Chapter 17
Analysis of Factors Causing High Radiocesium Concentrations in Brown Rice Grown in Minamisoma City

Takashi Saito, Kazuhira Takahashi, Toshifumi Murakami, and Takuro Shinano

Abstract Despite a concentration of exchangeable K of >208 mg kg^{-1} dry weight in soil, the brown rice grown in Minamisoma City in 2013 had a higher concentration of radiocesium than the new Japanese standard (100 Bq kg^{-1}) for food. To analyze the factors affecting the radiocesium concentration in brown rice, we carried out pot tests using paddy soil and irrigation water collected in Minamisoma City. Rice seedlings were planted in 5-L pots containing Minamisoma soil, in which the exchangeable K was 125 mg kg^{-1} dry weight, and were irrigated with tap water or irrigation water collected in Minamisoma City. There was no difference in the Cs-137 concentration in brown rice between the two types of irrigation. Then we grew rice in the Minamisoma soil and two soils collected in Nakadori, Fukushima Prefecture. Cs-137 uptake in the Minamisoma soil was intermediate between the uptake rates in the Nakadori soils, showing that the Minamisoma soil was not special in radiocesium uptake. Finally, we grew rice in soil without radiocesium near the Fukushima Daiichi Nuclear Power Plant in 2014. Although the maximum value of Cs-137 in brown rice was 18 Bq kg^{-1}, below the standard, radiocesium was attached to the surface of the foliage.

Keywords Cs-137 • Brown rice • Exchangeable K • Irrigation water • Minamisoma City

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T. Takahashi (ed.), *Radiological Issues for Fukushima's Revitalized Future*,
DOI 10.1007/978-4-431-55848-4_17

17.1 Introduction

Following the accident at the Fukushima Daiichi Nuclear Power Plant on 11 March 2011, soils became contaminated with radiocesium (Cs-134 and Cs-137). To reduce the uptake of radiocesium by rice, growers have been adding potassium (K) fertilizer to their paddy fields, as the uptake of Cs decreases with increasing K concentration ([K]) in the soil [1] and the concentration of radiocesium in brown rice decreases at increasing concentrations of exchangeable K in the soil and K^+ in the soil solution [2]. K fertilization offers an effective and practical way to reduce radiocesium uptake by rice from several soil types [3]. Following the accident, the Food Sanitation Law of 2012 reduced the standard for the concentration of radiocesium in food to 100 Bq kg^{-1}. A concentration of >208 mg kg^{-1} dry weight of exchangeable K in soil is recommended for keeping the radiocesium content in brown rice below the standard [4].

However, despite a concentration of exchangeable K of >208 mg kg^{-1} in soil, the brown rice grown in Minamisoma City in 2013 exceeded the new standard [5].

One possible source is irrigation water. Dissolved radiocesium moves more easily into plants from water than from soil [6]. The concentration of dissolved radiocesium in irrigation water drawn from the Ota River in Minamisoma City was higher than that in other parts of Fukushima Prefecture [7].

One possible source is soil. Although Tsumura et al. [8] reported the relationship between the concentration of exchangeable K in soils and Cs-137 uptake in brown rice, however, discussion on radiocesium uptake in brown rice has not been done in same levels of exchangeable K in soil.

Another possible source is dust. The Cs-137 concentration ([Cs-137]) in dust collected in Futaba Town on 19 August 2013 was clearly higher than that at other times [9].

To identify the cause of the high radiocesium concentration in brown rice grown in Minamisoma City, we conducted pot experiments comparing sources of irrigation water and soil types. In addition, we determined the [Cs-137] of rice grown near the nuclear plant.

17.2 Materials and Methods

17.2.1 Irrigation Water

We collected 20 L of water on six dates (shown in Fig. 17.1) from the Ota River and passed it through 0.45-μm filters. Suspended matter collected on the filters was compressed into cylindrical polystyrene containers (i.d. 5.0 cm, o.d. 5.6 cm, height 6.8 cm) for analysis. The filtrates were concentrated to 2 L by heat and then placed in 2-L Marinelli beakers for analysis as described below. The concentrations of dissolved Cs-137 were 0.15–0.42 Bq L^{-1}, and those of suspended Cs-137 were 0.09–0.17 Bq L^{-1} (Fig. 17.1). [Cs-137] in tap water was 0.02 Bq L^{-1}.

