Chapter 18
Radiocesium and Potassium Decreases in Wild Edible Plants by Food Processing

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Abstract It is more than 4 years since March 11, 2011, and, at this stage, foods that exceed the standard limits of radiocesium are mainly from the wild. Hence, one of the public's main concerns is how to decrease ingestion of radiocesium from foods they have collected from the wild as well as from their home-grown fruits because radioactivities in these food materials have not been monitored. In this study, we focused on wild edible plants and fruits, and the effects of washing, boiling, and pealing to remove radiocesium were observed. Samples were collected in 2013 and 2014 from Chiba and Fukushima Prefectures, e.g., young bamboo shoots, giant butterbur, and chestnuts. Wild edible plants were separated into three portions to make raw, washed, and boiled samples. For fruit samples (i.e., persimmon, loquat, and Japanese apricot), fruit parts were separated into skin, flesh, and seeds.

It was found that washing of plants is not effective in removing both ^{137}Cs and ^{40}K, and that boiling provided different removal effects on plant tissues. The retention factors of ^{137}Cs and ^{40}K for thinner plant body sample (leaves) tended to be higher than those for thicker plant body types, e.g., giant butterbur petiole and bamboo shoots. Thus, the boiling time as well as the crop thickness affects radiocesium retention in processed foods. For fruits, Cs concentration was higher in skin than in fruit flesh for persimmon and loquat; however, Japanese apricot showed different distribution.

Keywords Food processing retention factor • Radiocesium • Potassium • Wild edible plants • Fruits

18.1 Introduction

Radiocesium (^{134}Cs + ^{137}Cs) concentrations in foods are of great concern in Japan since the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, as people wish to avoid receiving additional internal doses from ingestion of the radionuclide.

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Food monitoring has been carried out since March 2011; provisional regulation values for total radiocesium concentration were applied at the time, and the values were 200 Bq kg^{-1} for water, milk, and daily products, and 500 Bq kg^{-1} for other food materials. From April 1, 2012, standard values for radiocesium have been employed in Japan, and the concentration in marketed raw food materials has been limited up to 100 Bq kg^{-1}, except baby foods (50 Bq kg^{-1}) and drinking water (10 Bq kg^{-1}). If any food exceeds these limits, the food name together with the producing district has been reported immediately by the Ministry of Health, Labour, and Welfare (MHLW). Every month, radioactivities of more than 20,000 samples are being measured and the recent data of February 2015 [1] have shown that foods exceeding the standard limits were mostly from the wild (i.e., not commercially grown or raised), i.e., meats of wild boar, Sika deer, Asian black bear, Japanese rock fish (marine), Japanese eel and char (freshwater). Not only meats but also edible wild plants in spring and autumn in 2014, e.g., giant butterbur flower-bud, Japanese angelica-tree shoot (*taranome*), and various mushrooms species, exceeded the limits.

One of the public's main concerns is how to decrease ingestion of radiocesium from foods, especially foods they have collected from the wild as well as from their home-grown garden fruits because radioactivities in these foods have not been measured for many cases. Unfortunately, radiocesium removal data by food processing, including culinary preparation, for crops commonly consumed in Japan have been limited because of little interest in the topic before the FDNPP accident. To remedy this, Japanese researchers have begun collecting such data [2–12]. Recently, the Radioactive Waste Management Funding and Research Center (RWMC) compiled the radiocesium removal ratios by food processing using open source data published mainly in 2011 and 2012 [13]. However, it is necessary to add more data to update the information.

In the present study, we focused on food processing effects on radiocesium removal in food plants from the wild to add more information and we compared obtained values with previously compiled values in the IAEA Technical Report Series No. 472 [14]. We also measured potassium-40 (^{40}K) concentrations in the same samples for comparison with radiocesium.

18.2 Materials and Methods

The following samples were collected in Chiba and Fukushima Prefectures in 2013 and 2014: giant butterbur (*Petasites japonicus*: flower-bud, 2; leaf blade, 4; petioles, 4), Japanese mugwort (*Artemisia indica var. Maximowiczii*: young shoots, 4), field-horsetail (*Equisetum arvense*: fertile stem, 3), water dropwort (*Oenanthe javanica*: young shoots, 1), Moso bamboo (*Phyllostachys heterocycla f. pubescens*: young shoots, 8), chestnut (*Castanea crenata*: nuts, 3), persimmon (*Diospyros kaki*: fruits, 2), loquat (*Eriobotrya japonica*: fruits, 4), and Japanese apricot (*Prunus mume*, 1). Sampling dates are listed in Tables 18.1, 18.2, and 18.3. Immediately after the

