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Surveillance of Strontium-90 in Foods after the Fukushima Daiichi Nuclear Power Plant Accident

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As a result of the Fukushima Daiichi nuclear power plant (NPP) accident, various radionuclides were released into the environment. In this study, we surveyed strontium-90 (^{90}Sr) concentrations in several foodstuffs. Strontium-90 is thought to be the third most important residual radionuclide in food collected after the Fukushima Daiichi, NPP accident after following cesium-137 (^{137}Cs) and cesium-134 (^{134}Cs). Results of ^{90}Sr analyses indicated that ^{90}Sr was detect in 25 of the 40 radioactive cesium (r-Cs) positive samples collected in areas around the Fukushima Daiichi NPP, ranging in distance from 50 to 250 km. R-Cs positive samples were defined as containing both ^{134}Cs and ^{137}Cs which are considered to be indicators of the after-effects of the Fukushima Daiichi NPP accident. We also detected ^{90}Sr in 8 of 13 r-Cs negative samples, in which ^{134}Cs was not detected. Strontium-90 concentrations in the r-Cs positive samples did not significantly exceed the ^{90}Sr concentrations in r-Cs negative samples or the ^{90}Sr concentration ranges in comparable food groups found in previous surveys before the Fukushima Daiichi NPP accident. Thus, ^{90}Sr concentrations in r-Cs positive samples were indistinguishable from the background ^{90}Sr concentrations arising from global fallout prior to the Fukushima accident, suggesting that no marked increase of ^{90}Sr concentrations has occurred in r-Cs positive samples as a result of the Fukushima Daiichi NPP accident.

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Key words: the Fukushima Daiichi nuclear power plant (NPP) accident; strontium-90 (^{90}Sr); radioactive cesium (Cs); surveillance

Introduction

According to the report by the Nuclear and Industrial Safety Agency (NISA) on 6 June, 2011, which was revised on 20 October, 2011*[1], many types of radionuclides were released during the Fukushima Daiichi nuclear power plant (NPP) accident, including xenon-133 (^{133}Xe; about 1.1×10^{19} Bq), iodine-131 (^{131}I; about 1.6×10^{17} Bq), ^{134}Cs (about 1.8×10^{16} Bq), ^{137}Cs (about 1.5×10^{16} Bq), strontium-89 (^{89}Sr; about 2.0×10^{15} Bq), and ^{90}Sr (about 1.4×10^{14} Bq). Though the released total quantities of ^{133}Xe, ^{131}I and tellurium-132 (^{132}Te) were substantial immediately following the accident, the half-lives of these radionuclides are very short, less than 10 days[1] (Table 1). Therefore, one year after the accident, residual radioactivity in the environment was predominantly due to ^{137}Cs. Cesium-134 and ^{90}Sr are the second and third most significant residual local radionuclides. Many foodstuffs were contaminated with radionuclides directly or indirectly, and on 17 March, 2011, immediately following the Fukushima Daiichi NPP accident, the Ministry of Health, Labour and Welfare began a program to control food destined for human consumption based upon provisional radioactive material regulations. These regulations established concentration values for radioactive iodine, radioactive cesium, uranium, plutonium and α-particle-emitting nuclides of transuranium elements. For example, previous regulatory levels for r-Cs (sum of ^{134}Cs and ^{137}Cs) in common foods such as seafood, vegetables and others had been limited to 500 Bq/kg. On 1 April, 2012, the new regulatory standards for radioactive materials in foods were enforced and continue to this day. These standards, for example, limit concentrations of r-Cs to 100 Bq/kg for foods except for drinking water, milk and infant foods. Although the new standard limits are represented as r-Cs concentrations, they include all radionuclides with half-lives ($T_{1/2}$) of more than one year, as summarized in Table 1[1]. These new regulatory concentration limits for r-Cs were established such that the effective dose to the populace from the target radionuclides would not exceed 1 mSv/year through the consumption of foods*[2]. In order to estimate the con-

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*[1] Ministry of Economy, Trade and Industry. Report of evaluation of reactor core state in Unit 1, 2 and 3 concerning the Tokyo Electric Power Company Fukushima Daiichi Nuclear Power Plant accident.
(6 June, 2011, revised version; 20 October, 2011). http://www.meti.go.jp/press/2011/06/20110606008/20110606008-2.pdf (revised version)

Table 1. Half-lives[1)] of major radionuclides released by the Fukushima Daiichi NPP accident