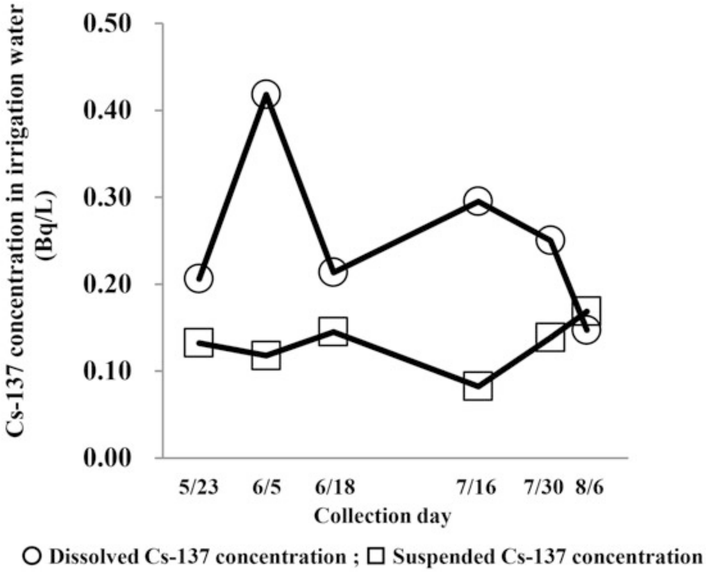


Fig. 17.1 [Cs-137] in irrigation water used for pot experiments. Error bars represent standard errors ($n = 3$). $*P < 0.05$ (Student's *t*-test) between tap water and irrigation water

Table 17.1 Chemical properties of soils used for pot experiment

				Exchangeable			
			Total C	K	Ca	Mg	CEC
Name	Soil types	pH(H_2O)	(g kg^{-1})	(mg kg^{-1})			(cmolc kg^{-1})
Minamisoma	Gray lowland soil	6.3	14	150	1640	390	9.1
Soil A	Andosol	5.6	79	25	1220	170	15.2
Soil B	Gray lowland soil	6.6	9.1	134	2160	553	11.4

17.2.2 Soils

Soils were collected from the field in April 2014. Experiment 1 used a contaminated Glay lowland soil collected from the top 15 cm of a paddy field in Minamisoma City where the concentration of radiocesium in brown rice grown in 2013 exceeded 100 Bq kg^{-1}.

Experiment 2 used three soils (Table 17.1): the Minamisoma Gray lowland soil; an Andosol collected from 0 to 15 and 15–30 cm depth in a paddy field in northern Fukushima Prefecture where the concentration of radiocesium in brown rice grown in 2011 exceeded 500 Bq kg^{-1} (soil A); and a Gray lowland soil collected from 0 to 15 and 15–30 cm depth in a paddy field at the Fukushima Agricultural Technology Centre (FATC), where the concentration of radiocesium in brown rice grown in 2011 was below the limit of quantification (<20 Bq kg^{-1}) (soil B).

Experiment 3 used an uncontaminated Gray lowland soil collected from the subsoil (beneath 15 cm depth) of a paddy field at FATC. The [Cs-137] in the soil was 35 Bq kg^{-1} dry weight.

17.2.3 Pot Experiments

Soils were air dried, thoroughly mixed, and passed through a 2-mm sieve. On 8 May 2014, rice seeds (*Oryza sativa* L. ‘Maihime’) were sown in granular culture soil. On 6 June, four seedlings were transplanted into each 5-L Wagner pot (diam. 16 cm, height 25 cm), which held 3.0 kg dry weight of soil. Each pot also received a basal dressing of 1.0 g of ammonium sulfate and calcium superphosphate and mixed into the soil. The water level was kept at a depth of 3–5 cm during the experiments. All experiments were conducted at FATC (and experiment 3 at Okuma Town also) under natural light. All treatments had three replicates. The rice plants were harvested on 9 October 2014, and samples of brown rice and leaves were oven dried at 40 °C for 24 h.

17.2.3.1 Experiment 1

The [Cs-137] in the contaminated Minamisoma soil was 1500 Bq kg^{-1} dry weight. No KCl was applied. Pots were watered with either tap water or irrigation water. Each pot received a total of 11–16 L during the experiment.

17.2.3.2 Experiment 2

The [Cs-137] in the contaminated Minamisoma soil was adjusted to 1500 Bq kg^{-1} as above. The [Cs-137] in soils A and B was adjusted to 1500 Bq kg^{-1} by mixing the topsoil and subsoil. The exchangeable K content of each was adjusted to 125 or 250 mg kg^{-1} with KCl. Pots were watered with irrigation water used in Experiment 1.

