Effects of Applying Potassium, Zeolite and Vermiculite on the Radiocesium Uptake by Rice Plants Grown in Paddy Field Soils Collected from Fukushima Prefecture

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Abstract: Radionuclides were released into the environment as a consequence of the Fukushima Daiichi Nuclear Power Plant accident that occurred on 11 March 2011. Radiocesium at an abnormal concentration was detected in brown rice produced in paddy fields located in northern part of Fukushima Prefecture. We examined several hypotheses that could potentially explain the excessive radiocesium level in brown rice in some of the paddy fields, including (i) low exchangeable potassium content of the soil, (ii) low sorption sites for cesium (Cs) in the soil, and (iii) radiocesium enrichment of water that is flowing into the paddy fields from surrounding forests. The results of experiments using pots with contaminated soil indicated that the concentration of radiocesium in rice plants was decreased by applying potassium or clay minerals such as zeolite and vermiculite. The obtained results indicated that high concentrations of radiocesium in rice are potentially a result of the low exchangeable potassium and sorption sites for Cs in the soils. Application of potassium fertilizer and clay minerals should provide an effective countermeasure for reducing radiocesium uptake by plants. Radiocesium-enriched water produced by leaching contaminated leaf litter was used to irrigate rice plants in the cultivation experiments. The results indicated that the radiocesium concentrations in rice plants increased when the radiocesium-enriched water was applied to the potted rice plants. This indicated the possibility that the radiocesium levels in brown rice will increase if the nuclide is transported with water into the rice paddy fields from surrounding forests.

Key words: Fukushima Daiichi Nuclear Power Plant, Potassium, Radiocesium, Rice, Vermiculite, Zeolite

A large amount of radionuclides was released into the environment as a consequence of the accident at Fukushima Daiichi Nuclear Power Plant (FNPP) that was triggered by the earthquake and subsequent tsunami that occurred on 11 March 2011. The released radionuclides were deposited on land and sea surfaces. Major radionuclides observed in surface soils were $^{131}\text{I}$ ($T_{1/2} = 8.02$ d), $^{134}\text{Cs}$ ($T_{1/2} = 2.065$ yr), $^{136}\text{Cs}$ ($T_{1/2} = 13.2$ d), $^{137}\text{Cs}$ ($T_{1/2} = 30.04$ yr) and $^{129m}\text{Te}$ ($T_{1/2} = 33.6$ d) (Chino et al., 2011; Tagami et al., 2011; Ohno et al., 2012; Yoshida and Takahashi, 2012). Radiocesium ($^{134}\text{Cs}$ and $^{137}\text{Cs}$) has a relatively long half-life and a high biological availability; and, consumption of food with radiocesium represents the principal route of human exposure to radiocesium (Zhu and Smolders, 2000). Therefore, the monitoring of radiocesium in agricultural products has been intensively performed by agencies within Fukushima Prefecture and by other organizations. Most of the brown rice (unpolished rice) produced during the 2011 growing season in Fukushima Prefecture had radiocesium at a concentration markedly lower than 100 Bq kg$^{-1}$ (on a fresh weight basis), which is much lower than the provisional regulation value in 2011 for food of 500 Bq kg$^{-1}$ established in 2011 (Ministry of Health, Labour and Welfare, 2012). Nonetheless, elevated radiocesium concentrations that exceeded the provisional guideline, were found in brown rice produced in some paddy fields located in northern part of Fukushima Prefecture (Saito et al., 2012). In order to provide countermeasures to reduce the radiocesium levels in future harvests of brown rice in Fukushima Prefecture, it is first necessary to find the source and causes of the higher radiocesium concentrations. In

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Abbreviations: FNPP, Fukushima Daiichi Nuclear Power Plant; RCEW, radiocesium-enriched water.
order to clarify the factors which influence the transfer of high levels of radiocesium to rice plants we have conducted studies using contaminated soils under several different conditions.