Radionuclides	Half-life
^{133}Xe	5.25 d
^{131}I	8.02 d
^{132}Te	3.20 d
^{134}Cs	2.06 yr
^{137}Cs	30.2 yr
^{90}Sr	28.8 yr
^{238}Pu	87.7 yr
^{239}Pu	24110 yr
^{240}Pu	6564 yr
^{241}Pu	14.35 yr
^{106}Ru	373.59 d

[1)] Japan Radioisotope Association. Radioisotope Pocket Data Book 11th Edition. Tokyo, Maruzen, 2011, p. 22–100. (ISBN 978-4-89073-211-1)

tribution from the target radionuclides other than r-Cs in the annual committed effective dose, radioactive concentration ratios of ^{137}Cs to the other radionuclides in various food groups were estimated based on the environmental monitoring data for the radionuclides, the estimates of radionuclide amounts released from the Fukushima Daiichi NPP reported by NISA, and the radionuclide transfer factors from environments to foods, etc.[2)]

Intensive monitoring of r-Cs in foods has been conducted since the Fukushima Daiichi NPP accident*[3]. Because r-Cs is a γ-ray-emitting radionuclide and is easy to measure, extensive information about r-Cs concentrations in foods is currently available*[4]. Besides r-Cs, ^{90}Sr has been recognized as one of the most prevalent target radionuclides in the Fukushima Daiichi NPP accident, due to its dose coefficient (higher than ^{137}Cs) and relatively long physical half-life. Once absorbed into the body, ^{90}Sr accumulates in bone and may cause bone cancer[3), 4)]. These findings stirred up public anxieties with regard to ^{90}Sr concentrations in foods. Unfortunately, it is time-consuming and costly to measure ^{90}Sr concentration in foods, since ^{90}Sr emits only β-rays. Also, because the amount of ^{90}Sr released was much smaller than that of r-Cs, it is difficult to apply other quick analytical techniques, such as ICP-MS. For these reasons, reports regarding ^{90}Sr concentration in foods after the Fukushima Daiichi NPP accident are fewer in number than reports regarding r-Cs. In the accident, multiple explosions of different types occurred in a number of units, which were in varying operational states. Because of the complexity of this accident, there is a poor correlation between the radioactive concentrations of highly volatile radionuclides such as ^{137}Cs and those of low-volatile radionuclides such as ^{90}Sr[5)]. Thus, it is critical in terms of food safety to identify current concentrations of ^{90}Sr as well as r-Cs in various foodstuffs. In this study, we measured concentrations of ^{90}Sr in various foods and we discuss the influence of the accident upon ^{90}Sr levels in foods. We also discuss the adequacy of the new regulatory limits by comparing the ratios of ^{137}Cs to ^{90}Sr in several food groups observed in this study to the estimated ratios used for establishment of the new limits.

*[2] Ministry of Health, Labour and Welfare, Summary of standard values of radioactive materials in foods. http://www.mhlw.go.jp/shinsai_jouhou/dl/20130417-1.pdf

*[3] Ministry of Health, Labour and Welfare, Regarding inspection plan of radioactive contaminants in foods by respective prefectures. (20 March, 2014) http://www.mhlw.go.jp/file/04-Houdouhappyou-11135000-Shokuhinanzenbu-Kanshianzenka/0000043037.pdf.

*[4] Ministry of Health, Labour and Welfare, Levels of radioactive contaminants in foods tested in respective prefectures. http://www.mhlw.go.jp/stf/kinkyu/0000045250.html

Materials and Methods

1. Samples

Food samples were collected between May 2011 and November 2013 from all over Japan. The concentrations of r-Cs (^{134}Cs and ^{137}Cs) in food samples were measured by utilizing a high-purity germanium (HPGe) γ-spectrometer (Canberra, Meriden, CT, USA). Food samples containing both ^{134}Cs and ^{137}Cs were considered to have been contaminated by the Fukushima Daiichi NPP accident and were defined as r-Cs positive samples. Food samples containing only ^{137}Cs or no detectable r-Cs were defined as r-Cs negative samples. Most of the samples classified as r-Cs positive were collected at distances of 50 to 250 km from the accident site. Most of the r-Cs positive samples were non-commercial samples, while most of the r-Cs negative samples were commercial samples. Food samples used for ^{90}Sr analysis in this study are listed in Tables 2 (r-Cs positive samples) and 3 (r-Cs negative samples).

