Chapter 16
Concentrations of $^{134, 137}$Cs and ^{90}Sr in Agricultural Products Collected in Fukushima Prefecture

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Abstract On April 1, 2012, new Standard Limits for radionuclide concentrations in food were promulgated, superseding the Provisional Regulation Values in Japan set in 2011. The new Standard Limits are calculated based on 1 mSv y^{-1} of annual internal radiation dose through food ingestion of ^{134}Cs, ^{137}Cs, ^{90}Sr, Pu and ^{106}Ru, which were detected or possibly released into the environment from the accident at the TEPCO Fukushima Daiichi Nuclear Power Stations (FDNPS). The concentrations of the radionuclides were based on the values of radiocesium ($^{134, 137}$Cs) and of the other radionuclides (^{90}Sr, Pu and ^{106}Ru); the ratio observed in the determination or predicted concentrations in the soils from the FDNPS accident was used for estimating the concentration of the other radionuclides by means of the ratio against ^{137}Cs. The new Standard Limit of radiocesium in general foods was defined to be 100 Bq kg^{-1} fresh weight by the Ministry of Health, Labour and Welfare. In the present study the concentration of radiocesium was measured in agricultural products collected mostly in Fukushima-shi and Date-shi, Fukushima Prefecture, in 2012 and 2013. The average concentration of radiocesium in agricultural plants in 2012 was 7.6 (<0.2–40) Bq kg^{-1} fresh weight, decreasing to 2.0 (<0.1–14) Bq kg^{-1} fresh weight in 2013, which was approximately one-fourth of the concentration in 2012. The concentration of ^{90}Sr in agricultural products

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collected in Fukushima Prefecture in 2013 was 0.0047–0.31 Bq kg^{-1} fresh weight, which was a similar range to those collected throughout Japan. The concentration ratio of $^{90}Sr/^{137}Cs$ in the agricultural plants collected from the area 5 km west from the Nuclear Power Stations (difficult-to-return zone) was lower than the predicted $^{90}Sr/^{137}Cs$ ratio, which was calculated using the ratio in the soils and soil-to-plant transfer factors.

Keywords Agricultural product • New standard limits • $^{134, 137}Cs$ • ^{90}Sr • $^{90}Sr/^{137}Cs$ ratio

16.1 Introduction

Significant quantities of radionuclides were released into the environment from the TEPCO's Fukushima Daiichi Nuclear Power Stations (FDNPS) accident in March 2011. Radiocesium ($^{134, 137}Cs$) is the major radionuclide released by the accident and an important radionuclide for the assessment of radiation exposure to the public. Other relatively long half-life radionuclides such as ^{90}Sr, Pu, etc. are also important radionuclides for radiation dose estimation through long-term food ingestion. The new Standard Limits for radionuclides in foods was established by the Ministry of Health, Labour and Welfare on April 1, 2012. The limits were determined on the basis of 1 mSv y^{-1}. The limit in general foods is 100 Bq kg^{-1} of radiocesium, including the contribution of ^{90}Sr, Pu and ^{106}Ru, determined by using the actual concentration in the soils from the accident (estimated data for ^{106}Ru) and soil-to-transfer factor. However, the public has been concerned about food contamination, especially ^{90}Sr. In the present study the concentrations of radiocesium and ^{90}Sr in agricultural and animal products produced in Fukushima Prefecture were determined, and compared with the values of the new allowable Standard Limits.

16.2 Materials and Methods

Agricultural and animal products, limited to those produced within the Fukushima Prefecture, were collected from markets located mostly in Fukushima-shi and Date-shi. Table 16.1 contains a list of 120–5000 g samples of 11 spices in 2012 (40 samples) and 2013 (42 samples). The agricultural plants were washed, peeled, and then the edible parts were cut into small pieces. Each sample was dried at 70 °C for 1 week and pulverized in a stainless steel cutter blender before being analyzed for radiocesium. The animal samples were also cut into small pieces. The samples were compressed into a plastic container (47 mm in diameter and 50 mm in height) and the concentration of radiocesium and ^{40}K determined with a Ge detector connected to a multichannel analyzer system by counting for 9400–33,000 s. The

Table 16.1 Collected agricultural and animal products in Fukushima Prefecture in 2012 and 2013

