Removal of Radiocesium from Food by Processing: Data Collected after the Fukushima Daiichi Nuclear Power Plant Accident – 13167

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ABSTRACT

Removal of radiocesium from food by processing is of great concern following the accident of TEPCO's Fukushima Daiichi Nuclear Power Plant accident. Foods in markets are monitored and recent monitoring results have shown that almost all food materials were under the standard limit concentration levels for radiocesium (Cs-134+137), that is, 100 Bq kg^{-1} in raw foods, 50 Bq kg^{-1} in baby foods, and 10 Bq kg^{-1} in drinking water; those food materials above the limit cannot be sold. However, one of the most frequently asked questions from the public is how much radiocesium in food would be removed by processing. Hence, information about radioactivity removal by processing of food crops native to Japan is actively sought by consumers. In this study, the food processing retention factor, F_r, which is expressed as total activity in processed food divided by total activity in raw food, is reported for various types of corps. For white rice at a typical polishing yield of 90-92% from brown rice, the F_r value range was 0.42-0.47. For leafy vegetable (indirect contamination), the average F_r values were 0.92 (range: 0.27-1.2) after washing and 0.55 (range: 0.22-0.93) after washing and boiling. The data for some fruits are also reported.

INTRODUCTION

Large releases of radionuclides occurred in Japan after the 2011 accident at TEPCO's Fukushima Daiichi Nuclear Power Plant (FDNPP). Consequently, the potential radiation effects due to contaminated food ingestion are of great concern to the public. In order to show how much radiocesium is retained after food processing, the IAEA uses the food processing retention factor (F_r), which is expressed as total activity in processed food divided by total activity in raw food [1]. The percentage of radiocesium removal by food processing can be estimated from these values. However, the reported ranges are wide, and most of the data are from European and North American countries; there are not enough data on agricultural crops that are frequently consumed in Asian countries, including Japan. Therefore, it is necessary to collect data for the Japanese situation. In this study, therefore, we measured F_r values using food crops grown in Japan.

MATERIALS AND METHODS

In this study, food processing data for paddy rice, root crops (turnip and radish), fruits (persimmon, apple and loquat) and nuts (chestnut and peanut) were collected by measuring Cs-134 and Cs-137 using a Ge detecting system (Seiko EG&G). Raw materials were mainly collected from our institute fields in Chiba Prefecture and some sites in Fukushima Prefecture. Information on sampling sites and processing methods has been reported elsewhere [2-4].

The food processing retention factor F_r is expressed as

$$F_r = \text{total activity in processed food / total activity in raw food}$$

and F_r can be calculated using the following equation as reported in IAEA-TECDOC-1616 [1]:

$$F_r = P_f \text{ x } P_e$$

where P_f is the ratio of the element activity concentrations and P_e is the ratio of the raw weight of the processed food to the weight of the original raw material.

The F_r cannot exceed 1 if a raw sample for radioactivity measurement is directly used for food processing. However, in this study, most raw samples had low radiocesium concentrations, so we separated each collected sample into three portions, i.e. raw, washed, and washed+processed samples to obtain concentrated samples after drying, separately. Thus, F_r sometimes exceeded 1. Both Cs-134 and Cs-137 data were obtained and the average value for each sample was used in this study.

RESULTS AND DISCUSSION

F_r values for rice

White rice is generally consumed in Japan, however, food monitoring has been carried out on a brown rice basis; it is well known that Cs concentration is high in rice bran [5, 6]. Previously F_r values were obtained for rice samples collected in Fukushima Prefecture in 2011 after polishing and washing the rice [2], and the results are summarized in Table I. In Japan, traditional rice cooking method is absorption method with boiling and white rice is used usually after washing and soaking. The polishing rate from brown rice to white rice is 90-92%; that is the P_e value is 0.90-0.92. The concentration ratio P_f observed was 0.49 on average (range: 0.46 - 0.52) and thus the average F_r value was 0.45 (range: 0.42-0.47). According to ref. [7], which compiles radionuclide removal rates by food processing and includes Japanese data, the Cs removal rate by polishing to white rice was 57-80% with 65% on average, which means that the F_r value was 0.35. When white rice was washed, the F_r value was 0.56 on average.

During the typical white rice cooking, the rice weight increases 2.3 times due to water absorption [8], and radiocesium is not removed by this process. Thus when brown rice with 100 Bq kg^{-1} (as radiocesium concentration) is used to make cooked white rice, after polishing at 90-92% and then washing the white rice, it contains 26 Bq kg^{-1} before cooking, but after cooking, the weight increases and thus the concentration is about 12 Bq kg^{-1}, which is much lower than the standard limit level.

TABLE I. Food processing efficiency (P_e), food concentration ratio (P_f) and food processing retention factor (F_r) of radiocesium for rice (average).

