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Title

Distribution of cesium-bearing microparticles in the paddy field and their effect on transfer factor of radioactive cesium for brown rice

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Abstract

Radioactive cesium-bearing microparticles (CsMPs) were insoluble glassy matrixes derived from Fukushima Daiichi Nuclear Power Plant accident. CsMPs may locally increase radioactive cesium (RCs) concentration of soils and crops. Furthermore, RCs in the CsMPs is included in insoluble grassy particle, it is considered that it may be difficult for crops to absorb. Transfer factor (TF) is used to evaluate transfer of RCs from soils to crops. However, the conventional TF is a simple ratio of RCs concentrations between them. This suggests that conventional TF may not be able to accurately assess RCs transfer from soils to crops when CsMPs are in the soil. Since there are many contaminated forests in Fukushima Prefecture, Japan, there are concerns about secondary contamination due to the inflow of CsMPs. In this study, we investigated the deposition of CsMPs in the paddy field located within specified reconstruction and revitalization base area in the first year of resumed cultivation following the accident. Furthermore, we evaluate the impact of CsMPs on RCs concentration in the soil and TF of brown rice.

The accumulation of RCs and CsMPs in our study field was not observed. RCs concentration of brown rice was less than the shipping limit defined by Japanese government. These suggested that the impact of secondary contamination was not large, and cultivation can be done safely. However, because there are many contaminated sites in the surrounding area, and it is necessary to continue to monitor the effects of radiation in the future. Furthermore, the proportion of CsMPs on RCs concentration in the soil was less than 10%. Therefore, CsMPs may not have a significant effect on transfer factor of RCs for brown rice.

Key word

Radioactive cesium, Radioactive cesium-bearing microparticles, Fukushima, Paddy soil, Transfer factor

1. Introduction

Lots of radionuclides were scattered around Fukushima Prefecture, Japan, due to the Fukushima Daiichi Nuclear Power (FDNPP) accident occurred on March 11, 2011[1,2]. Because Radioactive cesium-137 (^{137}Cs), which has a long half-life of approximately 30 years, is deposited in large quantities, it is necessary to grasp its long-term behavior and the effect of radiation in the environment [3]. After the FDNPP accident, decontamination activity such as removal of vegetation and topsoil were conducted in the farmland and residential area [4]. Furthermore, intensive decontamination has been carried out in some difficult-to-return zones where residential and activity had been restricted due to high radiation air dose rate. As a result, residential and agricultural activities have resumed in these areas as specified reconstruction and revitalization base areas since 2022 [5]. However, decontamination in the forest areas has not been completed because there are large target areas for decontamination in the prefecture and it is difficult to introduce heavy machinery on steep slopes. Therefore, there is a concern that ^{137}Cs may move from their forests to the surrounding area and contaminate soil, water, and agricultural crops [6, 7, 8, 9].

Radioactive cesium-bearing microparticles, hereinafter referred to CsMPs, are insoluble glassy matrixes derived from the FDNPP accident [10, 11, 12, 13]. CsMPs has a particle size of a few µm, but are estimated to have an ^{137}Cs concentration of 10^5 -10^6 times higher per unit mass than particulate ^{137}Cs in which ^{137}Cs attached to clay particles [10, 11]. Therefore, previous studies have shown that CsMPs may attach the surface of crops such as paddy rice and Japanese mustard spinach and increase their ^{137}Cs concentration [6, 7]. Ikehara et al. (2018) defined that CsMPs were particles with a size of 114 µm or less and radioactivity of 0.06 Bq or more. Ikehara et al. (2020) [12] investigated CsMPs in soils collected through out Fukushima Prefecture, and showed lots of CsMPs deposit in the soil in the highly contaminated areas located near and northwest of the FDNPP. They also showed that the proportion of CsMPs to radioactive cesium (RCs, sum of ^{137}Cs and ^{134}Cs) concentration in the soil collected from less contaminated areas located more than 30 km north west of the FDNPP was larger than that in the soil collected from near the FDNPP This was because the concentrations of RCs other than CsMPs in the soil were lower than those near the FDNPP. Tatsuno et al. (2023) [9] showed that CsMPs might cause a local increase in RCs concentration in the river water. Furthermore, they also indicated that CsMPs might significantly increase particulate RCs concentration, leading to an overestimation of the solid-liquid distribution coefficient for RCs in the river through the less contaminated area. These previous studies have suggested that CsMPs with high radioactivity have a great effect on the RCs concentration and transfer value between the soil and water reaction in the less contaminated area. The relatively large amount of CsMPs remained in the surface layer of forest soil in the area close to the FDNPP even in 2022-23 [14], and it is thought that these CsMPs might enter decontaminated farmland and affect the RCs concentration in the soil. Previous studies on agricultural land, especially paddy fields, have reported on the distribution trends of RCs within the field and their transfer to rice [15, 16, 17, 18]. However, the distribution and concentration of CsMP in the paddy soil are not clear. In addition, since CsMPs are insoluble matrixes, it is considered that RCs in them is difficult to release from themselves [10, 11]. Therefore, previous study on crop cultivation using contaminated soils suggested that RCs in CsMPs might not contribute to RCs uptake from the roots [8]. When evaluating RCs adsorption from soils to crops, a transfer factor, which is the ratio of RCs concentrations in soil and crops, is used [18]. However, the presence of insoluble matrixes such as CsMPs in soils may make it difficult to accurately evaluate the RCs transfer factor pf crops, as is the case with river water [9]. In order to correctly evaluate the safety of farmland and crops against radiation effects and the transfer of RCs in agricultural crops, it is necessary to understand the distribution of CsMPs in the farmland and their effects on crops.