There are a number of factors that potentially influence the transfer of radiocesium to rice plants, although three major factors seem to be most important: Firstly, the exchangeable potassium (K) content of soils should have some influence on the radionuclide concentrations in rice. Both the exchangeable K content (Bilo et al., 1993; Zhu and Smolders, 2000) and the total K content (Tsukada et al., 2002) of soils correlate negatively with soil-plant transfer factors (TFs). TF is, by definition, expressed as the ratio of the radionuclide concentration in the plant (expressed in Bq kg$^{-1}$) to the radionuclide concentration in soil, (also expressed in Bq kg$^{-1}$ on a dry weight basis). Secondly, the sorption of Cs to clay minerals and that of radiocesium transferred to rice plant is affected by the amount of exchangeable Cs in the soil (Tsumura et al., 1984). Thirdly, the environment surrounding the paddy fields should also be considered. Many of the paddy fields that have higher radiocesium concentrations in the brown rice are located near forests or mountains. Radiocesium was deposited both on the tree canopies and in the leaf litter on the forest floor following the FNPP accident. If radionuclides dissolve in water and flow from the forests to paddy fields, the radiocesium concentrations of rice would likely increase.

In this study, we examined some hypothetical scenarios that can potentially lead to enhancement of radiocesium levels in brown rice in paddy fields, including, (i) low K content of the soil, (ii) low clay content of the soil, and (iii) the influx of radiocesium-enriched water (RCEW) into the paddy field from surrounding forests. For these investigations, we carried out rice cultivation experiments using pots containing contaminated soil under different growing conditions.

### Materials and Methods

Pot experiments were conducted in a glass house at Fukushima Agricultural Technology Centre, Fukushima, Japan using the rice cultivar “Hitomebore” (*Oryza sativa* L. cv. Hitomebore). Six seedlings at the third leaf stage were transplanted to Wagner pots (200 cm$^2$, 20 cm height) on 29 November 2011 in three replicated pots per treatment. The air temperature during the experiment was maintained at 23.9°C on the average.

Soil used in the experiment was collected from a paddy field located in Obama/Nihonmatsu, Fukushima Prefecture. The soil sample was collected from a particular paddy field, where brown rice had a relatively high radiocesium concentration (470 Bq kg$^{-1}$), in October 2011. Each pot was filled with 3.4 kg of the soil at the dry ratio of 0.88. The amount of exchangeable K and the proportions of clay, silt and sand in the soil before fertilizer incorporation are shown in Table 1.

Fertilizer applied was 0.1 g of nitrogen and phosphorus as basal dressing, and 0.3 g of nitrogen as topdressing. The amount of fertilizer on an area basis applied to the control was the same as that used by the local farmer. Rice plants were cultivated in pots with different treatments, and three pots were used for each treatment. During the cultivation, water was added when the depth of standing water became level with the soil surface. The amount of K (60.0 potassium chloride powder, National Federation of Agricultural Cooperative Associations), zeolite (less than 2 mm in diameter, Super Z, ZEEKLITE Co., Ltd.) and vermiculite (Gold vermiculite, ASAHI INDUSTRIES CO., LTD.) applied to each pot is shown in Table 2. Vermiculite was powdered using a mixer before incorporating into the soil. K, zeolite and vermiculite were incorporated into the soil before transplanting. RCEW that was used in three treatments (Table 2) was prepared as follows: Fallen leaves were gathered in Fukushima Prefecture in October 2011 and were soaked in water for 12 hr. Then the leachate was filtered through cloth to obtain water enriched with radiocesium. 2 L RCEW at the average 390 Bq kg$^{-1}$ was applied to a pot before transplanting and 11 L RCEW at the average 40 Bq kg$^{-1}$ was applied to a pot after transplanting until sampling. The average 137Cs concentration of RCEW applied during the experiment was 94 Bq kg$^{-1}$.