2. Reagents and Materials

Reagents (special grade) were purchased from Wako Pure Chemical Industries, Ltd. and Kanto Chemical Co., Inc. Cation-exchange resin (Dowex 50W-X8) was purchased from the Dow Chemical Company (Midland, MI, USA). Prior to use, Dowex 50W-X8 resin was washed with distilled water and activated with HCl (1+1) and 24% (w/v) NaOH solution. A chromatography column 3 cm in diameter was filled with activated (H form) resin up to about 26 cm in height.

Filter papers used in quantitative analysis (No. 5A and No. 5C) were purchased from ADVANTEC MFS, Inc. (Dublin, CA, USA). Kiriyama filter paper and a Kiriyama funnel were purchased from Kiriyama Glass Works Co. A glass column (diameter; 3 cm, length; 30 cm), a discrete type filter and a long leg funnel were purchased from AGC Techno Glass Co., Ltd. Sample dishes (standard, diameter; 25 mm) were purchased from Chiyoda Technol Corporation and centrifuge bottles were purchased from Thermo Fisher Scientific Inc. (Waltham, MA, USA).

Table 2. Results of ^{90}Sr analysis (r-Cs positive samples)

Sample[1), 2)]	Food category	Weight of sample used for Sr analysis		Total Cs	^{137}Cs	^{90}Sr			Sr recovery rate
		Ash weight	Fresh weight (Corresponding value)	Radioactive conc.[3)]	Radioactive conc.[3)]	Radioactive conc.[3),4)]	Counting error	LOD[5)]	
		(g)	(g)	(Bq/kg)	(Bq/kg)	(Bq/kg)	(Bq/kg)	(Bq/kg)	
Codfish A	Sea fish and shellfish	3.72	300	34	24	*	*	0.07	0.923
Codfish B		8.44	500	99	63	*	*	0.04	0.979
Fat greenling		13.65	500	17	10	*	*	0.04	0.912
Flatfish*1		19.35	500	24	17	(0.46)[6)]	(0.05)[6)]	(0.09)[6)]	0.403
Marbled flounder*1		22.22	500	30	19	0.18	0.05	0.1	0.788
Pond smelt A*1	Freshwater fish and shellfish	17.28	800	28	19	0.073	0.02	0.03	0.731
Pond smelt B*1		17.12	800	34	22	0.11	0.02	0.03	0.839
Pond smelt C*1		11.25	500	28	17	0.15	0.03	0.04	0.892
Loach*1		27.14	1000	6.4	4.0	0.10	0.01	0.02	1.011
Rice bran A	Rice	10.79	100	720	390	*	*	0.2	0.841
Rice bran B		9.79	100	1000	560	0.27	0.08	0.2	0.874
Brown rice A		12.8	1000	98	54	0.036	0.009	0.02	0.800
Brown rice B		12.33	1000	160	90	0.040	0.009	0.02	0.805
Brown rice C		12.95	1000	77	44	*	*	0.02	0.840
Brown rice D		6.44	1000	89	54	0.071	0.02	0.04	0.914
Crude wheat A	Grains	17.50	1000	130	68	0.16	0.01	0.02	0.903
Crude wheat B		17.55	1000	150	81	0.057	0.01	0.02	0.799
Wheat bran		19.51	400	1100	580	0.57	0.04	0.06	0.848
Soy bean	Beans	26.51	500	250	140	0.28	0.02	0.04	0.947
Beef A	Beef	8.41	1000	550	300	*	*	0.02	0.976
Beef B		2.72	500	3500	1900	0.081	0.02	0.04	0.880
Beef C		6.91	1000	770	420	0.028	0.007	0.02	0.929
Beef D		8.12	1000	530	350	*	*	0.02	0.948
Beef E		8.34	1000	470	300	*	*	0.02	0.904
Shiitake mushroom A	Others	5.77	1000	39	25	0.029	0.008	0.02	0.896
Shiitake mushroom B		1.50	250	100	64	0.12	0.03	0.1	0.841
Shiitake mushroom C		4.30	800	43	30	*	*	0.03	0.892
Shiitake mushroom D		3.00	450	34	25	*	*	0.05	0.897
Shiitake mushroom E		4.41	500	110	71	*	*	0.04	0.854
Shiitake mushroom F		1.09	170	48	34	*	*	0.1	0.895
Dried shiitake mushroom A*2		5.43	120	2000	1300	0.42	0.08	0.2	0.837
Dried shiitake mushroom B*2		13.70	300	240	170	0.80	0.05	0.07	0.862
Nameko mushroom		0.43	95	590	410	*	*	0.2	0.924
Kuritake mushroom		0.6	140	970	690	0.52	0.07	0.2	0.864
Tawny milkcap mushroom		0.2	22	31000	19000	*	*	0.9	0.898
Mixture of some kinds of mushrooms		0.81	250	120	87	*	*	0.1	0.916
Green tea*2		10.38	200	3500	1800	0.60	0.06	0.1	0.986
Mulberry leaf tea*2		24.74	200	310	180	3.7	0.2	0.1	0.737
Horsetail		3.20	160	800	500	0.87	0.08	0.1	0.881
Bracken		8.60	1000	430	300	0.30	0.02	0.02	0.923