17.2.3.3 Experiment 3

The exchangeable K content of the uncontaminated soil was adjusted to 208 mg kg^{-1} with KCl. All plants were watered with tap water. Treatment pots were moved from the FATC to Okuma Town on 9 July and back to the FATC on 20 August. Control pots remained at the FATC. The pots were protected with a multi-film so that only the leaves were exposed to fallout. Radiocesium contaminations of rice foliage were analyzed by a gamma-ray spectrometry and autoradiography visually. In the autoradiogram analysis, powdered shoot (3 g) was put into a small polyethylene bag (10 × 7 cm) and the bag was put on the cardboard (40 × 20 cm, 0.6 mm thickness).

Markers made with potassium chloride (contain 10–26 mg) were attached on the corners of the cardboard samples to obtain a superposition of the autoradiogram and the visible image. An imaging plate (BAS-SR2040 (40 × 20 cm), Fuji-film, Japan) was contacted with the cardboard, and they were put into a paper case together and sandwiched between two lead plates of 4 mm thickness in a dark room. After 7 days exposure the imaging plate was scanned by image scanner (Typhoon FLA 7000, GE Healthcare Bio-Science Co., Ltd., USA) at a spatial resolution of 25 μm. The autoradiography and its visible image were overlapped on image processing software (Photoshop CS4 ver. 11.0, Adobe Co., Ltd., USA).

17.2.4 Soil and Plant Analyses

The chemical properties of the soils were analyzed according to the Editorial Boards of Methods for Soil Environment Analysis [10] (Table 17.1). Soil pH (H_2O) was measured at a soil-to-water ratio of 1:2.5 (w/w). The total carbon content was determined by dry combustion on a Sumigraph NC Analyzer NC-220 F (Sumika Chemical Analysis Service, Ltd., Osaka, Japan). Exchangeable K, calcium, magnesium were determined by the semi-micro Schollenberger method on an atomic absorption spectrophotometer (AA280FS; Varian Technologies Japan Ltd., Tokyo, Japan), and cation exchange capacity is calculated as the sum of these component ions.

The brown rice and leaf samples were compressed into cylindrical polystyrene containers as above, and the [Cs-137] was measured with a Ge gamma-ray detector connected to a multichannel analyzer (GC2020, GC3020, GC3520, GC4020, Canberra USA) for 36,000 s.

17.2.5 Statistical Analyses

Statistical analyses were performed in StatView 5.0 J software (SAS Institute, Berkeley, CA, USA). Analysis of variance (ANOVA) followed by *t*-test or Tukey's multiple comparison test was used to determine the significance of differences in a pairwise comparison matrix.

17.3 Results

17.3.1 Effect of Irrigation Water on Cs-137 Uptake in Rice

In both watering treatments (irrigation and tap water), the concentration of exchangeable K was 125 mg kg^{-1} before planting and 45 mg kg^{-1} after harvest (data not shown). When watered with irrigation water, [Cs-137] in brown rice and

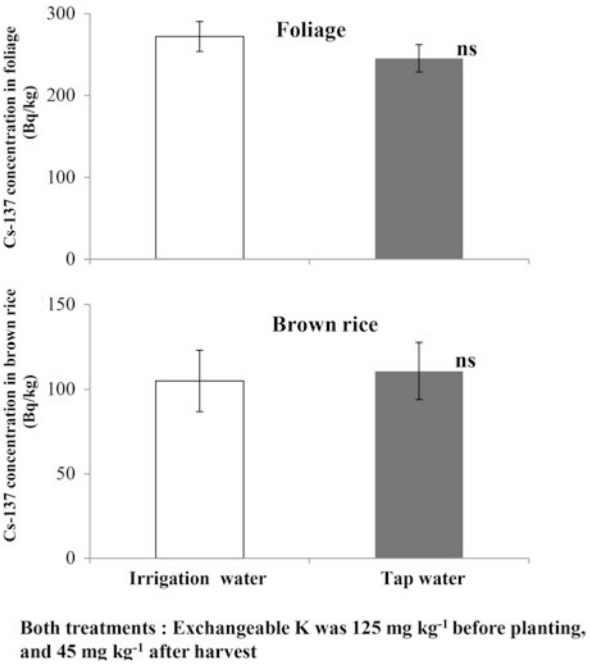


Fig. 17.2 [Cs-137] in foliage and brown rice of plants watered with irrigation or tap water. Error bars represent standard error ($n = 3$). ns, not significantly different; $*P > 0.05$ (Student's t-test) between irrigation water and tap water

foliage were 105 and 272 Bq kg^{-1} and watered with tap water were 111 and 245 Bq kg^{-1}, respectively (Fig. 17.2). There was no significant difference in the [Cs-137] of brown rice or foliage between treatments.