In this study, we investigated the distribution of RCs and CsMPs in the paddy soil immediately after planting and before harvesting in the first year after the resumption of cultivation. Furthermore, we evaluated the RCs transfer factor from soil to brown rice, both in the presence and absence of CsMPs, in order to assess the impact of CsMPs on estimation for RCs transfer.

2. Experimental

2.1 Study site and sampling

The study site is a paddy filed located in the specified reconstruction and revitalization base areas, approximately 12.5 km northwest from FDNPP. Farming resumed in the field in April 2023. The paddy field with an area of approximately 752 m^2 and was divided into 16 plots (approximately 40.8-53.2 m^2 of each plot) for collecting soil and crop samples (Fig.1). In this study, to compare the values such as RCs concentration of soil and crop by the locations in the paddy field, three to four plots each near the water intake, center of the field, and water outlet were defined as the following three sampling area; intake, center, and outlet (Fig. 1). Furthermore, the average of all 16 plots was defined as the entire paddy field. The cultivated rice sample was "Tenno Tsubu," a recommended variety in Fukushima Prefecture, which is also widely applied in other paddy fields along the coast of the Prefecture [19].

Soil sampling was conducted twice. First time was conducted after just planting on May 23, 2023, while the second time was just before harvesting on September 18, 2023. Each sampling period was defined as the planting and the harvesting period, respectively. In planting period, soil column samples were collected from each plot in the paddy filed under flooding using an acrylic cylinder with a length of 40 cm and a diameter of 4.2 cm. In the harvesting period, soil column sample was collected from the ground surface using a liner soil collector with a length 30 cm and the diameter of 5 cm (DIK-110C, Daiki, Japan). In both sampling periods, soil column samples were taken to a depth that allowed each sampler (i.e., acrylic cylinder and liner soil collector) to be inserted without soil disturbing. Therefore, the length of column sample varied from 11 cm to 21 cm for each plot. For crop samples, three aboveground rice plants were collected near the soil sampling point of each plot in the harvesting period.