1) *1 indicates that samples were analyzed whole (including bones).
2) *2 indicates dried samples. Weight of these samples was not fresh weight but dried weight.
3) Radioactivity of cesium (^{134}Cs and ^{137}Cs) and ^{90}Sr was corrected for attenuation (reference date was the date of sample collection).
4) In the case where the counting rate was more than 3-times higher than the counting rate error, concentration of ^{90}Sr is shown to two significant figures. In the case where the counting rate was less than 3-times higher than the counting rate error, concentration of ^{90}Sr is shown as *.
5) Limit of detection (LOD) of ^{90}Sr is shown to one significant figure.
6) ^{90}Sr concentration was regarded as a reference value because Sr recovery rate was significantly low.

3. Measurement of r-Cs

R-Cs concentrations in food samples were measured with an HPGe γ-spectrometer. The fresh food samples were chopped and homogenized then poured into the measurement containers. According to the sample weight, density and concentration of r-Cs, the measurement time was adjusted between 10 min and 12 hr. To calculate the r-Cs concentration from the counting data acquired by the HPGe γ-spectrometer, analytical software (Gamma Explorer; Canberra) was used. The matrix was set to "water" or "ash (dry food)" and self-absorption was corrected by an algorithm within the analytical software. Results were corrected for the sum effect and for attenuation (reference date was the date of collection). According to the method described in Radioactivity measurement series No. 7[6], the concentrations of ^{134}Cs and ^{137}Cs were calculated. We define the concentration of radioactive cesium as the sum of the individual concentrations of ^{134}Cs and ^{137}Cs.

4. Measurement of ^{90}Sr

Measurement of ^{90}Sr was carried out in accordance with the ion-exchange method or ferric chloride co-precipitation method as described in Radioactivity measurement series No. 2[7]. The flow chart corresponding to the ^{90}Sr analysis method is summarized in Fig. 1.

Food samples were ashed at 600℃ for 27 hr. Ash samples corresponding to 1 kg fresh weight were used for ^{90}Sr analysis. If the sample amount was small or included high levels of calcium, the amount of sample ash used for ^{90}Sr analysis was reduced. β-Rays emitted by ^{90}Y were counted for 60 min per sample by a low back-