17.3.2 *Effect of Soil Type on Cs-137 Uptake in Rice*

When exchangeable K in Minamisoma soil of pot was 34.9 and 52.1 mg kg^{-1} after harvest, the [Cs-137] in the foliage of rice plant grown were 272 and 119 Bq kg^{-1}, respectively (Fig. 17.3). On the other hand, when exchangeable K in soil A of pot was 15.6 and 46.5 mg kg^{-1}after harvest, the [Cs-137] in the brown rice grown was 594 and 215 Bq kg^{-1}, respectively, and higher than that of Minamisoma soil. When exchangeable K in soil B of pot was 56.4 and 78.8 mg kg^{-1}, the [Cs-137] in the brown rice was 30.4 and 12.0 Bq kg^{-1}, respectively, and lower than that of the Minamisoma soil.

When exchangeable K in Minamisoma soil of pot was 34.9 and 52.1 mg kg^{-1} after cultivating, the [Cs-137] in the brown rice grown were 105 and 54.1 Bq kg^{-1}, respectively. On the other hand, when exchangeable K in soil A of pot was 15.6 and 46.5 mg kg^{-1}, the [Cs-137] in the brown rice grown was 206 and93.8 Bq kg^{-1},

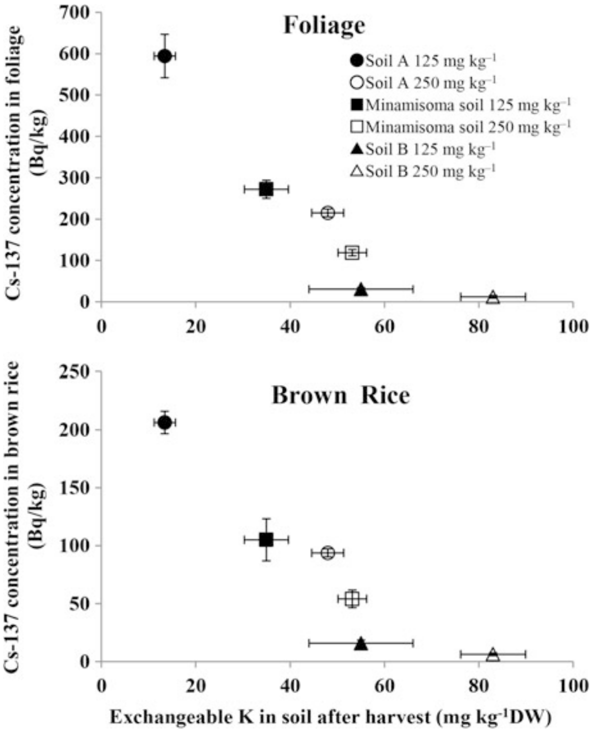


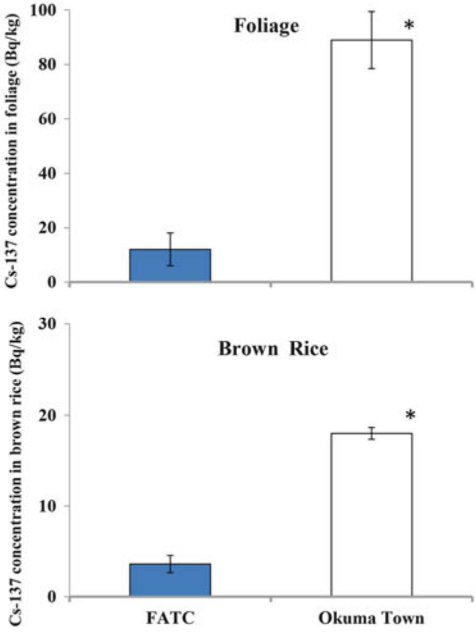
Fig. 17.3 Relationship between [Cs-137] in rice and exchangeable K after harvest in different kind soils or in different K soils. Error bars represent standard errors ($n = 3$)

respectively, and higher than that of the Minamisoma soil. When exchangeable K in soil B of pot was 56.4 and 78.8 mg kg^{-1}, the [Cs-137] in the brown rice was 15.8 and 6.3 Bq kg^{-1}, respectively, and lower than those of Minamisoma soil.