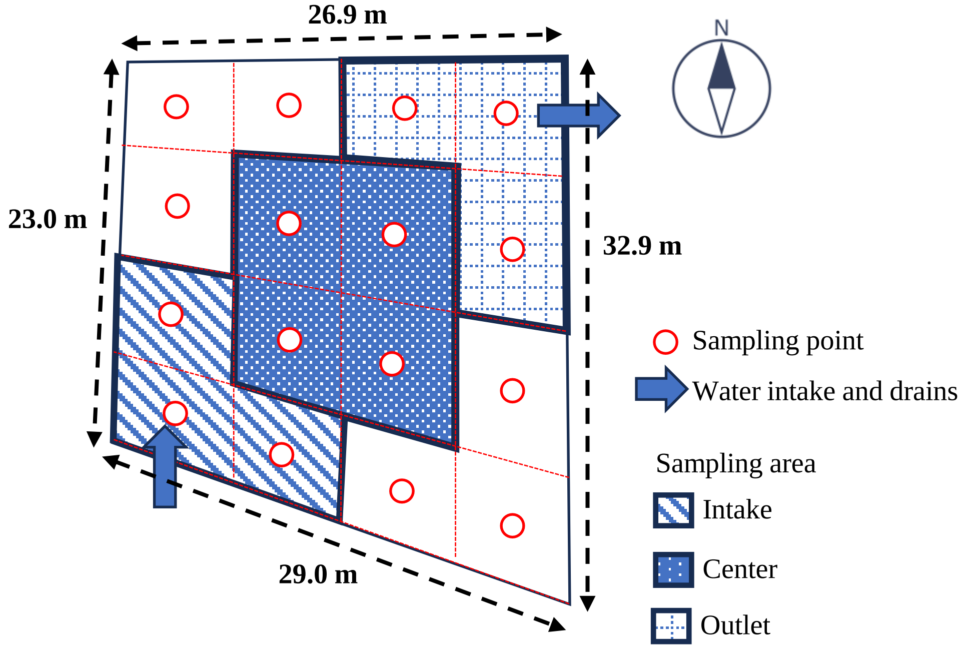

Fig.1 Sampling point and areas

2.2 Sample analysis for RCs and CsMPs in the soil

After taking the samples, soil column samples were cut into the soil layers according to depth. For the top 0-5 cm layer, samples were cut at the following intervals: 0-0.25, 0.25-0.5, 0.5-1, 1-2, 2-3, 3-4, and 4-5 cm. For soil deeper than 5 cm, samples were cut at 2 cm intervals. Each soil layer was dried in an oven at 105℃ and collected through the sieve with porosity of 2 mm. Sieved soil layer was put into a 100 mL of U8 container (Pla tsubo 3–20, Umano Kagaku Youki Co., Ltd., Osaka, Japan). RCs concentration (i.e., sum of ^{134}Cs and ^{137}Cs concentration) of the soil layer was measured by germanium semiconductor detector (GC4020, CANBERRA Industries Inc., USA, relative efficiency 32%). Gamma-ray peaks of 604 and 662 keV were used to measure concentration of ^{134}Cs and ^{137} Cs, respectively. The counting efficiency was calibrated using the standard source (MX033U8PP and MX035; The Japan Radioisotope Association, Tokyo, Japan.

Based on previous studies [9, 11, 14], the concentration of RCs derived from CsMPs, hereinafter called to CsMPs concentration, was quantified for each soil layer using the following procedure. After measuring the RCs concentration, a part of soil sample was taken from each soil layer, and mixed with pure water. After thoroughly stirring the mixture with a shaker and ultrasonic disperser, the suspended particles were passed through a mesh with a porosity of 114 µm and then collected on a filter with porosity of 0.45µm. It should be noted that there may be some CsMPs with size smaller than 0.45 µm [11]. However, most CsMPs in previous studies were only a few µm in size [11], and CsMPs in river water samples remained on the 0.45 µm filter [9]. Therefore, the soil particles remaining on the filter with the porosity of 0.45 µm was analyzed for CsMPs in this study. The suspended particles on the filter was dried in the oven at 60 ℃ for a few hours and put into a zip bag to avoid losing particles, hereinafter called to CsMPs sample. CsMPs samples of each soil layer and standard samples (0.05-2.00 Bq per spot) were placed on a plastic sheet and covered with an imaging plate (IP; BAS IP MS 2025; Cytiva, Japan) for 24 hours. After contacting, the highly radioactive spot recorded on the IP was read on the IP reader (FLA-9500, Ge Healthcare Bio-Science AB, Sweden). The radioactivity of each spot was calculated based on the calibration curve using standard samples [9]. In this study, CsMPs were defined as a particle with radioactivity of 0.06 Bq or more using an image processing software (Image J, National Institute of Health, USA) [11]. CsMPs concentration in the soil layer was obtained using the equation 1 (eq.1);

$$C_{CsMPs} = \frac{R_{CsMPs}}{m_{CsMPs}} \quad \text{(eq. 1)}$$

where R_{CsMPs} is the total radioactivity of CsMPs detected in each CsMPs sample. m_{CsMPs} was the mass of samples taken from each soil layer when preparing CsMPs samples.

In the vertical distribution RCs and CsMPs concentration in the soil, the proportion of CsMPs concentration to RCs concentration, hereinafter called to proportion of CsMPs, in each soil layer was calculated from the following equation (eq.2);

$$P_{CsMPs\_i} = \frac{C_{CsMPs\_i}}{C_{RCs\_i}} \cdot 100 \quad \text{(eq. 2)}$$

where P_{CsMPs} (%) indicate the proportion of CsMPs of *i* soil layer. C_{CsMPs_i} (Bq kg^{-1}) and C_{RCs_i} (Bq kg^{-1}) were CsMPs and RCs concentration in *i* soil layer.

The centroid depth of RCs and CsMPs concentration in the vertical direction, $\langle x \rangle$ (cm), were calculated from the following equation to grasp the trend of vertical distribution for them in the soil column samples [14, 21].

$$\langle x\rangle = \frac{\sum_i x_i C_i \Delta x_i}{\sum_i x_i \Delta x_i} \quad \text{(eq. 3)}$$

where c_i (Bq kg^{-1}) indicates the concentration of Cs or CsMPs in i-soil layer. x_i (cm) is the intermediate depth of the i-soil layer. Δx_i (cm) is the thickness of i-soil layer.

2.3 RCs concentration in the brown rice

After sampling the rice plants, they were air-dried, threshed and husked to collect the brown rice. The collected brown rice was put in to the U8 container. RCs concentration in the brown rice was measured using the germanium semiconductor detector. In this study, the RCs concentration per unit mass of brown rice was described as the value of fresh weight (Bq $kgFW^{-1}$) assuming the moisture content of 15%.