Table 18.1 Food processing retention factor (F_r) of ^{137}Cs and ^{40}K for giant butterbur tissues collected in 2013–2014

Tissue	Sampling date	Method of processing	Wet mass ratio	F_r of ^{137}Cs	F_r of ^{40}K
Flower bud	2013/3/13	Boiling	1.1	0.19 ± 0.11	0.56 ± 0.10
Flower bud	2013/3/18	Boiling	1.2	0.40 ± 0.18	0.65 ± 0.14
Leaf blade	2013/4/5	Washing	1.0	0.90 ± 0.10	1.24 ± 0.08
Leaf blade	2013/4/19	Washing	1.0	1.01 ± 0.08	1.10 ± 0.07
Leaf blade	2013/4/30	Washing	1.0	1.04 ± 0.08	0.99 ± 0.05
Leaf blade	2014/4/8	Washing	1.0	0.98 ± 0.10	0.99 ± 0.04
Leaf blade[a]	*2012–2014*	*Washing*	–	*0.95 (n = 8)*	*1.02 (n = 8)*
Leaf blade	2013/4/5	Boiling	1.1	0.43 ± 0.05	0.51 ± 0.04
Leaf blade	2013/4/19	Boiling	1.0	0.39 ± 0.03	0.60 ± 0.04
Leaf blade	2013/4/30	Boiling	1.0	0.46 ± 0.04	0.56 ± 0.03
Leaf blade	2014/4/8	Boiling	0.95	0.40 ± 0.05	0.44 ± 0.02
Leaf blade[a]	*2012–2014*	*Boiling*	–	*0.40 (n = 8)*	*0.46 (n = 8)*
Petiole	2013/4/5	Washing	1.0	0.69 ± 0.16	1.20 ± 0.07
Petiole	2013/4/19	Washing	1.0	1.34 ± 0.26	0.90 ± 0.05
Petiole	2013/4/30	Washing	1.0	1.10 ± 0.18	1.01 ± 0.05
Petiole	2014/4/8	Washing	1.0	0.91 ± 0.20	0.94 ± 0.04
Petiole[a]	*2012–2014*	*Washing*	–	*0.97 (n = 8)*	*1.01 (n = 8)*
Petiole	2013/4/5	Boiling	0.83	0.59 ± 0.12	0.82 ± 0.05
Petiole	2013/4/19	Boiling	0.84	0.93 ± 0.18	0.73 ± 0.04
Petiole	2013/4/30	Boiling	0.84	0.66 ± 0.10	0.66 ± 0.03
Petiole[b]	2014/4/8	Boiling	1.0	0.37 ± 0.10	0.35 ± 0.02
Petiole[a]	*2012–2014*	*Boiling*	–	*0.80 (n = 8)*	*0.80 (n = 8)*

± shows error from counting
[a]Averaged value for the data collected from 2012 (published in [2]) to 2014
[b]The sample was kept in water for 1 h after boiling

collection, samples were transferred to a laboratory and weighed to obtain the fresh weight.

In order to obtain the food processing effect, giant butterbur, Japanese mugwort, and water dropwort samples were separated into three portions to make raw, washed, and boiled (2.5 min) subsamples. One giant butterbur petiole sample was soaked in water for 1 h at room temperature after boiling. Field horsetail, young bamboo shoot, and chestnut samples were separated into two portions to make raw and boiled subsamples (boiling times depended on samples). All samples were weighed before processing. Washing was carried out with tap water in a washing bowl by changing the water five times, and then, finally, the samples were rinsed with reverse osmosis (RO) water. For the boiling process, edible parts of the plants were cooked in RO water after washing with tap water five times. For fruits, a whole fruit was washed with running tap water and rinsed with RO water. Then the water was removed with paper towels from the fruits, and each tissue part (skin, flesh, and seeds) was separated and weighed.