Process	Material	P_e	P_f	F_r
Polishing	Brown rice	0.92	0.49	0.45
Washing	Brown rice	0.99	0.94	0.93
Washing	White rice	0.96	0.59	0.56

F_r values for vegetables

Table II summarizes data obtained in this study as well as our other recent data obtained after the FDNPP accident. The F_r and P_e values are listed for washing, washing and boiling, pickling and peeling of different types of vegetables. When we compared the results for both washing and boiling for leafy vegetables contaminated indirectly or directly, F_r values were usually small for directly contaminated samples. For example, by washing, the average F_r for indirect contaminated leafy vegetables was 0.92 and that for directly contaminated leafy vegetables was 0.75. By washing + boiling for 2.5 minutes, the average F_r for indirectly contaminated leafy vegetables was 0.55 and that for directly contaminated leafy vegetables was 0.35. The differences between direct and indirect contamination pathways were possibly seen because surface contamination was easily removed by both methods.

F_r values have been compiled by Kashparove *et al.* [1] using published data collected mainly after the Chernobyl nuclear power plant accident; F_r by washing vegetables, berry, and fruits (all data) ranged from 0.6-1.0, and that for direct deposition (outside) ranged from 0.1-0.9. The F_r by boiling ranged from 0.4-0.9 for all data and 0.1-0.5 for direct deposition. Thus the tendency observed in our study was the same as that observed before the FDNPP accident.

There is a tendency that F_r values of Cs in thinner plant body crop types, e.g. leafy vegetables, were lower than those of thicker plant body crop types, e.g. root crops. Boiling of bamboo shoot and turnip (indirect pathway) resulted in higher F_r values, that is, 0.70 and 0.82, respectively, compared to those of leafy vegetables, 0.55 on average. Fig. 1 compares values for (a) washing and (b) washing + boiling processes for Japanese butterbur leaves and petioles (n=5 for each, indirect pathway). Petioles were about 1-2 cm in diameter. Although F_r values after washing did not differ between leaves and petioles, washing + boiling process (plus peeling for petiole samples), geometric mean of F_r for leaves of Japanese butterbur was 0.37, while that for petioles was 0.87. Therefore, we thought that the boiling time as well as the crop thickness would affect radiocesium removal rates.

Table II. Food processing efficiency (P_e), and food processing retention factor (F_r) of radiocesium for vegetables

Sample	Contamination pathway	Process	N	P_e (Range)	F_r (Range)	Ref.
Leafy vegetables	Indirect	Washing	14	1.0	0.92 (0.27-1.2)	[3]
Leafy vegetables	Direct	Washing	8	1.0	0.75 (0.49-1.2)	[4]
Turnip	Indirect	Washing	1	1.0	0.48	This study
Bamboo shoot	Indirect	Washing + 10-15 min Boiling	4	1.0	0.70 (0.67-0.74)	[3]
Leafy vegetables	Indirect	Washing + 2.5 min Boiling	22	0.96 (0.83-1.1)	0.55 (0.22-0.93)	[3]
Leafy vegetables	Direct	Washing + 2.5 min Boiling	6	0.96 (0.77-1.1)	0.35 (0.15-0.58)	[4]
Turnip	Indirect	Washing + 2.5 min Boiling	1	0.94	0.82	This study
Garlic	Indirect	Pickling in soy sauce, 2 months	1	0.93	0.38	[3]
Someiyoshino cherry leaves	Indirect	Pickling in salt, 3-5 days	2	Not measured	0.18 (0.13-0.22)	[3]
Turnip	Indirect	Pickling in 9% (w/w) salt, 14 h	2	0.69 (0.64-0.73)	0.49 (0.32-0.65)	This study
Japanese radish	Indirect	Peeling	1	0.89	0.16	This study

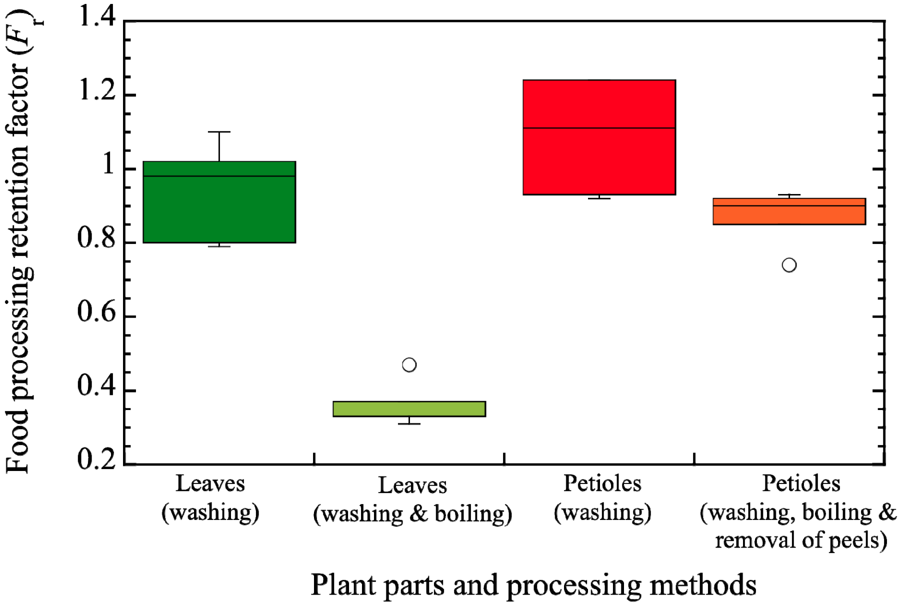

Fig. 1. F_r values for Japanese butterbur leaves and petioles (indirect contamination pathway).

