Chapter 4 Cesium Accumulation in Paddy Field Rice Grown in Fukushima from 2011 to 2013: Cultivars and Fertilization

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Abstract After the accident at the Fukushima Daiichi Nuclear Power Plant, radioactive cesium (Cs) was released and the agricultural fields in Fukushima were contaminated. It became important to obtain data for radioactive Cs accumulation in rice grown in contaminated fields. We conducted a 3-year investigation in a Fukushima paddy field of radioactive Cs concentrations in various rice cultivars, and in two commercial rice cultivars grown under four different nutrient conditions. Our studies demonstrated substantial variation in radioactive Cs concentrations among the rice cultivars, and an increase in radioactive Cs concentrations in straw and brown rice under high nitrogen and low potassium conditions. Our 3-year investigations of radioactive Cs-contaminated rice in Fukushima paddy field shows that the rice grown in Fukushima is now well-monitored and contains much less than the allowed levels of radiation (100 Bq kg⁻¹).

Keywords Radioactive cesium • Straw • Brown rice • Rice cultivars • Fertilizer effect • Fukushima paddy field

4.1 Introduction

The accident at the Fukushima Dai-ichi Nuclear Power Plant in March 2011 released radionuclides to the broader area including the paddy fields around the nuclear power plant. The radioactive isotopes of cesium (Cs) have relatively long half-lives among the released radionuclides (2.06 years for ¹³⁴Cs and 30.2 years for ¹³⁷Cs) (Matsumura et al. 2011). Contamination of agricultural products by radioactive Cs will thus be a serious problem for a long time. We have previously reported the accumulation of radioactive Cs among different rice cultivars, and the

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effects of fertilizer on the accumulation of radioactive Cs in rice (Ohmori et al. 2014a, b). Here, we review these reports.

Cesium is an alkali metal, which is absorbed from the soil by roots and transported to various parts of rice plants, such as the brown rice and straw, which are served as foods for human and livestock, respectively. To reduce the Cs accumulation in rice, we need to understand the mechanism of Cs uptake and transportation in rice plants. Cs accumulation in rice is thought to be determined by both genetic and environmental factors. We measured the radioactive Cs concentration in 85 rice cultivars to find the genetic factors, and we investigated the effects of fertilizer on radioactive Cs accumulation in rice plants to reveal the environmental factors.

4.2 Difference in Radioactive Cesium Accumulation Among Rice Cultivars Grown in the Paddy Field at Fukushima from 2011 to 2013

4.2.1 Radioactive Cesium Accumulation Among 85 Rice Cultivars Grown in Fukushima Paddy Fields in 2011

Cesium is an alkali metal that is not essential to plant growth, but is toxic. Potassium (K) also belongs to alkali metal group, and it is an essential element for plant growth. It is believed that both Cs uptake and transport are mediated by K transporters. In *Arabidopsis*, one of the KUP/HAK/KT type transporters, AtHAK5, plays a role in non-radioactive cesium (133 Cs) absorption under low K conditions (Qi et al. 2008). In addition, *AtCNGC1* is a candidate gene for determining the natural variation of Cs concentrations (Kanter et al. 2010). However, the whole mechanism of Cs uptake and transport remains unclear.

To reduce Cs accumulation in rice, it is crucial to understand that there is variation in Cs uptake among different rice cultivars. The difference in ¹³³Cs concentrations in brown rice among different rice cultivars has been reported (Yamaguchi et al. 2012), and the concentration of ¹³⁷Cs in rice grown in Aomori Prefecture before the Fukushima accident has also been reported (Tsukada et al. 2002). However, the amount of radioactive Cs fallout from the Fukushima Dai-ichi Nuclear Power Plant after the earthquake in 2011 was much higher than that derived from past fallout. Thus, a reinvestigation of the accumulation levels of radioactive Cs in rice is needed in the Fukushima area.

We selected 85 rice cultivars from the World Rice Core Collection (WRC), the Japanese Rice Landrace Mini Core Collection (JRC), and other domestic varieties (Table 4.1). The WRC consists of 67 varieties and covers 91 % of the genetic variation in about 37,000 rice landraces. The JRC consists of 50 varieties and covers 87.5 % of genetic variation in about 2000 Japanese rice landraces (Ebana et al. 2008; Kojima et al. 2005). We planted the 85 rice cultivars in the Fukushima paddy field on May 31st, 2011, and harvested them on September 23rd, October

