilution effects. Further examinations in detail are required to reveal the effect of crop growth on ^{137}Cs accumulation.

5. Conclusions

The main conclusions derived from the results of crop cultivation using soils of decontaminated field in coastal (Hamadori) area of Fukushima Prefecture were as follows:

(1) The application of CMC to fields decontaminated of radiocesium enhanced the growth and grain yield of buckwheat, soybean, and Komatsuna through the increase in the levels of macro and micronutrients and improved soil fertility.
(2) The increased soil Ex-K, due to CMC application, led to a reduction in the transfer of ^{137}Cs from soil to crops in both field and pot experiments.
(3) The higher crop yields and lower transfer of ^{137}Cs from soil to crops in CMC plots persisted for at least three years after the cessation of CMC application for two years.
(4) TF-Cs were lower in CMC-treated soil compared with CF-treated soil with the same Ex-K level owing to the lower Ex-Cs content and higher residual Cs content in CMC-treated soil.

Regarding (1) to (3), similar results were observed in another experiment conducted over five years (2016–2020) in a different field after the decontamination of radioactive materials (Supplementary Table 4).

Unfortunately, we could not determine whether the fertility had returned to the level before contamination due to lack of data on the soil chemical properties of the fields used in this study before decontamination; however, the application of CMC to the fields after decontamination improved fertility and various nutrients. In particular, the supply of K from CMC to the soil led to a reduction in the transfer of ^{137}Cs from the soil to crops, contributing to ensuring food safety; soybean, which had shown radiocesium concentration close to standard value (100 Bq kg^{-1}) in nearby areas of the field conducted this study immediately after the nuclear power plant accident in 2011 (Fukushima prefecture, 2023), had a grain radiocesium concentration below the standard values and the values were sufficiently low in the CMC-treated plot in this study. As previously mentioned, it is considered that CMC application is a sustainable method for restarting farming after the decontamination of radiocesium in Fukushima Prefecture. A stock farm expected to produce 12,000 t of CMC per year is scheduled to be completed in the coastal area of Fukushima Prefecture in 2025 (Dairy cooperative in Fukushima, 2021). The use of locally produced CMC on decontaminated fields holds promise as a desirable method for implementing the agricultural resumption while reducing the use of chemical fertilizer and recycling local resources, and for achieving environmental restoration in these areas.

In contrast, the relationship between CMC application and Ex-^{137}Cs and TF-^{137}Cs needs to be elucidated further, as, in some cases, the application of CMC enhances the transfer of ^{137}Cs from the soil to crops. Further analyses and measurement of actual soil chemical properties (including Ex-K and humus levels) in decontaminated fields may be necessary to make decisions on CMC application.

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CRediT authorship contribution statement

Masataka Suzuki: Conceptualization, Investigation, Data curation, Formal analysis, Writing–original draft. **Katashi Kubo:** Conceptualization, Investigation, Data curation, Formal analysis, Project administration, Visualization, Writing–original draft, Writing–review and editing. **Mayumi Hachinohe:** Investigation, Resource, Validation, Writing–review and editing. **Takashi Sato:** Investigation, Validation, Supervision, Writing–review and editing. **Hirofumi Tsukada:** Methodology, Validation, Writing–review and editing. **Noriko Yamaguchi:** Methodology, Validation, Writing–review and editing. **Toshihiro Watanabe:** Investigation, Resource, Validation, Writing–review and editing. **Hayato Maruyama:** Investigation, Validation, Writing–review and editing. **Takuro Shinano:** Funding acquisition, Supervision, Writing–review and editing.

Declaration of competing interest

The authors declare no conflict of interest.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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