Agricultural and animal products	Collected sample
Rice	Brown rice
Potatoes	Potato, Sweet potato, Eddoe
Savory herbs	Japanese ginger
Leaf and stem vegetables	Komatsuna, Malabar spinach, String bean, Cabbage, Welsh onion, Leek, Spinach, Japanese honeywort, Turnip (leaf and stem), Turnip rape, Mugwort, Asparagus, Onion
Root vegetables	Turnip, Burdock, Radish, Carrot
Pulses	Green soybean, Green bean, Cowpea, Black soybean
Fruity vegetables	Cucumber, Tomato, Green pepper, Eggplant, Pumpkin, Small green pepper, Okra, Broccoli, Snap garden pea, Zucchini, Wax gourd
Fruits	Pear, Apple, Persimmon, Huckleberry, Plum, Peach, Blue berry, Japanese plum
Wild vegetables	Udo, Butterbur, Momijigasa
Other agricultural plants	Jew's-ear mushroom, Shiitake mushroom, Hen-of-the-woods mushroom, Edible chrysanthemum, Japanese pepper (leaf)
Animal products	Chicken, pork, egg

detection efficiency of the Ge detector was dependent on the sample thickness and was obtained using the mixed standard radionuclides material made by the Japan Radioisotope Association. Counting statistics standard deviations for ^{137}Cs concentration in the sample were less than 10 % of the value.

Soil and agricultural samples were collected from 10 agricultural fields located both outside and within the more than 50 mSv y^{-1} of the external radiation dose zone (difficult-to-return zone, Okuma) in Fukushima Prefecture in 2013 and 2014 (Fig. 16.1) . Shiitake mushroom and its mushroom bed for cultivation was also collected. A stainless steel core sampler was used to collect soil cores 5 cm in diameter and 20 cm in depth at 5 points evenly distributed in each field. Twenty kilogram of each agricultural sample was collected from each field. The soil core samples collected from each field were dried at 50 °C for 1 week and then passed through a 2 mm sieve. The soil samples in each field were thoroughly mixed. The agricultural samples were washed, peeled, and then the edible parts were cut into small pieces. Approximately 100 g of dried sample was pulverized in a stainless steel cutter blender before being analyzed for radiocesium. The rest of the dried agricultural samples were washed at a temperature below 450 °C for analysis of ^{90}Sr. The dried soil and agricultural samples were compressed into plastic containers and the concentrations of radiocesium and ^{40}K determined. A radioanalytical method for ^{90}Sr was performed according to the previously reported method [1]. The ash plant samples (20–50 g) were decomposed with HNO_3, H_2O_2 and HCl after the addition of the Sr carrier. The soil samples (100 g) were heated at 450 °C and then extracted with 12 M HCl after the addition of the Sr carrier. The solution was filtered and the residue discarded. The solution was adjusted to >pH 10 with

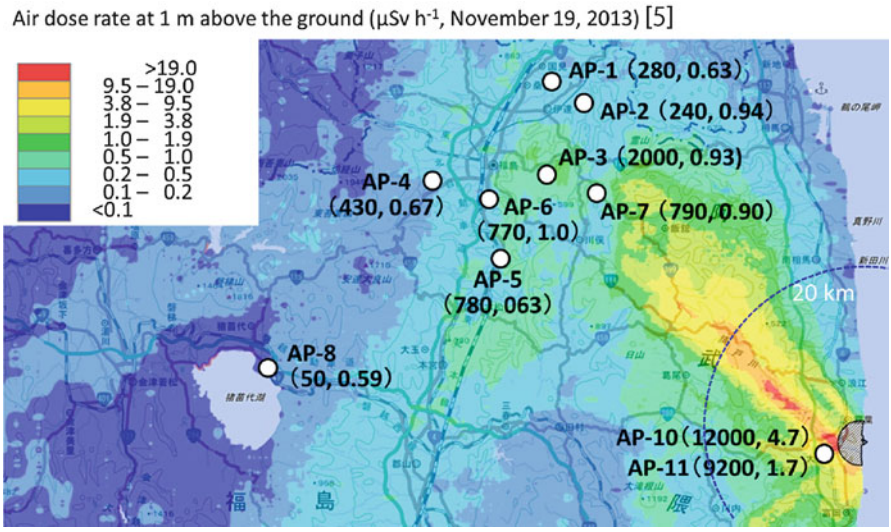

Fig. 16.1 Concentrations of ^{137}Cs and ^{90}Sr (Bq kg^{-1} dry weight) in cultivated soil ($n = 10$) collected from Fukushima Prefecture in 2014. Point numbers show the sampling agricultural plants indicated in Table 16.3 and the values in parentheses are the concentrations of ^{137}Cs (former) and ^{90}Sr (latter) in cultivated soil