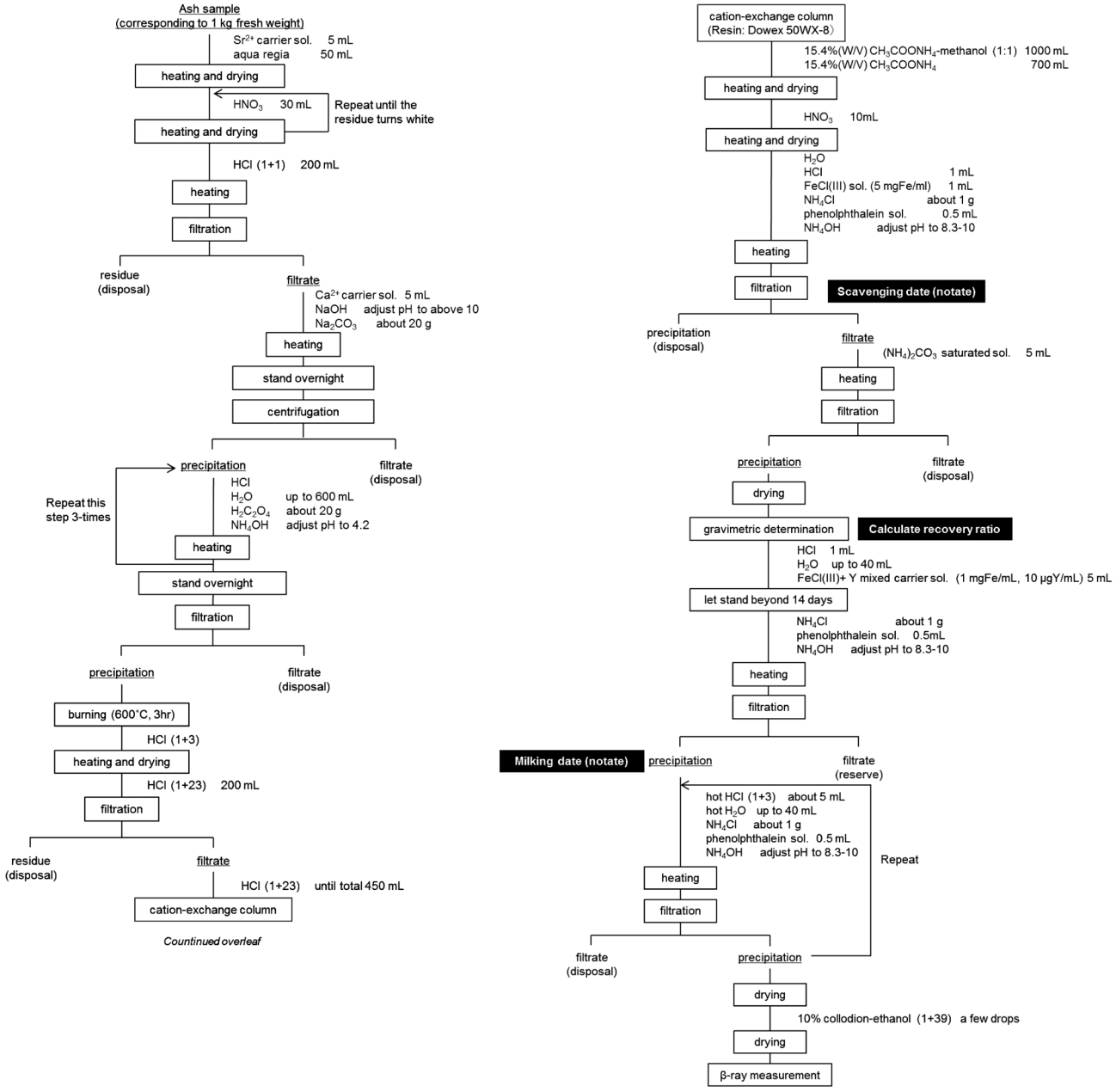

Fig. 1. Flow chart fo ^{90}Sr analysis method used in this study

Table 3. Results of ^{90}Sr analysis (r-Cs negative samples)

Sample[1), 2)]	Food category	Weight of sample used for Sr analysis		Total Cs	^{137}Cs	^{90}Sr			Sr recovery rate
		Ash weight	Fresh weight (Corresponding value)	Radioactive conc.[3)]	Radioactive conc.[3)]	Radioactive conc.[3),4)]	Counting error	LOD[5)]	
		(g)	(g)	(Bq/kg)	(Bq/kg)	(Bq/kg)	(Bq/kg)	(Bq/kg)	
Pond smelt NA*[1]	Freshwater fish and shellfish	14.94	780	＜0.2	＜0.1	0.052	0.008	0.02	1.357
Pond smelt NB*[1]		15.94	740	＜0.3	＜0.2	0.028	0.008	0.02	1.178
Jpanese trout (small size) NA*[1]		19.50	820	＜0.3	＜0.2	0.17	0.01	0.02	0.900
Brown rice NA	Rice	12.26	1000	＜2.2	＜0.8	*	*	0.02	0.837
Well-milled rice NA		4.36	1000	＜1.7	＜0.8	*	*	0.02	0.920
Well-milled rice NB		4.45	1000	＜1.5	＜0.7	*	*	0.02	0.941
Soy bean NA	Beans	18.14	300	3.4	3.4	0.15	0.03	0.06	0.934
Beef NA	Beef	3.98	580	＜0.2	＜0.09	*	*	0.03	0.942
Beef NB		4.08	520	＜0.2	＜0.09	*	*	0.04	0.886
Dried shiitake mushroom BA*[2] (Harvested in 2010)	Others	12.60	300	20	20	0.88	0.06	0.07	0.848
Mulberry leaf tea NA*[2]		15.00	100	＜2.8	＜1.5	2.0	0.1	0.2	0.865
Mulberry leaf tea NB*[2]		15.00	130	＜1.5	＜0.8	3.0	0.1	0.2	0.958
Mulberry leaf tea NC*[2]		15.00	110	＜1.8	＜0.9	5.5	0.2	0.2	0.973