Three plants per pot were sampled at the heading stage (27 February 2012). Cultivation was carried out in autumn/winter and therefore the growth of the plants was poorer than normal, even though they were cultivated in a glasshouse. We did not allow the plants to reach maturity due to the lack of time in this study, since we wish to verify the effectiveness of such treatments immediately, in preparation for the upcoming rice-growing season, in the spring of 2012. The above-ground part (foliage) of each

### Table 1. Content of exchangeable potassium (K) and relative proportions of clay, silt and sand in soil.

| Soil type | Exchangeable K (cmol, kg$^{-1}$) | Proportion (%) | Soil classification |
|-----------|---------------------------------|----------------|---------------------|
| Gley soil | 0.07                            | 15.3 14.2 70.5 | Sandy Clay Loam     |

29 November 2011 in three replicated pots per treatment. The air temperature during the experiment was maintained at 23.9°C on the average.
The plant was dried for 48 hr at 80°C in a ventilated oven. Before the fertilizer incorporation 100 g of soil per pot was sampled in order to determine the concentrations of $^{137}\text{Cs}$. Concentrations of $^{137}\text{Cs}$ in plant and soil were determined by germanium detectors (Lead Shield Model747, CANBERRA and HPGe-Detector Type PGC3519, Dr. Westmeier Gmbh). The concentration of $^{137}\text{Cs}$ was shown on a dry weight basis. The detection limit was $25 \text{ Bq kg}^{-1}$ on a dry weight basis. The amount of absorbed $^{137}\text{Cs}$ by rice plants as shown in Table 3 was defined as the concentration of $^{137}\text{Cs}$ multiplied by dry weight of the foliage. The results summarized in Table 3 are expressed as an average of the values obtained for the triplicate samples of each treatment.

The data were statistically analyzed using ANOVA followed by Tukey's HSD test as a completely randomized design.

## Results

Table 3 shows the results obtained for the uptake of $^{137}\text{Cs}$ by rice plants grown in pots under different treatments. Concentrations of $^{137}\text{Cs}$ in soils before cultivation and biomass production in each pot are also listed in this table. In order to compare the transfer rate of radiocesium from soil to plant with the results in other pots, we have calculated TF, which is defined as "$^{137}\text{Cs}$ concentrations in plants" divided by "$^{137}\text{Cs}$ concentrations in soil".

### Table 3. Concentration of soil and rice plants and dry matter production in rice plants at heading stage under different treatments.

| Treatment                     | Concentration of $^{137}\text{Cs}$ (Bq kg$^{-1}$) | TF plant/soil | Dry weight of foliage (g per pot) | The amount of $^{137}\text{Cs}$ in foliage (Bq per pot) |
|-------------------------------|-----------------------------------------------|---------------|----------------------------------|----------------------------------------------------|
|                               | soil                                         | plant         |                                  |                                                   |
| Control                       | $1900 \pm 60 \text{ a}$                      | $230 \pm 27 \text{ bc}$ | 0.116                            | $16.7 \pm 2.4 \text{ b}$                           |
| High K                        | $2000 \pm 110 \text{ a}$                     | $90 \pm 12 \text{ de}$  | 0.045                            | $21.0 \pm 0.8 \text{ a}$                           |
| Zeolite at 0.5 kg m$^2$       | $2000 \pm 30 \text{ a}$                     | $140 \pm 12 \text{ d}$  | 0.065                            | $20.7 \pm 1.1 \text{ a}$                           |
| Zeolite at 1 kg m$^2$         | $2100 \pm 10 \text{ a}$                     | $50 \pm 10 \text{ e}$  | 0.024                            | $21.0 \pm 1.5 \text{ a}$                           |
| Zeolite at 5 kg m$^2$         | $1900 \pm 90 \text{ a}$                     | ND (25)       | < 0.013                          | $21.7 \pm 1.3 \text{ a}$                           |
| Vermiculite at 1 kg m$^2$    | $1800 \pm 140 \text{ a}$                    | $170 \pm 9 \text{ e}$  | 0.089                            | $20.2 \pm 1.6 \text{ a}$                           |
| Vermiculite at 5 kg m$^2$    | $1900 \pm 80 \text{ a}$                     | $70 \pm 6 \text{ e}$   | 0.037                            | $21.8 \pm 1.2 \text{ a}$                           |
| RCEW†                         | $2000 \pm 60 \text{ a}$                     | $780 \pm 10 \text{ a}$ | $16.0 \pm 0.6 \text{ b}$         | $12.5 \pm 0.47 \text{ a}$                          |
| High K with RCEW              | $1900 \pm 110 \text{ a}$                    | $310 \pm 26 \text{ b}$ | $21.0 \pm 0.3 \text{ a}$         | $6.5 \pm 0.64 \text{ b}$                           |
| Zeolite at 1 kg m$^2$ with RCEW | $2000 \pm 130 \text{ a}$                  | $250 \pm 22 \text{ b}$ | $21.1 \pm 0.9 \text{ a}$         | $5.3 \pm 0.61 \text{ bc}$                          |