NaOH and then $SrCO_3$ precipitated by adding Na_2CO_3. The $SrCO_3$ precipitate was dissolved in HCl and then the oxalates re-precipitated at pH 4.2 by adding oxalic acid. The supernatant was decanted and the oxalate precipitation dissolved in HNO_3. Strontium in the solution was separated from Ca by the cation ion-exchange method, and then the filtrated precipitate was dissolved with water. Any radioactive impurity was eliminated by scavenging on $BaCrO_4$ and $Fe(OH)_3$. An ammonium carbonate solution was added to the solution after scavenging, and the $SrCO_3$ precipitate was filtered using a cellulose filter paper. The recovery of Sr was estimated by measuring the stable Sr with ICP-AES and then a disk sample prepared for beta-counting for 6000–60,000 s.

16.3 Results and Discussion

The concentrations of radiocesium in agricultural products collected in Fukushima Prefecture in 2012 and 2013 are listed in Table 16.2. The average concentration of radiocesium collected in 2013 was 2.0 ± 2.7 (<0.046–14) Bq kg^{-1} fresh weight, which was one-fourth of that in 2012 (7.6 ± 10, <0.11–40 Bq kg^{-1} fresh weight), and the concentrations of radiocesium in all the samples were less than the new Standard Limits. The concentrations of radiocesium in agricultural products decrease with time elapsed, and the reasons are as follows:

Table 16.2 Concentration ranges of ^{134}Cs, ^{137}Cs and ^{40}K in agricultural and animal products (grouping noted in Table 16.1) collected from Fukushima Prefecture in 2012 and 2013

Agricultural and animal products	2012				2013			
	^{134}Cs	^{137}Cs	^{40}K	n	^{134}Cs	^{137}Cs	^{40}K	n
	Concentration (Bq kg^{-1} fresh weight)				Concentration (Bq kg^{-1} fresh weight)			
Rice	1.0–2.5	1.4–4.9	34–55	3	<0.58	0.66	82	1
Potatoes	0.25–2.8	0.53–4.4	120–190	3	0.42–1.6	0.86–3.5	110–160	3
Savory herbs				–	1.2	2.4	130	1
Leaf and stem vegetables	0.08–2.0	0.17–3.8	39–140	8	<0.11–2.8	<0.11–4.7	42–280	15
Root vegetables				–	<0.022–0.41	<0.024–0.83	78–140	5
Pulses	5.9–15	10–25	160–560	3	1.1	2.2	210	1
Fruity vegetables	<0.063–2.3	<0.052–3.6	48–180	8	<0.046–0.85	<0.10–2.1	53–180	9
Fruits	0.16–13	0.25–23	26–170	7	0.77–0.92	1.6–2.1	45–47	2
Wild vegetables					0.26–4.4	0.51–9.6	94–160	4
Other agricultural plants	1.4–5.4	2.4–8.8	18–100	4	1.1	2.1	110	1
Animal products	<0.85	<0.68	60–110	4				–

1. Decay of radiocesium activities, especially ^{134}Cs (half-life, 2.1 y)
2. Countermeasure with the application of K fertilizer for reducing uptake of radiocesium in plants
3. Aging effect in soil, which radiocesium in exchangeable fraction decreases and that in strongly bound fraction increases with time elapsed [2, 3]
4. Decreasing radiocesium contents in soil by erosion, etc.

The concentration of radiocesium in agricultural plants drastically decreases immediately after the accident, and the rate of the decrease of radiocesium concentration in plants has gradually slowed as more time has lapsed. The reported mean concentration of radiocesium in a duplicate diet collected from Fukushima Prefecture in December 2011 was 1.5 Bq kg^{-1} [4], which was lower than that in 2013 as determined in this study. This is because the concentration of radiocesium in foods determined by a duplicate method decreased during processing and cooking and market dilution effect.