1) *[1] indicates that samples were analyzed whole (including bones).
2) *[2] indicates dried samples. Weight of these samples was not fresh weight but dried weight.
3) Radioactivity of cesium (^{134}Cs and ^{137}Cs) and ^{90}Sr was corrected for attenuation (reference date was the date of sample collection).
4) In the case that the counting rate is more 3-times higher than the counting error, concentration of ^{90}Sr is shown to two significant figures. In the case that the counting rate is less than 3-times higher than the counting error, concentration of ^{90}Sr is shown as *.
5) Limit of detection (LOD) of ^{90}Sr is shown to one significant figure.

ground 2π gas-flow counter (LBC-4302B; Hitachi-Aloka Medical). Radioactivity concentration of ^{90}Sr was calculated by counting the β-rays emitted by ^{90}Y. The concentration of ^{90}Sr was corrected for attenuation (reference date was the date of collection) and the Sr recovery rate. In this study, Sr recovery rates in most samples were above 75% (Tables 2 and 3). Strontium-90 concentration in flatfish was used only for reference purposes (a reference value) because the Sr recovery rate was significantly low. Prior to analyzing food samples, certification samples such as IAEA-152, -156 and -330 were analyzed to validate our analytical method. All obtained values were very close to the certified values, within the limits of uncertainty.

Results and Discussion

1. *General*

Strontium-90 analysis results are shown in Table 2 (r-Cs positive samples) and Table 3 (r-Cs negative samples). Strontium-90 was detected in 25 of 40 r-Cs positive samples. Detected ^{90}Sr concentrations in fresh and dried samples were 0.028–0.87 Bq/kg fresh weight (fw) and 0.42–3.7 Bq/kg dry weight (dw), respectively. Strontium-90 concentration in dried samples tended to be higher than in fresh samples due to the lower water content. Among fresh samples, edible wild plants and wild mushrooms contained relatively high concentrations of ^{90}Sr. The limits of detection (LODs) in some non-detectable samples were 1.5- to 10-fold higher than the lowest LOD (0.02 Bq/kg) in the ^{90}Sr analysis due to the small initial sample size. The ^{90}Sr levels were generally relatively low (up to 3.7 Bq/kg), being approximately 1 to 4 orders of magnitude lower than the ^{137}Cs levels in the respective samples.

Strontium-90 was also detected in 8 of 13 r-Cs negative samples, considered to be unaffected by the Fukushima Daiichi NPP accident. Dried shiitake mushrooms harvested in 2010, soybeans and mulberry leaf teas contained relatively high concentrations of ^{90}Sr. Also, it appeared that Japanese trout contained relatively high concentrations of ^{90}Sr, because this sample was analyzed as the whole body including bones. In fact, according to the results of the radioactivity and radiation surveys carried out since 1957 by the Japan Chemical Analysis Center, ^{90}Sr was detected in many kinds of foods before the Fukushima Daiichi NPP accident*[5]. The results of the surveys before the accident were divided into two periods (1957–1999 and 2000–2010) and were categorized according to the food category in our study, as shown in

*[5] Japan Chemical Analysis Center, Environmental Radioactivity and Radiation in Japan., http://search.kankyo-hosha-no.go.jp/servlet/search.SelectMain?paraSelectKind=0&pageSID=215453534.