In pickling, ion exchange of Cs^+ and K^+ by Na^+ would occur [9], thus, Cs concentration decreased. However, it was difficult to remove all of the radiocesium by this process. According to the data compiled by Kashparove et al. [1], F_r values ranged from 0.1-1 for picked vegetables; the efficiency of radionuclide removal through the process varies widely, however, from the results in Table II, we concluded that ion exchange is an effective processing method to remove radiocesium. When salt is used, Na is the only cation that can replace Cs and K. However, for pickling with soy sauce, which contains ca. 60g kg^{-1} of Na and ca. 3.5 g kg^{-1} of K, there might be some effect on F_rs for K and Cs due to the difference in exchange between K and radioactive Cs.

Previously, we studied the applicability of K data using edible plant samples for food processing by comparing F_r values for ^{137}Cs and ^{40}K [3]. The values were found to be close to the 1:1 line and highly correlated (R=0.96, p<0.001). Thus, K can be used as an analogue to estimate radiocesium F_r by food processing of vegetables for the case of the indirect contamination pathway. For some vegetables which are peeled before boiling, the removal rate for radiocesium might be underestimated because the K and Cs distributions in a plant differ; the peel may contain more Cs than K. However, the radiocesium concentration in crop samples is steadily decreasing due to radioactive decay and it is difficult to obtain F_r; the values obtained using K would provide supportive data although K behavior is not a perfect analogue for Cs behavior in food processing.

F_r values for fruits and nuts

For fruits and nuts, peeling (apple, persimmon and loquat), pickling (persimmon) and boiling (chestnut and peanut) processes were carried out and the results are listed in Table III. Since the samples were contaminated through roots/plant above surface uptake, the contamination pathway was indirect, thus, complete removal of radiocesium by peeling is difficult; the average F_r values ranged from 0.3-0.74. According to data compiled by the Radioactive Waste Management Center [7], it was possible to remove 97% of the radiocesium when pear fruit was contaminated on its surface, but that result was not comparable to our present findings.

Table III. Food processing efficiency (P_e), and food processing retention factor (F_r) of radiocesium for fruits and nuts.

Sample	Process	N	P_e (Range)	F_r (Range)
Apple	Peeling	1	0.89	0.74
Persimmon		3	0.79 (0.76-0.81)	0.57 (0.51-0.67)
Loquat		2	0.48 (0.44-0.51)	0.3 (0.2-0.4)
Apple Juice	Concentrating (at 80˚C)	1	0.23	1.0
Persimmon	Drying (5˚C)	1	0.23	1.0
Persimmon	Pickling with 5% w/w salt, 30 min	1	0.8	0.9
Chestnut	Boiling with hard shell, 10 min	1	1.0	1.0
	Boiling after peeling hard shell, 5 min	1	1.0	0.69
Peanut	Boiling after peeling hard shell, 50 min	1	Not measured	0.39

When apple juice and persimmon (peeled) were processed to remove some of their water content by heating or cooling to a certain percent, no radiocesium removal was found. Radiocesium was just concentrated in these water-reduced materials. Since cesium boiling point is 678˚C, it is removing it by heating at a relatively low temperature is difficult.

Boiling of chestnut with the hard shell did not remove any radiocesium, however, when the hard shell was removed before boiling, some radiocesium removal was observed as was found for vegetables. The hard shell prevented radiocesium removal from inside. We also checked the concentration change in the hard shell before and after boiling and the F_r values was 0.95. Thus radiocesium in the hard shell was also difficult to remove by boiling.

CONCLUSIONS

Food processing retention factors were evaluated after the FDNPP accident and the results are summarized in this study. In most cases, radiocesium could be removed by food processing; however, removing water (by drying) from raw materials did not lead to any drop in radiocesium concentration. Also, when the edible part was protected by a hard shell (chestnut), boiling also could not remove Cs. Since the concentration level of radiocesium decreases naturally by radioactivity decay, data collection for food processing becomes difficult over time. According to some results [3, 7] it might be useful to apply F_r data on K as an indicator of radiocesium because their removal rates are close, although there is some differences due to their different distributions in crops. These collected data will provide information to lessen worries about contaminated foodstuffs among consumers.

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