2.4 Deposition of RCs and CsMPs in the topsoil

To compare the deposition of RCs and CsMPs of the topsoil with the depth of 0-5 cm between different sampling areas and periods, their deposition volumes were calculated using the equivalent soil mass (ESM) basis [22, 23]. ESM basis is used to compare the amount of nutrients for the reference mass of soil in the field where the dry bulk density has changed due to tillage or trampling by livestock. In this study, unlike the dry soil in the harvesting period, the soil in the planting period was under flooded condition, and water might enter into the soil porosity, causing the soil particles to float. It was expected that the dry bulk density (i.e., amount of dry soil per depth) would be different between the two sampling periods even under the same condition of soil depth and plot. Therefore, ESM basis was applied to compare to the deposition of RCs and CsMPs of the topsoil between different sampling areas and periods. The reference soil mass was determined as the average dry mass per unit area of topsoil collected from depth of 0 – 5 cm in 16 plots in the harvesting period (53.2 kg m^{-2}). The accumulated RCs and CsMPs up to the reference soil mass of each plot were calculated using the following equations;

$$TI_{RCs\_R} = TC_{RCs}(n) + (M_R - M(n)) \cdot C_{RCs}(n+1) \quad \text{(eq. 4)}$$

$$TC_{RCs}(n) = \sum_i^n M_i \cdot C_{RCs\_i} \quad \text{(eq. 5)}$$

$$TI_{CsMPs\_R} = TC_{CsMPs}(n) + (M_R - M(n)) \cdot C_{CsMPs}(n+1) \quad \text{(eq. 6)}$$

$$TC_{CsMPs}(n) = \sum_i^n M_i \cdot C_{CsMPs\_i} \quad \text{(eq. 7)}$$

where TI_{RCs_R} (kBq m^{-2}) and TI_{CsMPs_R} (kBq m^{-2}) are the deposition of RCs and CsMPs per unit area in reference soil mass, hereinafter called to as RCs inventory and CsMPs inventory, respectively. TC_{RCs_R} (kBq m^{-2}) and TC_{CsMPs_R} (kBq m^{-2}) are the cumulated values of RCs or CsMPs up to the soil layer n just before the reference soil mass is reached. M_R is the reference soil mass. *M(n)* is the cumulative soil mass per unit area until the reference mass is reached (kg m^{-2}). M_i is the mass of dry soil layer-i per unit area (kg m^{-2}). C_{RCs} (n+1) and C_{CsMPs} (n+1) indicate RCs and CsMPs concentrations of the soil layer that reach or exceed the reference soil mass (kBq kg^{-1}). C_{RCs_i} (kBq kg^{-1}) and C_{CsMPs_i} (kBq kg^{-1}) are RCs and CsMPs concentrations of the soil layer-i, respectively.

2.5 Apparent and net transfer factor of RCs to brown rice

To calculate the transfer factors from soil to brown rice, the RCs and CsMPs concentrations of the topsoil with the depth of 0–5 cm in the harvesting period were determined using the following equations;

$$C_{RCs\_0-5} = \frac{\sum_i m_i \cdot C_{RCs\_i}}{\sum_i m_i} \quad \text{(eq. 8)}$$

$$C_{CsMPs\_0-5} = \frac{\sum_i m_i \cdot C_{CsMPs\_i}}{\sum_i m_i} \quad \text{(eq. 9)}$$

where $C_{RCs\_0\text{-}5}$ and $C_{CsMPs\_0\text{-}5}$ indicate the RCs and CsMPs concentration of the soil with

0-5 cm depth, respectively (Bq kg^{-1}). m_i is the dry mass of soil layer-i (kg). It should be noted that the RCs concentration of dry weight for brown rice (Bq kgDW^{-1}) was used in the calculation of the transfer factor. In order to evaluate the transfer of RCs from surface soil to brown rice, the transfer factor was obtained for each sampling plot using the following equation (eq. 10) [15, 18];

$$TF_{apparent} = \frac{C_{RCs\_rice}}{C_{RCs\_0-5}} \quad \text{(eq. 10)}$$

where $TF_{apparent}$ indicates the transfer factor of RCs from the surface soil to brown rice. C_{RCs_rice} (Bq kg^{-1}) is the RCs concentration in the brown rice. In addition, the net transfer factor, TF_{net}, which subtracts the contribution of CsMPs from RCs concentration in the soil, was calculated using the following equation (eq.11).

$$TF_{net} = \frac{C_{RCs\_rice}}{C_{RCs\_0-5} - C_{CsMPs\_0-5}} \quad \text{(eq. 11)}$$

3. Results

3.1 Vertical distribution of RCs and CsMPs in the paddy soil

Fig. 2 shows the vertical distribution of RCs and CsMPs concentrations and proportion of CsMPs in each sampling area collected in the planting and harvesting periods. In each area and period, the RCs concentration was almost the same order between the soil depth of 0-15 cm, or tended to decrease gradually with depth. This distribution trend was similar to that in the previous study after puddling [17]. On the other hand, CsMPs were distributed heterogeneously in the soil depth of 0-21 cm. Proportion of CsMPs in most soil layer was less than 5%, while that in some was greater than 10% (e.g. the soil layers with the depth of 0-0.25 cm of the center area and 11-13 cm of outlet area in the planting period, and those with the depth of 3-4 cm of the intake area, 2-3 cm of center area, 4-5cm and 13-15 cm of outlet area in the harvesting period). Table 1 indicates the centroid depth of RCs and CsMPs concentrations by sampling areas and periods. When RCs and CsMPs were evaluated independently, the centroid depth did not differ significantly among areas during the same period, nor between periods within the same area.