Number	Cultivar name	Number	Cultivar name
1	Karahoushi	44	Kabashiko
2	Houmanshindenine	45	Jamaica
3	Mansaku	46	Shichimenchou Mochi
4	Himenomochi	47	Khauk Yoe
5	Akage	48	Tachisugata
6	Hassokuho	49	Shiroine
7	Kahei	50	Mizuhochikara
8	Shinyamadaho 2	51	Akamai
9	Aichiasahi	52	Kusanohoshi
10	Hamasari	53	Sekiyama
11	Hakamuri	54	Fukoku
12	Shinriki Mochi	55	Shinriki
13	Raiden	56	Leaf Star
14	Puluik Arang	57	Chinya
15	Ginbouzu	58	Gaisen Mochi
16	Vary Futsi	59	Meguro Mochi
17	Ishijiro	60	Senshou
18	Nipponbare	61	Moritawase
19	Mogumoguaoba	62	Momiroman
20	Bekogonomi	63	Yamada Baka
21	Nishiaoba	64	Hetadawee
22	Nagoyashiro	65	Taichung 65
23	Aikoku	66	Mack Kheua
24	Kameji	67	Rikutou Rikuu 2
25	Yumeaoba	68	Omachi
26	Moroberekan	69	Chinsurah Boro 2
27	Hosogara	70	Hiyadachitou
28	Kasalath	71	Okka Mososhi
29	Kyoutoasahi	72	Daw Dam
30	Co 13	73	Deng Pao Zhai
31	Tachiaoba	74	Basilanon
32	Bekoaoba	75	Koshihikari
33	Kusahonami	76	Badari Dhan
34	Dango	77	Hoshiaoba
35	Tupa 121-3	78	Asominori
36	Hirayama	79	Kaneko
37	Naba	80	Oiran
38	Hinode	81	Khau Mac Kho
39	Muha	82	Joushuu
40	Touboshi	83	Wataribune
41	Bouzu Mochi	84	Iruma Nishiki
42	Fukuhibiki	85	Shinshuu
43	Okabo		

 Table 4.1
 List of the 85 rice cultivars tested in this study

ultivars grown at Fukushima in 2011	rice	n (SD) Median Range	(4.7) 6.7 0.7 - 20.3	$(5.8) \qquad 10.2 \qquad 2.7 \smile 26.6$
n in different rice culti	Cs concentration in b		¹³⁴ Cs (Bq/kg)	¹³⁷ Cs (Bq/kg) 1
cesium concentration		Range	$19.4 \sim 73.4$	$10.3 \smile 100.3$
n, and range for		Median	35.8	35.5
riation), media		(SD)	(13.5)	(17.2)
(standard dev	in straw	Mean	38.9	39.0
Table 4.2 Mean	Cs concentration		¹³⁴ Cs (Bq/kg)	¹³⁷ Cs (Bq/kg)

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4th, and October 18th. The radioactive Cs concentrations in harvested rice straw and brown rice were independently determined. The ¹³⁴Cs and ¹³⁷Cs concentrations in the straw were 19.4–73.4 and 10.3–100.3 Bq kg⁻¹, respectively (Table 4.2). In addition, the mean concentrations were 38.9 and 39.0 Bq kg⁻¹, respectively, and the medians were 35.8 and 35.5 Bq kg⁻¹, respectively (Table 4.2). In brown rice, the ¹³⁴Cs and ¹³⁷Cs concentrations were 0.7–20.3 Bq kg⁻¹ and 2.7–26.6 Bq kg⁻¹, respectively (Table 4.2). The means were 8.1 and 11.6 Bq kg⁻¹, respectively, and the medians were 6.7 and 10.2 Bq kg⁻¹, respectively (Table 4.2). Both the straw and brown rice from the selected rice cultivars showed a large variation in radioactive Cs concentration. This variation can be used to isolate either Cs uptake or transport-related factors.

Next, we correlated the Cs concentration between straw and brown rice among the 85 rice cultivars. Both ¹³⁴Cs and ¹³⁷Cs concentrations correlated significantly and positively between straw and brown rice (Fig. 4.1), that are $p = 1.2 \times 10^{-6}$ and

Fig. 4.1 Correlation diagram for Cs concentration. (a) Straw versus brown rice for 134 Cs. (b) Straw versus brown rice for 137 Cs. The corresponding coefficients of determination (\mathbb{R}^2) are shown. The *black line* represents the linear regression line corresponding to the least square adjustment of all the data



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 $p = 4.9 \times 10^{-7}$ for ¹³⁴Cs and ¹³⁷Cs, respectively (Fig. 4.1). The coefficients of determination (R²) were 0.33 and 0.35 for ¹³⁴Cs and ¹³⁷Cs, respectively. Thus, we concluded that the Cs concentrations in brown rice might be estimated from the Cs concentrations in straw, although there were some exceptions.