Table 4. Results of ^{90}Sr analysis before the Fukushima Daiichi NPP accident extracted from environmental radioactivity database[1)]

Surveillance period	1957–1999			2000–2010		
Sample	Minimal radioactive conc. (Bq/kg)[2)]	Maximal radioactive conc. (Bq/kg)[2)]	Number of samples	Minimal radioactive conc. (Bq/kg)[2)]	Maximal radioactive conc. (Bq/kg)[2)]	Number of samples
Sea fish	ND	2.6751	2548	ND	0.094	1328
Freshwater fish	ND	29.6	354	ND	3.2	139
Beef	ND	ND	16	ND	ND	38
Well-milled rice	ND	0.444	1812	ND	0.11	1002
Brown rice	ND	0.851	596	ND	0.093	121
Crude wheat	ND	5.6166	261	ND	0.352	57
Green tea	ND	151.922	778	ND	1.5	354
Soybean	0.16	0.94	10	—	—	0
Shiitake mushroom	ND	2.5	15	—	—	0

[1)] This data are summarized data from two databases:
http://search.kankyo-hoshano.go.jp/servlet/search.SelectMain?paraSelectKind=0&pageSID=215453534
http://search.kankyo-hoshano.go.jp/food/servlet/food_in;jsessionid=2c30a3d29b79514d8bdd617a1c6951b1d792?pageSID=1006_2c30a3d29b79514d8bdd617a1c6951b1d792
[2)] ND means not detected. LOD was not given in the databases.