Table 1 Centroid depth of RCs and CsMPs concentration in the soil column

		intake	center	outlet	entire
Planting	RCs (cm)	6.45 (±0.37)	5.94 (±0.94)	6.90 (±1.10)	6.46 (±0.29)
	CsMPs (cm)	7.62 (±0.81)	3.55 (±1.83)	4.57 (±2.73)	5.50 (±0.90)
Harvesting	RCs (cm)	7.18 (±0.82)	8.85 (±1.44)	8.33 (±0.23)	8.16 (±0.43)
	CsMPs (cm)	6.06 (±0.98)	7.68 (±2.93)	7.04 (±3.52)	6.16 (±1.09)

The number of parentheses indicates the standard error calculated from three or four column samples in each sampling area.

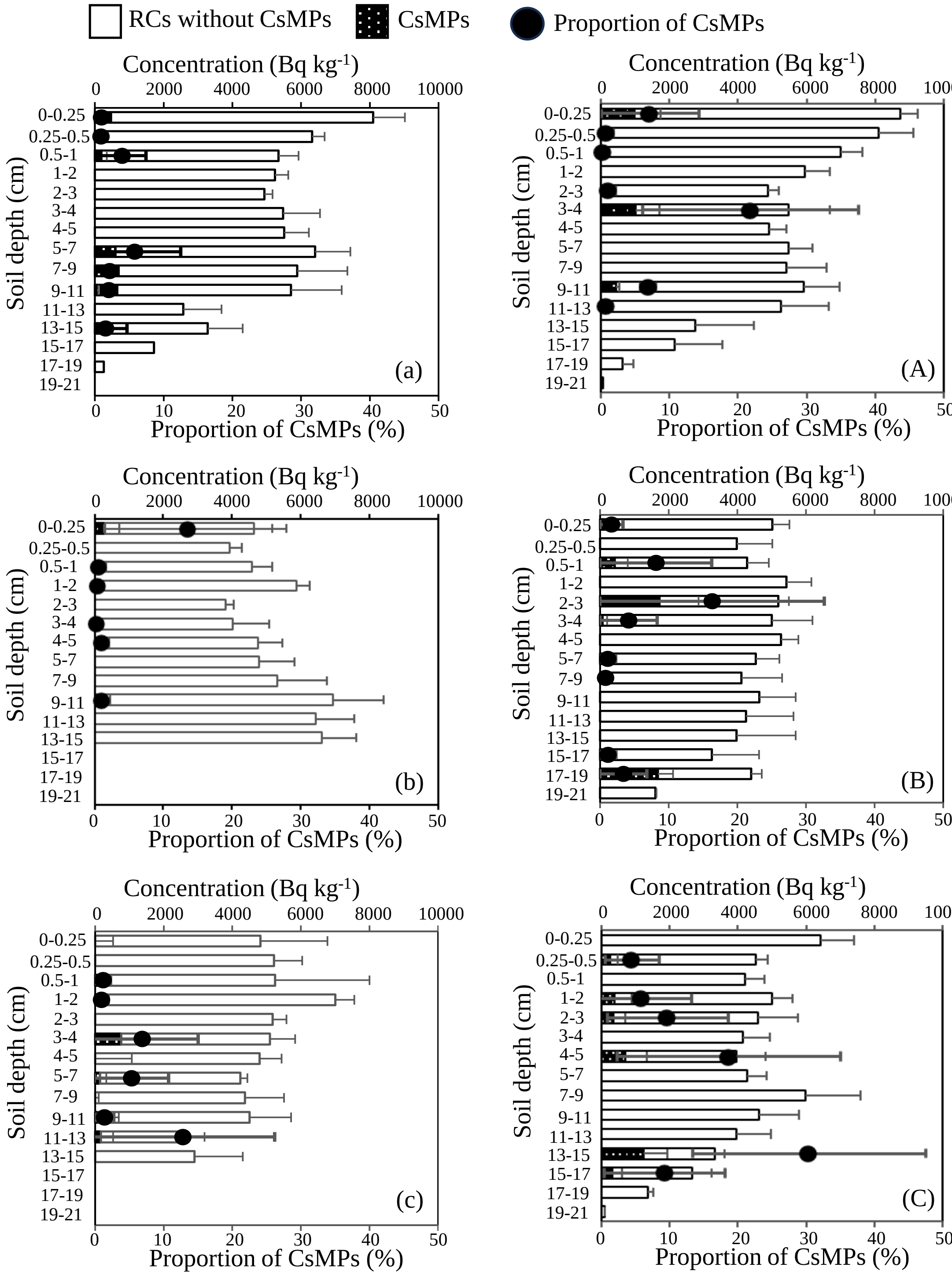

Fig. 2 Vertical distribution of RCs and CsMPs concentration and Proprtion of CsMPs in the soil layers collected in the planting and harvesting periods

Horizontal error bars indicate the standard deviations calculated from three or four column samples. The small letters in parentheses indicate the following sampling area in the planting period; (a) intake, (b) center, and (c) outlet areas. The small letters in parentheses indicate the following sampling area in the harvesting period ; (A) intake, (B) center, and (C) outlet areas.