4.2.2 Radioactive Cesium Accumulation Among 15 Selected Rice Cultivars Grown in a Fukushima Paddy Field in 2012 and 2013

On the basis of the Cs concentration of brown rice in 2011, we selected 15 rice cultivars to test the reproducibility of Cs uptake, and planted them at a Fukushima paddy field in 2012 and 2013. Khau Mac Kho, Asominori, Kaneko, and Deng Pao Zhai were selected as high Cs accumulating cultivars; whereas, Kasalath, Hamasari, Kameji, Aichiasahi, Wataribune, Mansaku, Akage, and Hassokuho were selected as low Cs accumulating cultivars. In addition, Koshihikari, Nipponbare, and Taichung 65 were selected as typical Japanese cultivars. In 2012, we planted the selected 15 rice cultivars at a Fukushima paddy field on May 23rd, and sampled them on October 13. In 2013, the planting and harvesting dates were May 14th and October 10th, respectively. Khau Mac Kho, Asominori, and Deng Pao Zhai showed relatively higher concentrations of ¹³⁷Cs in brown rice among different rice cultivars (Fig. 4.2). On the other hand, Hamasari, Aichiasahi, and Mansaku showed relatively lower ¹³⁷Cs concentrations in brown rice compared with the other cultivars (Fig. 4.2). These results were comparatively conserved in 3-year investigations.

Our results significantly provide data for Cs accumulation levels among different rice cultivars in a Fukushima paddy field. A molecular genetic approach to rice cultivars with different Cs accumulation may enable identification of genes that regulate Cs uptake and transportation in rice.

4.3 Fertilizer Effects on Cs Accumulation in Rice

4.3.1 General Information of Fertilizer Effects on Cs Accumulation in Plants

Both K and Cs are alkali metals, and Cs transportation is known to be mediated by several K transporters (Qi et al. 2008; Jabnoune et al. 2009). Thus, Cs uptake and transportation by K transporters compete with K uptake and transportation in plants. It has been reported that Cs uptake is enhanced under low K conditions in various plant species (Shaw 1993). K fertilizer applications can reduce Cs absorption in crops such as wheat, barley, rye, and potato under K deficient conditions

Fig. 4.2 Comparison of 137 Cs concentrations among 2011–2013 data in straw and brown rice. (a) 137 Cs concentrations (Bq kg⁻¹) in straw from selected cultivars. (b) 137 Cs concentrations (Bq kg⁻¹) in brown rice from selected cultivars. *Blue, red,* and *green boxes* indicate 2011, 2012, and 2013 data, respectively. Means and standard deviations are shown (n = 3)



(Lemmbrechts 1993). On the other hand, fertilizer application has little effect on Cs absorption under adequate K conditions.

Ammonium (NH_4^+) is known to affect the elution of Cs from soil by replacing NH_4^+ with Cs⁺. Therefore, high concentrations of NH_4^+ in the soil enhance Cs⁺ elution, resulting in the promotion of Cs⁺ absorption by plants. It has been reported that the application of nitrogen fertilizer enhances Cs uptake by plants in the field, although the degree of this effect depends on the soil type and other conditions (Lemmbrechts 1993; Smolders et al. 1997).

Before March 11, 2011, studies on radioactive Cs accumulation had been mainly conducted at the site of the Chernobyl Nuclear Power Plant accident in Russia.

Therefore, the behavior of radioactive Cs affected by fertilizers in paddy fields and andosols, which are the common field condition and soil-type in Japan, were not well elucidated. Thus, it is important to investigate the effects of K and N fertilizer on radioactive Cs absorption in rice grown in Japanese paddy fields.

In the next chapter, we will describe radioactive Cs concentrations in rice grown in paddy fields under four different fertilizer conditions at Ishidairayama, Yamakiya, Kawamata-cho in Fukushima in 2011 and 2012.

4.3.2 Radioactive Cs Concentrations in Rice Grown in Paddy Fields Under Four Different Fertilizer Conditions at Fukushima

To investigate the effect of fertilizer applications on radioactive Cs concentrations in rice, we cultured two commercial rice cultivars, Koshihikari and Hitomebore, in 2011 and 2012 under four different fertilizer conditions: normal, -K, -K + 2N, and no fertilizer. Under normal condition, a commercial fertilizer containing 8:18:16 (N:P:K; equivalent to 6, 9, and 8 kg per 10 a) was applied as a basal fertilizer. Under K-depleted conditions, N and P were given as urea and monocalcium phosphate, respectively. Under -K condition, N, P, and K were given as 6, 9, and 0 kg per 10 a, respectively. Under -K + 2N condition, N, P, and K were given as 12, 9 and 0 kg per 10 a, respectively. Under no fertilizer condition, no fertilizers were applied.