$P$-value: 0.72 < 0.001 0.067 0.04 < 0.001

†RCEW: radiocesium-enriched water (see also text).

Concentrations of $^{137}\text{Cs}$ (on a dry weight basis) are shown as mean values of three pots together with standard error. ND indicates that $^{137}\text{Cs}$ was not detected and the number in parentheses indicates the detection limit. $P$-value was obtained by ANOVA. Values followed by the same letters ( a − f) are not different at the 5% level by Tukey’s HSD. The statistical analysis was conducted without results for the treatment with zeolite at 5 kg m$^2$, since $^{137}\text{Cs}$ was not detected at this level of zeolite addition.

TF is defined as $^{137}\text{Cs concentrations in plants}$ divided by $^{137}\text{Cs concentrations in soil}$.
study we use the term TF for not edible part in rice plant, although this is commonly applied if the plants had reached maturity and produced edible part (rice grains).

The application of K significantly increased dry weight of foliage when the rice plant was cultivated without RCEW. On the other hand, the amount of $^{137}$Cs uptake was decreased by the application of K although the difference was not significant. As a result, the application of K caused a significant decrease in the TF of rice plants. The TF of rice plants decreased from 0.116 to 0.045 when treated with high K. A similar tendency was observed when rice plant was cultivated with RCEW, i.e., the amount of $^{137}$Cs uptake was significantly decreased from 12.5 Bq per pot to 6.5 Bq per pot and the concentration of $^{137}$Cs was also markedly decreased as a result of the high K treatment.

Radiocesium uptake was markedly depressed in plants grown in pots without RCEW where zeolite was applied to the soil. The amount of $^{137}$Cs uptake by rice plants was decreased according to the amount of zeolite added to the pot. Since dry weight of foliage was significantly increased by the application of zeolite, the decreases in TF was rather large when compared with the decrease in the amount of $^{137}$Cs uptake. TF dropped from 0.116 in the control to 0.065, 0.024 and < 0.013 in the zeolite applications of 0.5, 1 and 5 kg m$^{-2}$, respectively. A similar tendency was observed when rice plant was cultivated with RCEW.

Similarly, the application of vermiculite also decreased the amount of $^{137}$Cs uptake by the rice plants. The effects of vermiculite application on the TF were also larger than those observed in the amount of $^{137}$Cs uptake due to the increase in dry weight of foliage by the application of vermiculite. TF observed in pots with the application of 5 kg m$^{-2}$ and 1 kg m$^{-2}$ vermiculite were 0.089 and 0.037, respectively.

The application of RCEW leached from leaf litter significantly enhanced the concentration of $^{137}$Cs in rice plants. The $^{137}$Cs concentration in rice plants applied with RCEW was increased by 140%, 140% and 300% in the control, high K and zeolite at 1 kg m$^{-2}$, respectively. We have not calculated TF for this treatment, because the source of radiocesium was not only in soil but also in water added several times during the cultivation. The rate of uptake of $^{137}$Cs derived from water was calculated as the difference between the amount of $^{137}$Cs observed in the rice plant with RCEW and that without RCEW divided by the amount of $^{137}$Cs applied with water during the experiment (1200 Bq per pot). We obtained an estimated rate of 0.7%, 0.4% and 0.4% in the control, high K and zeolite at 1 kg m$^{-2}$, respectively.