Table 18.2 Food processing retention factor (F_r) of ^{137}Cs and ^{40}K for eight bamboo shoots collected in 2013

Tissue	Sampling date	Method of processing	Wet mass ratio	F_r of ^{137}Cs	F_r of ^{40}K
New shoot-1	2013/4/9	Boiling	0.97	0.94 ± 0.04	0.80 ± 0.05
New shoot-2	2013/4/9	Boiling	0.95	0.66 ± 0.04	0.67 ± 0.06
New shoot-3	2013/4/9	Boiling	0.95	0.65 ± 0.02	0.55 ± 0.06
New shoot-4	2013/4/9	Boiling	–	0.68 ± 0.03	0.69 ± 0.05
New shoot-5	2013/4/9	Boiling	–	0.65 ± 0.03	0.69 ± 0.04
New shoot-6	2013/4/9	Boiling	0.99	0.67 ± 0.02	0.63 ± 0.06
New shoot-7	2013/4/9	Boiling	0.97	0.88 ± 0.04	0.75 ± 0.06
New shoot-8	2013/4/9	Boiling	0.97	0.79 ± 0.05	0.80 ± 0.07
New shoot[a]	*2012–2013*	*Boiling*		*0.71 (n = 12)*	*0.73 (n = 12)*

± shows error from counting
[a]Averaged value for the data collected from 2012 (published in [2]) to 2013

Table 18.3 Food processing retention factor (F_r) of ^{137}Cs and ^{40}K for mugwort (M), field horsetail (F), water dropwort (W), and chestnut (C) collected in 2013–2014

Tissue	Sampling date	Method of processing	Wet mass ratio	F_r of ^{137}Cs	F_r of ^{40}K
M, New shoots	2013/5/8	Washing	1.0	0.87 ± 0.37	1.14 ± 0.07
M, New shoots	2013/5/24	Washing	1.0	1.09 ± 0.02	0.86 ± 0.13
M, New shoots	2013/9/26	Washing	1.0	0.89 ± 0.03	0.97 ± 0.32
M, New shoots	2014/12/9	Washing	1.0	0.77 ± 0.36	0.97 ± 0.05
M, New shoots[a]	*2012–2014*	*Washing*	–	*0.98 (n = 9)*	*0.97 (n = 9)*
M, New shoots	2013/5/8	Boiling	1.0	0.36 ± 0.21	0.57 ± 0.04
M, New shoots	2013/5/24	Boiling	1.1	0.25 ± 0.01	0.45 ± 0.08
M, New shoots	2013/9/26	Boiling	1.0	0.47 ± 0.02	0.64 ± 0.19
M, New shoots	2014/12/9	Boiling	0.94	0.29 ± 0.20	0.26 ± 0.02
M, New shoots[a]	*2012–2014*	*Washing*	–	*0.39 (n = 8)*	*0.46 (n = 8)*
F, Fertile stem	2013/3/8	Boiling	0.81	0.62 ± 0.29	0.43 ± 0.0.5
F, Fertile stem	2013/3/20	Boiling	0.86	0.47 ± 0.25	0.57 ± 0.08
F, Fertile stem	2013/3/26	Boiling	0.75	0.16 ± 0.33	0.44 ± 0.04
F, Fertile stem[a]	*2012–2014*	*Boiling*	–	*0.46 (n = 6)*	*0.52 (n = 6)*
W, New shoots	2013/5/24	Washing	1.0	0.90 ± 0.06	0.96 ± 0.22
W, New shoots	2013/5/24	Boiling	0.83	0.61 ± 0.04	0.32 ± 0.12
C, nuts-1[b]	2013/9/26	Boiling	1.0	1.30 ± 0.01	1.04 ± 0.17
C, nuts-2[b]	2013/9/26	Boiling	1.0	0.68 ± 0.01	0.78 ± 0.15
C, nuts-3[b]	2013/9/26	Boiling	1.0	0.81 ± 0.01	0.77 ± 0.19

± shows error from counting
[a]Averaged value for the data collected from 2012 (published in [2]) to 2014
[b]Samples taken from different trees

Then, all samples were oven-dried at 80 °C to decrease the sample volume. Each oven-dried sample was pulverized and mixed well, and then transferred to a plastic container (U8 container). Radioactivity concentration in each sample was measured by a Ge detecting system (Seiko EG&G) and the gamma spectrum was analyzed using Gamma Station software (Seiko EG&G) to obtain activity on wet mass basis (Bq kg^{-1}-wet mass). The detection limit was about 0.5–1.0 Bq kg^{-1}-wet mass with the counting time of 40,000–80,000 s. The radiocesium concentrations in samples were usually low and sometimes ^{134}Cs could not be detected 2–3 years after the accident. Therefore, in this study, only ^{137}Cs data are presented together with ^{40}K.

Food processing retention factor (F_r) was determined by using the following equation as defined in IAEA TRS-472 [14]:

$$F_r = A_{after}\,(Bq)\ /\ A_{before}\,(Bq)$$

where A_{after} is the total activity of ^{137}Cs or ^{40}K retained in the food after processing (Bq), and A_{before} is the total activity in the food before processing (Bq). The wet mass of the processed subsample before processing (W_{bp}, kg) was recorded, thus, A_{before} was calculated using the following equation:

$$A_{before} = C_{raw} \times W_{bp}$$

where C_{raw} is the radioactivity concentration of ^{137}Cs or ^{40}K in the raw subsample (Bq kg^{-1}-wet mass).