3.2 Inventories of RCs and CsMPs in the planting and harvesting periods

Fig. 3 shows the inventories of RCs and CsMPs of entire and each area in the planting and harvesting periods. There was no significant difference in RCs inventory when comparing the entire and each sampling area at the same sampling period (Fig. 3-(a)). Furthermore, there was no significant difference in RCs inventory when comparing the planting and harvesting periods in the same sampling area. The average CsMPs inventories of each sampling area in the harvesting period tended to increase more than those in the planting period. However, this difference was not statistically significant ($p > 0.05$). As in the case of RCs inventory, there were no significant difference in CsMPs inventory when comparing between the sampling areas at same sampling period or the periods in the same area.

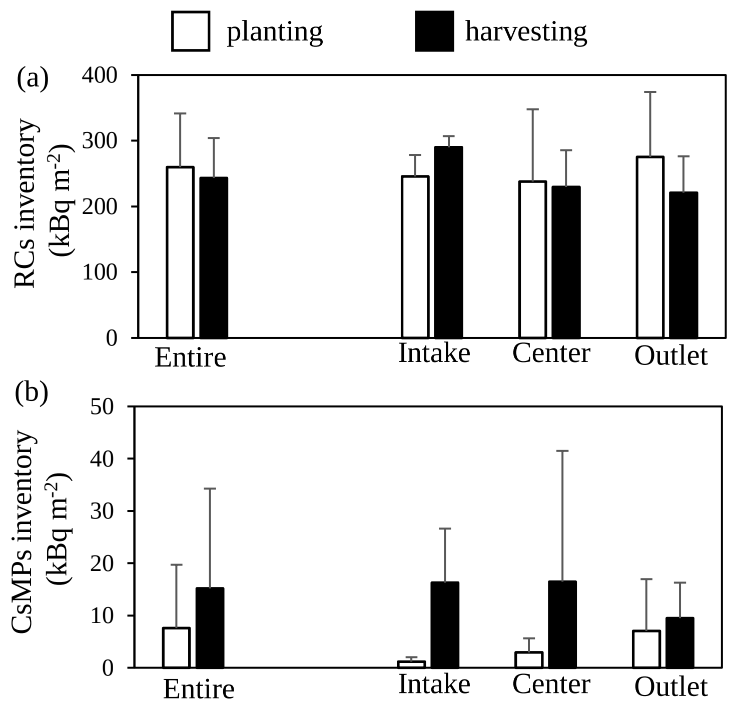

Fig. 3 Inventories of (a) RCs and (b) CsMPs for all and each sampling area in the planting and harvesting periods

Vertical error bars indicate the standard deviation.

3.3 RCs concentration in brown rice

Fig. 4 shows the average RCs concentration of brown rice collected in the entire and each sampling area. The RCs concentration of brown rice in the intake area was greater than the entire and the center areas. The RCs concentration of the brown rice in each sampling plot ranged from 8.0 to 26.2 Bq $kgFW^{-1}$, which were significant below the shipping limit (100 Bq $kgFW^{-1}$) [24].

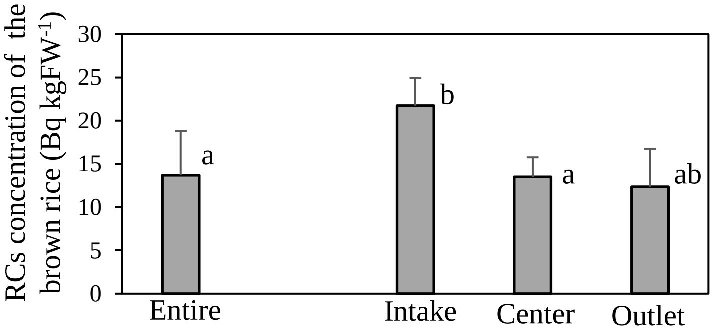

Fig. 4 Average RCs concentration in the brown rice in all plots and each sampling area
Vertical error bars indicate the standard deviation.