17.3.3 Effect of Site on Acquisition of Cs-137 by Foliage and Brown Rice

There was no significant change in the amount of exchangeable K in either treatment before and after the experiment (data not shown). The [Cs-137] in the brown rice and leaves of plants grown at Okuma Town for 6 weeks was 5–7 times that in the plants kept at the FATC (Fig. 17.4).

Autoradiographs of brown rice and foliage revealed no contamination of either in plants grown at the FATC or in brown rice of plants grown at Okuma Town (Fig. 17.5). However, foliage of plants grown at Okuma Town showed radioactive contamination (Fig. 17.6).



FATC : Exchangeable K was 208 mg kg^{-1} before planting, and 63 mg kg^{-1} after harvest.

Okuma Town: Exchangeable K was 208 mg kg^{-1} before planting, and 67 mg kg^{-1} after harvest.

Fig. 17.4 Translocation of Cs-137 from foliage to brown rice between two sites. Error bars represent standard errors ($n = 3$). *$P < 0.05$ (Student's t-test) between TATC and Okuma Town

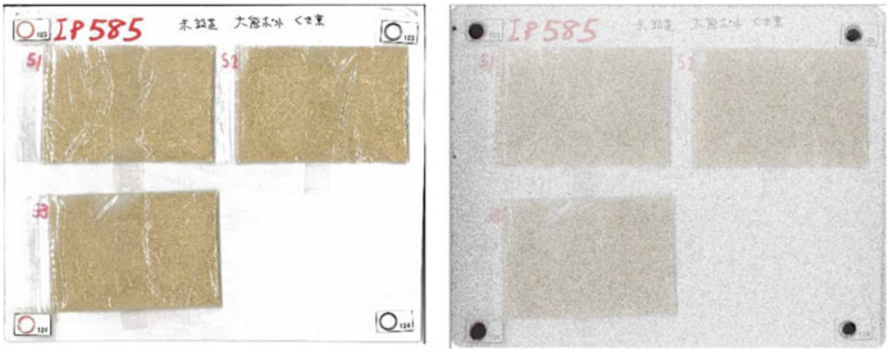


Fig. 17.5 Autoradiographs of foliage rice by FATC

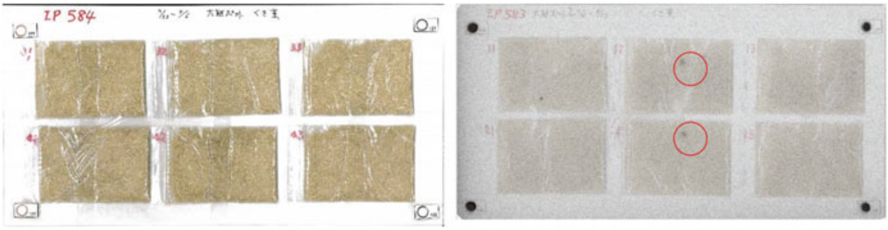


Fig. 17.6 Autoradiographs of foliage rice by Okuma Town

17.4 Discussion

17.4.1 Effect of Irrigation Water on Cs-137 Uptake in Rice

In hydroponic culture with 0.1, 1.0, or 10 Bq L^{-1} Cs-137, the [Cs-137] in brown rice increased with increasing [Cs-137] in the culture solution [6]. However, nutrient uptake can be greater in hydroponic culture than in soil culture because all nutrients are in solution. In contrast, a high level of exchangeable K in soil can limit the transfer of Cs-137 from irrigation water containing low levels of Cs-137 (0.1–1.0 Bq L^{-1}) [11]. We found no significant difference in uptake between water sources; therefore, low levels of exchangeable K in both soils limited Cs-137 uptake by rice plants.

17.4.2 Effect of Soil Type on Cs-137 Uptake in Rice

The Minamisoma soil did not have superior ability to promote Cs-137 uptake (Fig. 17.3). To reduce the [Cs-137] below 100 Bq kg^{-1} would require 40 mg kg^{-1} of exchangeable K in the Minamisoma soil and 50 mg kg^{-1} in soil A. Thus, at a similar level of exchangeable K in the soil, Cs-137 uptake depended on soil type. The high carbon content and low clay content of soil A may have helped to inhibit Cs-137 uptake. Therefore, the Minamisoma soil was not the cause of high [Cs-137] in 2013.

17.4.3 Effect of Site on Acquisition of Cs-137 by Foliage and Brown Rice

The [Cs-137] of rice plants increased greatly during 6 weeks' culture in Okuma Town. Cs-137 derived from the Fukushima accident is still distributed widely around the town. Thus, the rice plants could have taken up more radiocesium from the outside environment.

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