For fruit samples, distribution percentages of wet mass, ^{137}Cs and ^{40}K in skin, flesh, and seeds were calculated and these distributions were compared.

18.3 Results and Discussion

The F_r values of ^{40}K and ^{137}Cs for each wild edible plant sample are shown in Tables 18.1, 18.2, and 18.3. Some F_r data exceeded 1.0 although that is impossible from the above equation; however, because of the low concentrations in samples giving a large counting error, and the samples for raw and processed samples being not completely the same, values of more than 1.0 were inevitable.

The mean F_rs of ^{137}Cs by washing for butterbur leaf blade ($n = 4$), petiole ($n = 4$), mugwort ($n = 4$), and water dropwort ($n = 1$) were 0.98, 1.0, 0.91, and 0.90, respectively and that of ^{40}K were 1.1, 1.0, 0.98, and 0.96, respectively. From this result, it was clear that washing plants is not effective to remove both ^{40}K and ^{137}Cs, because both elements are distributed inside the plant body. Boiling provided different removal effects by plant tissues. The mean F_rs of ^{137}Cs for leaf-blade of giant butterbur ($n = 4$), mugwort ($n = 4$), water dropwort ($n = 1$), and fertile stem of field horsetail ($n = 3$) were 0.40, 0.41, 0.61, and 0.42, respectively, i.e., about 40–60 % of the ^{137}Cs was removed by this process. Similar results were observed for ^{40}K for these samples. Unfortunately, however, the number of samples treated in 2013–2014 was not enough for statistical analysis between F_rs of ^{40}K and ^{137}Cs in each plant species. Only analytical results for bamboo shoots ($n = 8$) are reported here; when ANOVA test was carried out, no statistical difference was observed between F_rs of ^{40}K and ^{137}Cs. For some sample types, all the data from 2012 (reported in [2]) to 2014 were summarized (Tables 18.1, 18.2, and 18.3) and the values for ^{40}K and ^{137}Cs did not show any statistical differences by ANOVA test. Thus, we concluded that K could be used as an analogue for radiocesium to calculate F_rs.

Compared to leafy samples, bamboo shoots and petioles of giant butterbur (except soaking in water for 1 h) showed slightly higher F_rs of ^{137}Cs of 0.74 and 0.73, respectively; since these tissues are thicker than leaf tissues, it is reasonable to expect that more radiocesium would be retained in these tissues. However, when the giant butterbur sample soaked in water for 1 h showed lower F_r values, therefore, boiling followed by soaking would effectively remove ^{137}Cs, although K was also removed by this process. For chestnut, mean values of F_r of ^{137}Cs and ^{40}K were 0.95 and 0.87, respectively. Removal of these two elements by boiling of chestnut was difficult, because the hard shell prevented extracting ^{137}Cs and ^{40}K from the nuts into the boiling water. In total, most of the F_r values presented in this study and our previous data [2] were within the range of values compiled by IAEA [14], as shown in Fig. 18.1.