The concentrations of $^{134, 137}$Cs and ^{90}Sr were determined in surface soils and agricultural plants collected from both outside and within the difficult-to-return zone in Fukushima Prefecture. The concentration of radiocesium in the agricultural soils outside the zone is decreasing because of plowing, migration, erosion, etc. However, the soil within the difficult-to-return zone (experimental field 5 km west from the

Table 16.3 Concentrations of ^{134}Cs, ^{137}Cs, ^{40}K and ^{90}Sr in agricultural plants collected from Fukushima Prefecture in 2012 and 2013

Sample no.	Agricultural plant	Dry matter content	Concentration			
			^{134}Cs	^{137}Cs	^{40}K	^{90}Sr
		(%)	(Bq kg^{-1} fresh weight)			
AP-1	Komatsuna	4.5	0.030 ± 0.0036	0.055 ± 0.0044	100 ± 0.34	0.054 ± 0.0027
AP-2	Cucumber	4.2	0.063 ± 0.0074	0.11 ± 0.008	66 ± 0.57	0.013 ± 0.0011
AP-3	Brown rice	89	0.74 ± 0.054	1.6 ± 0.077	65 ± 1.9	0.013 ± 0.0018
AP-4	Potato	19	1.7 ± 0.026	3.9 ± 0.039	130 ± 0.88	0.012 ± 0.00093
AP-5	Carrot	9.1	0.36 ± 0.032	0.78 ± 0.040	130 ± 1.7	0.031 ± 0.0022
AP-6	Soy bean	90	3.7 ± 0.32	8.8 ± 0.47	540 ± 14	0.30 ± 0.014
AP-7	Persimmon	14	1.5 ± 0.047	3.6 ± 0.074	56 ± 1.2	0.0086 ± 0.00050
AP-8	Edible chrysanthemum (flowers)	8.8	0.072 ± 0.0040	0.17 ± 0.0059	86 ± 0.32	0.044 ± 0.0039
AP-9	Shiitake mushroom	11	2.2 ± 0.093	5.1 ± 0.14	85 ± 2.3	0.0047 ± 0.00032
AP-10[a]	Pumpkin	15	27 ± 0.79	80 ± 1.3	75 ± 5.9	0.31 ± 0.0061
AP-11[a]	Cabbage	5.8	17 ± 0.38	50 ± 0.68	64 ± 3.5	0.21 ± 0.0057
	Various agricultural plants[b]		ND[c] ~ 4.9	ND ~ 10		ND ~ 0.91

The errors indicate one standard deviation of counting statistics
[a]Collected from the difficult-to-return zone (Okuma)
[b]Samples collected throughout Japan excluding Fukushima Prefecture in 2013 (data from Environmental Radioactivity and Radiation in Japan [6])
[c]Not detected

Nuclear Power Stations in Okuma) is still highly contaminated with radiocesium (Fig. 16.1). The concentration of ^{90}Sr in the soils collected outside the zone is low with no differences among the values. In contrast, ^{90}Sr concentration in the soils collected within the zone was several times higher than that outside the zone. However, the concentration of ^{90}Sr in the soils collected both outside and within the zone is within the range collected throughout Japan except Fukushima (ND-5.9 Bq kg^{-1}, data from Environmental Radioactivity and Radiation in Japan [6]). The concentration of radiocesium in the plants collected outside of the difficult-to-return zone (Table 16.3) is similar to the range shown in Table 16.2. The concentration of radiocesium in the agricultural plants within the zone was still higher than that outside the zone (Table 16.3); however, part of the plants cultivated