4.3.2.1 Radioactive Cs Concentration in Rice Straw Grown in a Paddy Field at Kawamata-cho

To assess the effect of fertilizer conditions on radioactive Cs concentrations in rice, we determined the radioactive Cs (134 Cs and/or 137 Cs) concentrations in straw harvested at the ripening stage.

In 2011, the ¹³⁴Cs concentration in straw under the -K + 2N condition was 1.5 times higher than that under the normal condition (Fig. 4.3a). The ¹³⁴Cs concentrations in straw were also high under the -K condition compared to those under the normal condition (Fig. 4.3a). On the other hand, there was no difference in the ¹³⁴Cs concentrations in straw under the normal and no fertilizer conditions. Similar trends were also observed for the ¹³⁷Cs concentrations (Fig. 4.3b).

In 2012, we replanted Koshihikari and Hitomebore at the same paddy field and investigated the reproducibility of the ¹³⁷Cs concentrations in the straw. The ¹³⁷Cs concentrations in the straw were highest under the -K + 2N condition (Fig. 4.3c). The ¹³⁷Cs concentrations in straw were also high under the -K condition (Fig. 4.3c). Under the no fertilizer condition, the ¹³⁷Cs concentrations in straw were similar to those under the normal fertilizer condition. All patterns of



Fig. 4.3 Cesium concentrations in straw under different fertilizer conditions in 2011 and 2012. (a) 134 Cs concentrations in 2011. (b) 137 Cs concentrations in 2011. (c) 137 Cs concentrations in 2012. *White and black boxes* indicate Koshihikari and Hitomebore, respectively. Means and standard deviations are shown (n = 3). The concentrations are presented on a dry-weight basis. Normal, the normal fertilizer condition; -K + 2 N, twofold nitrogen without potassium condition; -K, normal nitrogen and phosphorus but no potassium condition; and no, no fertilizer condition

radioactive Cs concentrations in rice straw were similar between Koshihikari and Hitomebore in 2011 and 2012 (Fig. 4.3).

4.3.2.2 Radioactive Cs Concentrations in Brown Rice Grown in a Paddy Field at Kawamata-cho

We determined the radioactive Cs concentrations in brown rice grown at Kawamata-cho in 2011 and 2012. In 2011, the trends for radioactive Cs accumulation in brown rice were very similar between ¹³⁴Cs and ¹³⁷Cs (Fig. 4.4a, b). The radioactive Cs concentrations in brown rice were highest under the -K+2N condition, being about twice that under the normal condition. Under the -K condition, the radioactive Cs concentrations in brown rice were also higher than those under the normal condition in Koshihikari (Fig. 4.4a, b). The radioactive Cs concentrations in brown rice were also higher than those under the normal condition in Koshihikari (Fig. 4.4a, b). The radioactive Cs concentrations in brown rice were the lowest under the no fertilizer condition.

In 2012, the trends of ¹³⁷Cs accumulation in brown rice under the four different fertilizer conditions were very similar to those observed in 2011 (Fig. 4.4). The¹³⁷Cs concentrations in brown rice were highest under the -K + 2N condition, and lowest under the no fertilizer condition both in 2011 and 2012 (Fig. 4.4b, c).



Fig. 4.4 Cesium concentrations in brown rice under different fertilizer conditions in 2011 and 2012. (a) ¹³⁴Cs concentrations in 2011. (b) ¹³⁷Cs concentrations in 2011. (c) ¹³⁷Cs concentrations in 2012. *White and black boxes* indicate Koshihikari and Hitomebore, respectively. Means and standard deviations are shown (n = 3). Fertilizer conditions are the same as described in the legend of Fig. 4.3

However, in contrast to the results of 2011, the 137 Cs accumulation in brown rice under the -K condition in 2012 was no different to that under the normal condition (Fig. 4.4c).

In our study, low K conditions tended to increase the radioactive Cs concentrations both in straw and brown rice grown in the contaminated paddy field at Kawamata-cho in Fukushima. This result may be caused by chemical competition between K and Cs. In addition, it is noteworthy that nitrogen fertilizer affects radioactive Cs concentrations in rice. The fertilizer condition that caused the highest radioactive Cs concentration in rice was -K + 2N in both 2011 and 2012. This result suggests that not only K fertilizer, but also N fertilizer affects radioactive Cs concentrations in rice grown in a Japanese paddy field.

The mechanism that increases radioactive Cs concentrations in rice by N fertilizer application is still unknown. One hypothesis is that N fertilizers elute radioactive Cs from the soil surface and enhance the radioactive Cs uptake by rice. To avoid unexpectedly high-levels of radioactive Cs in rice (over 100 Bq kg⁻¹; the governmental new safety standards for radioactive Cs in food products in Japan), we may need to implement the N and K fertilizer conditions.

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