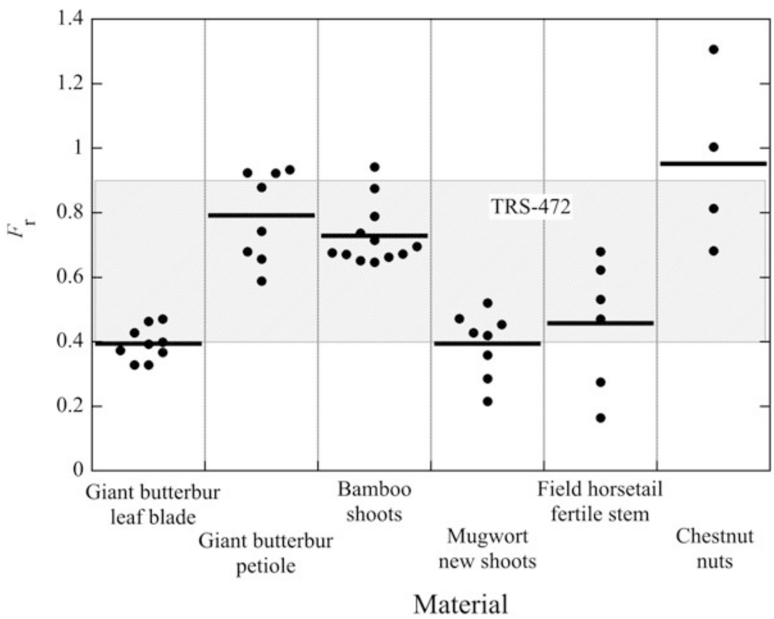

Fig. 18.1 Comparison of food processing retention factor (F_r) of ^{137}Cs for several plant materials collected in 2011–2014 by boiling. Both previous [2] and present study's data are plotted. The *shaded area* shows Fr range for vegetables, berries, and fruits boiled in water reported in TRS-472 by IAEA [14]

Table 18.4 Relative proportions (%) of wet mass, ^{137}Cs and ^{40}K in fruit tissues (skin, flesh, and seeds) to whole fruits for four species collected in 2013–2014 at harvest

Sample	Sampling date	Wet mass, %			^{137}Cs, %			^{40}K, %		
		Skin	Flesh	Seeds	Skin	Flesh	Seeds	Skin	Flesh	Seeds
Persimmon[a]	2011/10/4	13	83	4	23	67	10	–	–	–
Persimmon[a]	2011/10/20	9	91	–	19	81	–	–	–	–
Persimmon	2013/10/8	11	85	4	16	74	11	22	71	7
Persimmon	2014/10/9	11	85	4	16	81	2	19	76	5
Loquat	2011/6/7	22	47	31	34	44	21	–	–	–
Loquat-1[b]	2013/6/19	18	59	22	29	50	21	20	39	41
Loquat-2[b]	2013/6/19	12	69	18	19	69	12	16	50	34
Loquat	2014/6/4	16	62	22	22	56	22	22	43	35
Loquat	2014/6/11	11	68	22	15	72	13	13	48	39
Japanese apricot[c]	2012	12	75	13	12	83	6	–	–	–
Japanese apricot	2014/6/4	17	69	14	11	82	7	22	70	8

[a]Data from [15]
[b]Samples were taken from different trees on the same date
[c]Data from [13]

The results for fruits are listed in Table 18.4. For persimmon and loquat fruits, compared to the relative proportions of skin mass to the total fruits, partitioning percentages of ^{137}Cs and ^{40}K in the skin samples were 1.4–2.2 and 1.2–2.0 times higher, respectively, and those in flesh were 0.8–1.1 and 0.6–0.9, respectively. Thus, ^{137}Cs concentrations in skin were higher than those in flesh for both species. On the other hand, Japanese apricot showed higher percentage of ^{137}Cs distribution than wet mass proportion, although ^{40}K distributions were similar to those of wet mass proportion. In this study, we measured only one sample, however, similar results were obtained in 2012 [13], as listed in the same table.

In 2011, loquat and persimmon fruits were also measured and the ^{137}Cs distribution percentages in skin samples [15] were higher than those observed in 2013 and 2014 for both species. It was assumed that immediately after the ^{137}Cs was taken up through these trees' aboveground parts, it transferred to their growing tissues including the fruits; during development of fruits, fruit flesh mass increases at the middle-late ripening stage [16], but probably ^{137}Cs supply reduced due to the smaller amount of ^{137}Cs uptake through tree surface. By this process, ^{137}Cs distributions in fruit parts differed from those we observed in 2013 and 2014, although more studies are necessary to understand the mechanisms of Cs transfer in fruit trees.

18.4 Conclusions

The effects of washing and boiling of wild edible plants, and peeling of home-grown fruits were studied. It was found that washing plant surface is not effective in removing both ^{40}K and ^{137}Cs, because both elements are in the plant tissues. Food processing retention factor, F_r, decreased by boiling, and the average values ranged from 0.40 to 0.61 for leaf-blade of giant butterbur, mugwort, water dropwort, and fertile stem of field horsetail. Bamboo shoots and petioles of giant butterbur (except soaking in water for 1 h), however, showed slightly higher F_rs of ^{137}Cs of 0.59–0.94, because these tissues are thicker than leaf tissues, and thus, it is reasonable to expect that more radiocesium would be retained in these tissues. For fruits of persimmon and loquat, ^{137}Cs concentrations in skin were higher than those in flesh, but different trend was observed in Japanese apricot. Interestingly, the distributions of ^{137}Cs in skin, flesh, and seeds observed in this study differed from those observed in 2011. The mechanism is not clarified yet; therefore, further studies are necessary to understand the radiocesium transfer to fruits after direct deposition to fruit tree surfaces.

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