Discussion

The effect of applying K, zeolite and vermiculite as well as RCEW to paddy soils was evaluated, with the objective of evaluating factors responsible for high concentrations of radiocesium in rice produced in northern part of Fukushima Prefecture. Our results indicate that the TFs decrease through the application of K, zeolite and vermiculite. These results also suggest that, conversely, low-K and low sorption sites for Cs in paddy fields can potentially result in high radiocesium concentrations in rice. Therefore, application of these materials may be effective strategy in reducing the transfer of radiocesium to rice plants, including cases in which the soils are exposed to RCEW.

Our results are consistent with previous findings that the uptake of radiocesium decreases with increasing content of both exchangeable K (Bilo et al., 1993; Zhu and Smolders, 2000) and total K (Tsukada et al., 2002) in soils. Since Cs is a group-I alkali metal with chemical properties similar to K, plant roots uptake Cs through the K pathways (Qi et al., 2008). Therefore, the presence of potassium affects the uptake of cesium through the competitive effect in metabolic processes (Zhu and Smolders, 2000). The threshold of exchangeable K content has been reported to be around 0.2 cmol kg$^{-1}$ in cereals (Bilo et al., 1993). Immediately after the finding of the high radiocesium values in brown rice, radiocesium concentrations in rice were measured together with exchangeable K contents in soils of respective paddy fields. A negative correlation was found between the concentration of exchangeable K contents and the radiocesium concentrations in brown rice (Saito et al., 2012). The exchangeable K contents were 0.07 cmol kg$^{-1}$ in the soil used in this study. At this level of exchangeable K, applying K more than the control would result in decreasing the concentration of radiocesium in rice plants.

Zeolite and vermiculite decrease radiocesium uptake in plants (Sawhney, 1966; Paasikallio, 1999). The content of clay in the soil used in this study was markedly low (15.3%), while the sand content was high (70.5%). Therefore, there may be less Ce sorption sites in the soil used in this study. The addition of zeolite and vermiculite increased the Cs sorption sites, subsequently decreasing absorbability of radiocesium by rice plants. Zeolite and vermiculite also generally contain K, which should provide an additional inhibitory effect on the uptake of radiocesium by plants, as mentioned above. K dissolved from zeolite and vermiculite may have lowered the radiocesium uptake by rice plants in this study. The individual effect of K from zeolite and vermiculite on the exchangeable K content and the uptake of Cs by rice plants will be evaluated in near future.

Our results show that applying RCEW to the soil increases the concentration of radiocesium in rice plants. Higher radiocesium concentrations observed in brown rice produced in rice fields and terraces located near forests may be explained, in part, by this pathway. In the present study, RCEW at the $^{137}$Cs concentration of 94 Bq kg$^{-1}$ was applied to the soil and $^{137}$Cs concentration in rice
plants was increased by 200 – 550 Bq kg$^{-1}$. This suggested that water at the radiocesium concentration of 1.7 – 4.7 Bq kg$^{-1}$ could increase radiocesium concentration in rice plants by 10 Bq kg$^{-1}$. Attention should be paid to the influence of the radiocesium retained in forest ecosystems as a potential source of contamination of the rice paddy fields. Radiocesium concentrations in water flow into paddy fields should be measured during the cultivation.

The results obtained in this study indicate that low K and small number of sorption sites for Cs in paddy fields are responsible for the high concentrations of radiocesium observed in rice plants. Our work suggests that radiocesium derived from water will increase the uptake of radiocesium in rice plants. The applying K, zeolite and/or vermiculite should provide useful countermeasures in inhibiting the transfer of radiocesium from soil to rice plants.

**Acknowledgements**

The authors wish to thank Eurofins Food Testing Japan K.K., CREATERRA INC. Mr. M. Akiyama and Mr. T Yaginuma (Fukushima Agricultural Technology Center) for data collecting, and Mr. S. Fujita and Mr. Y. Suzuki (Fukushima Agricultural Technology Center) for discussion. We would like to express our thanks to Dr. G. Snyder (Rice University) for his critical reading of the manuscript, useful suggestions and improvement of English.

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