3.4 RCs and CsMPs concentrations in the topsoil and transfer factors of RCs for brown rice

Fig. 5 shows the RCs and CsMPs concentrations and proportion of CsMPs of the topsoil with 0-5 cm depth in the harvesting period. The RCs concentration in the topsoil ranged from 4.55×10^3 to 5.73×10^3 Bq kg^{-1} in each sampling area, and the average concentration in the entire paddy field was 4.98×10^3 Bq kg^{-1}. CsMPs concentration of each area was between 283 to 310 Bq kg^{-1}, and the average concentration in the entire paddy field was 304 Bq kg^{-1}. The proportion of CsMPs in each sampling area was 4.4 to 6.5 %. The average proportion of CsMPs was 5.8% for the entire paddy field. When comparing between sampling areas, there were no significant differences in the concentration of RCs and CsMPs and the proportion of CsMPs in the topsoil.

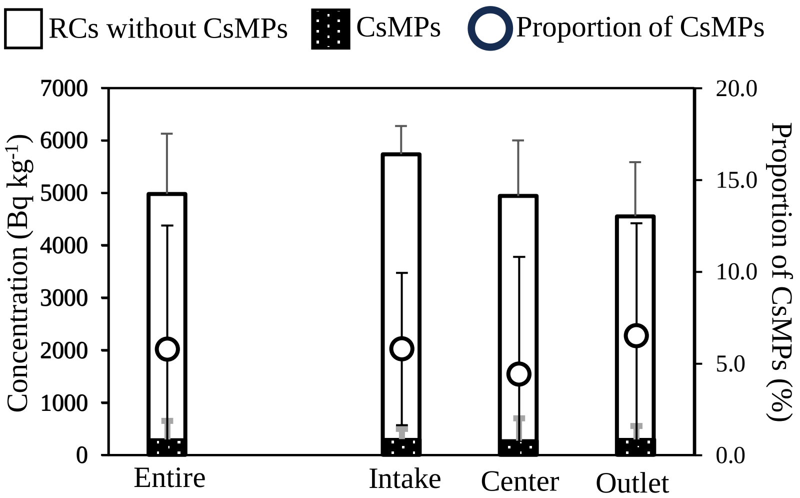

Fig. 5 RCs and CsMPs concentration and proportion of CsMPs in the soil with depth of 0-5 cm in the cultivation period

Vertical error bars indicate the standard deviation.

Fig.6 shows the apparent and net transfer factors of RCs to brown rice in the entire and each sampling area. The apparent transfer factors in the intake, center, and outlet areas were 3.24×10^{-3}, 2.47×10^{-3}, and 2.27×10^{-3}, respectively. The net transfer factors in the intake, center, and outlet areas were 3.45×10^{-3}, 2.55×10^{-3}, and 2.42×10^{-3}, respectively. For the entire paddy filed, the apparent and net transfer factors were 2.41×10^{-3}, and 2.54×10^{-3}, respectively. There was no significant difference between apparent and net transfer factors in each sampling area and entire paddy field.

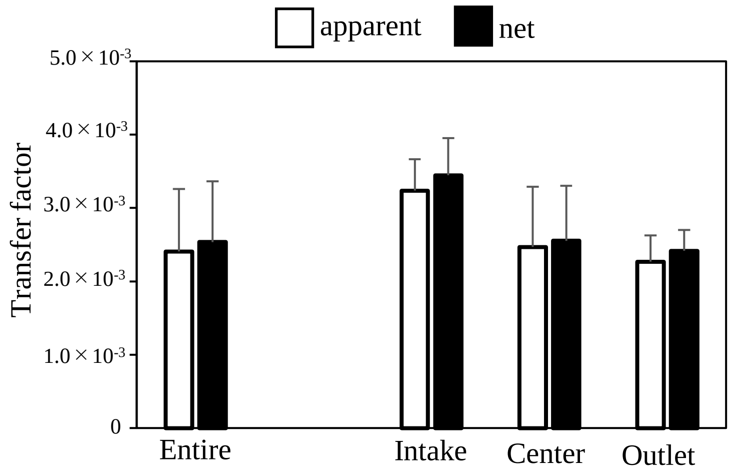

Fig. 6 Apparent and net transfer factor of Cs

Vertical error bars indicate the standard deviation.

4. Discussion

4.1 Secondary contamination in the paddy field

Some previous studies showed that there was RCs deposition trends to decrease from water intake to outlet in the paddy field [15, 17]. However, in both sampling periods, there were no significant difference in inventories and centroid depth of RCs and CsMPs between sampling points, respectively (Table1 and Fig. 3). Furthermore, at the same sampling area, no significant difference was observed between two sampling periods. These results suggest that deposition of RCs and CsMPs might not significantly change during our observation period, and that external contaminants might be rarely introduced to this study field via irrigation water and atmospheric deposition. The RCs concentration of brown rice was less than the shipping limit. This study suggested that the impact of secondary contamination on farmland was not large even within the specified reconstruction and revitalization base area. However, it should be noted that these results are based solely on the period and location of this study. It is necessary to continue to investigate the dynamics of Cs and their effects on secondary contamination for soils and crops.