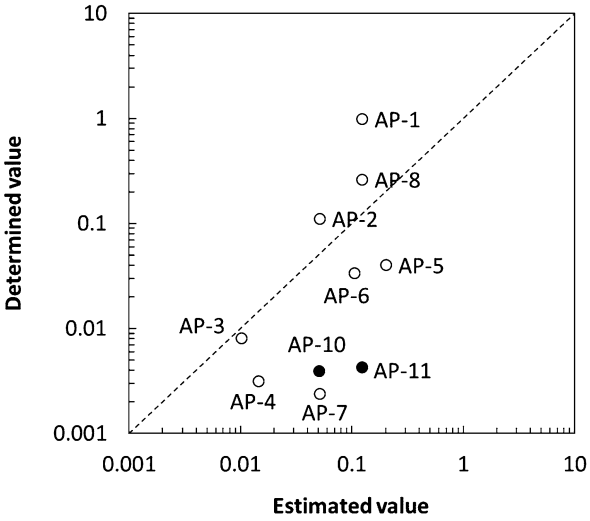

Fig. 16.2 Comparison of ^{90}Sr/^{137}Cs concentration ratio between the estimated value and the determined value in agricultural plants. *Plotted numbers* indicate sample no. in Table 16.2. The *solid-black circles* (AP-10 and AP-11) were collected within the difficult-to-return zone

in the experimental field was lower than the new Standard Limits [7]. The range of ^{90}Sr concentration in the plants collected outside the zone is 0.0047–0.30 Bq kg^{-1} fresh weight, and those collected within the zone were 0.31 (pumpkin) and 0.21 (cabbage). These data are also within the range collected throughout Japan except Fukushima (ND-0.91 Bq kg^{-1} fresh weight, data from Environmental Radioactivity and Radiation in Japan [6]).

The new Standard Limits of radiocesium include the contribution of ^{90}Sr concentration in general foods. The concentration ratio of ^{90}Sr/^{137}Cs in the foods was predicted by using observed ^{90}Sr/^{137}Cs concentration ratio (0.003) [8] in the soils from the FDNPS accident and the reported soil-to-plant transfer factor [9, 10], and the concentration of ^{90}Sr in foods was determined by multiplying the predicted ^{90}Sr/^{137}Cs ratio in foods by the measured ^{137}Cs value in foods. Therefore, the propriety between the predicted and the measured ^{90}Sr/^{137}Cs ratio in foods (Fig. 16.2) needs to be evaluated. The measured concentration ratio of ^{90}Sr/^{137}Cs in the plants, except for three samples (AP-1, komatsuna; AP-2, cucumber; AP-8, edible chrysanthemum), was lower than the predicted ^{90}Sr/^{137}Cs ratio, and the determined ratio in the two samples collected within the difficult-to-return zone (Okuma), which may have a large contribution from the accident, was also lower than the predicted ^{90}Sr/^{137}Cs ratio. The concentration of ^{90}Sr in the soils collected from the three fields, where the ^{90}Sr/^{137}Cs concentration ratio in the plants overestimated the predicted ratio, was similar in range to the global fallout deposited in the soil. Therefore it is necessary to attribute the ^{90}Sr contents in the plants as being derived from the global fallout from several decades ago.

Internal radiation doses from radiocesium through food ingestion for adult males and females (over the age of 19) were estimated. Measured and predicted data for the radiocesium concentration in food categories were used. The concentration of ^{137}Cs in the animal products including milk collected in 2012 and 2013 was not

detected, and the average value of the detection limits (0.6 Bq kg^{-1}) in the animal products was used for the dose estimation. Drinking water pathway was not included for the dose estimation because it was lower than the detection limit. The estimated internal radiation doses through food ingestion for males and females were 0.066 and 0.052 mSv y^{-1} in 2012, and those in 2013 were 0.016 and 0.012, respectively, reflecting the decreases in the concentration of radiocesium in foods with time elapsed. It was also reported that the internal radiation dose from radiocesium in Fukushima Prefecture in 2012 was 0.0039–0.0066 mSv y^{-1} by the market basket method [11], which was one order of magnitude lower than that in this study. This is attributed to the fact that the collected foods by the market basket method usually included products both within and outside of Fukushima Prefecture, and the concentration of radiocesium in the foods decreased by market dilution. On the other hand, the samples collected in this study were produced only in Fukushima Prefecture and were not influenced by the market dilution effect. The internal radiation doses from radiocesium by the duplicate diet method in Fukushima Prefecture were reported as 0.026 mSv y^{-1} in 2011 [4] and 0.0022 mSv y^{-1} in 2012 [11]. The internal radiation dose from radiocesium through food ingestion determined by the duplicate diet method is lower than that by the market basket method because of processing and cooking, and it is assumed that those values decrease with time elapsed.

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