4.2 Effect of CsMPs on RCs transfer factor of brown rice

Previous studies showed that the apparent transfer factor of RCs ranged from 1.50×10^{-3} to 6.80×10^{-3} in the decontaminated paddy field located from 40 km northwest of FDNPP [17]. In addition, the average value of apparent transfer factor for polished rice surveyed in Aomori Prefecture, Japan, in 1996-1997 was 0.0016 [18]. The net transfer coefficients in this study were of a same order of magnitude (Fig.5). Furthermore, there was no significant difference in the transfer factor of RCs for brown rice between apparent and net values (Fig. 5). This may be because the proportion of CsMPs was low (< 10 %) as in the case of the previous study conducted in the highly contaminated area [9]. As mentioned above, in a field such as the one in this study, where there is no inflow of CsMPs from external source, it is considered that the current transfer factor can be appropriately used to evaluate the transfer of RCs to brown rice. However, it is possible that soil disturbance due to plowing or irrigation water may change the RCs distribution at the paddy field in the future [15, 17]. In addition, in forest soils located within 5 km of the study site, CsMPs are distributed in large amounts in the surface layer within 5cm depth, and it has been pointed out that there is a possibility that they may be remobilized by soil erosion [14]. There is a possibility that CsMPs may be transported from the surrounding area in the future due to the movement of sediment by water and wind [8, 9]. On the other hand, it is considered that soil disturbance caused by agricultural work may change the distribution of CsMPs in the soil or cause them to run off outside the field [15].

Therefore, it is necessary to continue to understand the dynamics of CsMPs in paddies, taking into account changes over time and at other sites.

5. Conclusion

We investigated the distribution of CsMPs in the paddy soil and their effect of Cs transfer factor to brown rice during planting and harvesting period. During the observation period, the depositions of RCs and CsMPs in the field did not change significantly. This result indicated that there was little secondary contamination in this study site. Furthermore, the proportion of CsMPs in the RCs concentration in the soil was less than 10% in both periods. These results suggest that CsMPs might not have a significant impact on the conventional transfer coefficient. Furthermore, the RCs concentration of brown rice was well below the shipping limit. This study demonstrated that rice cultivation can be safely conducted within the specified reconstruction and revitalization base area. It should be noted that the inflow of CsMPs from the surrounding area will need to be further considered, and that it is important to continue investigation.

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Supplementary

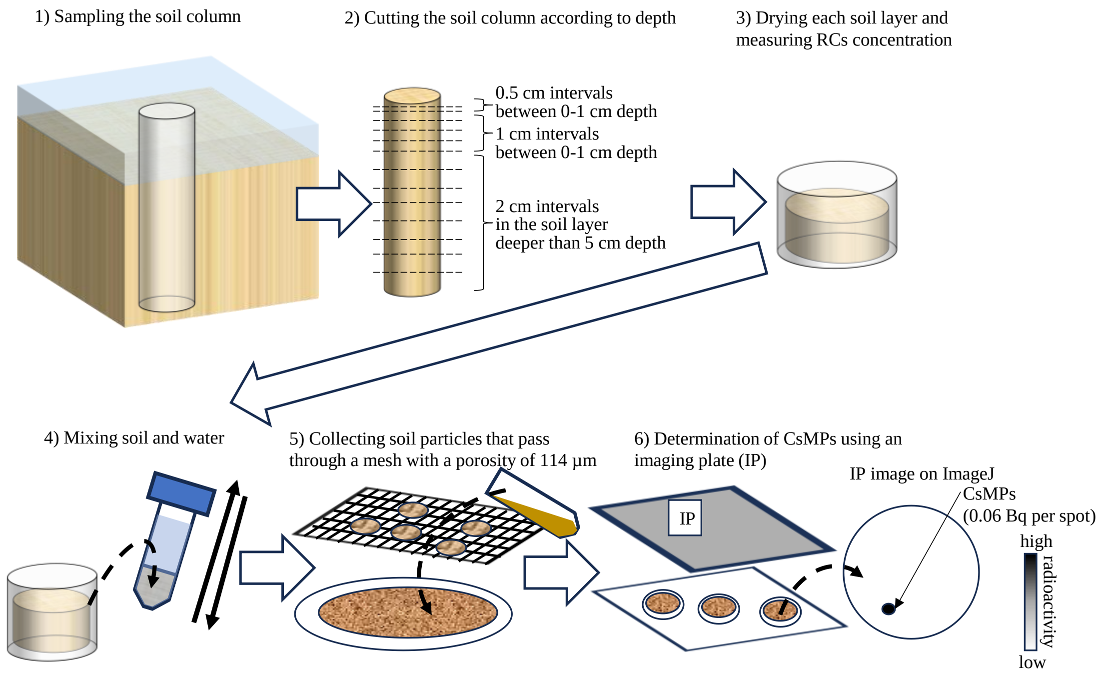

Fig. S1 Procedure from soil sampling to CsMPs measurement