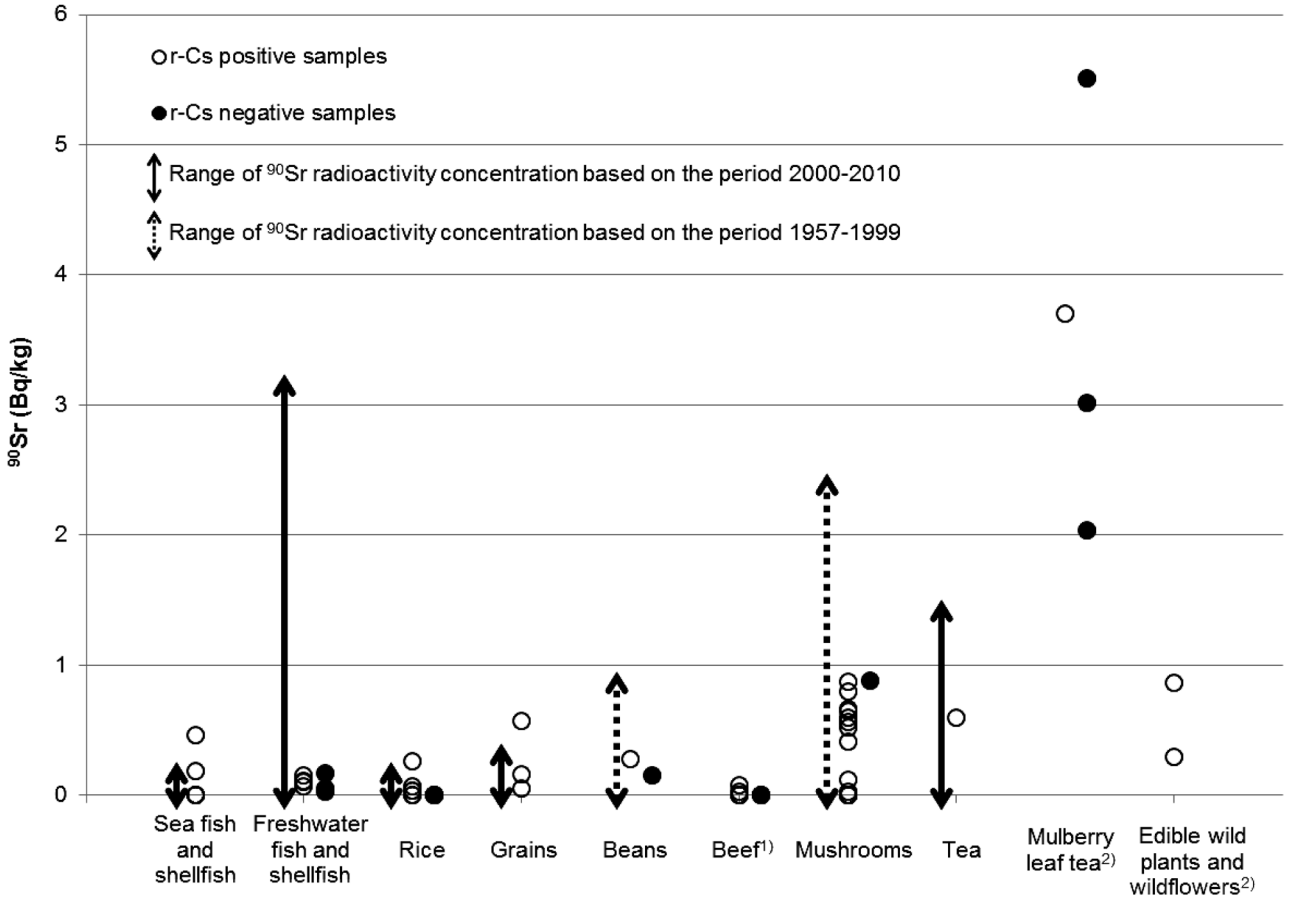

Fig. 2. Concentration of ^{90}Sr radioactivity each food category

Radioactive concentration of ^{90}Sr was plotted for each food category. Open circles (○) indicate radioactive r-Cs positive samples and filled circles (●) indicate radioactive r-Cs negative samples. Double-headed arrows indicate range of ^{90}Sr radioactivity concentration based on past data (before the Fukushima Daiichi NPP accident) as reported in the environmental radioactivity database.
[1)] Even though ^{90}Sr radioactive concentration in this food classification was measured, ^{90}Sr radioactive concentration was below the lowest detection limit in all of samples.
[2)] No past data (before the Fukushima Daiichi NPP accident) were reported.

Table 5. Concentration ratios of ^{90}Sr and ^{137}Cs in r-Cs positive samples

Sample[1), 2)]	Food category	^{90}Sr/^{137}Cs Ratio of conc. (Observed value)[3)]	^{90}Sr/^{137}Cs Ratio of conc. (Estimated value)[4)]
Codfish A	Sea fish and shellfish	0.0029	—[5)]
Codfish B		0.00063	
Fat greenling		0.0039	
Flatfish*1		0.027	
Marbled flounder*1		0.0095	
Pond smelt A*1	Freshwater fish and shellfish	0.0039	0.000024
Pond smelt B*1		0.0050	
Pond smelt C*1		0.0086	
Loach*1		0.025	
Rice bran A	Rice	0.00059	0.010
Rice bran B		0.00047	
Brown rice A		0.00065	
Brown rice B		0.00044	
Brown rice C		0.00053	
Brown rice D		0.0013	
Crude wheat A	Grains	0.0024	0.0060
Crude wheat B		0.00071	
Wheat bran		0.00099	
Soy bean	Beans	0.0020	0.10
Beef A	Beef	0.000067	0.0096
Beef B		0.000043	
Beef C		0.000067	
Beef D		0.000057	
Beef E		0.000067	
Shiitake mushroom A	Others	0.0012	—[5)] (0.033)[6)]
Shiitake mushroom B		0.0019	
Shiitake mushroom C		0.00087	
Shiitake mushroom D		0.0021	
Shiitake mushroom E		0.00061	
Shiitake mushroom F		0.0036	
Dried shiitake mushroom A*2		0.00032	
Dried shiitake mushroom B*2		0.0047	
Nameko mushroom		0.00051	
Kuritake mushroom		0.00076	
Tawny milkcap mushroom		0.000047	
Mixture of several kinds of mushrooms		0.0012	
Green tea*2		0.00033	
Mulberry leaf tea*2		0.021	
Horsetail		0.0017	
Bracken		0.0010	

1) *1 indicates that samples were analyzed whole (including bones or shell).
2) *2 indicates dried samples. Weight of these samples was not fresh weight but dried weight.
3) In the case that radioactive concentration of ^{90}Sr was less than its lower detection limit, the ratio of concentration between ^{90}Sr and ^{137}Cs (^{90}Sr/^{137}Cs) was calculated using the lower detection limit value of ^{90}Sr.
4) Estimated ratio of concentration between ^{90}Sr and ^{137}Cs was calculated based on information from ref. 2.
5) — indicated that ^{90}Sr/^{137}Cs ratio was not established.
6) The reference ratio in parentheses was calculated as described in Results and Disccusion.

Table 4. Because the previous data from the 1950's to 1980's were heavily influenced by nuclear bomb tests and the Chernobyl accident, our data were instead compared to the results of the survey for the decade previous to the Fukushima Daiichi NPP accident except where data were absent. Beans and mushrooms, which were not surveyed from 2000 to 2010, were compared to the results of the survey from 1957–1999. The ^{90}Sr concentrations of freshwater fish, rice, beans and mushrooms in r-Cs negative samples were within the range of the results from the surveys before the Fukushima Daiichi NPP accident. Our results suggest that the influence of past global fallout still remains in the r-Cs negative samples. Similarly, it is suspected that some ^{90}Sr

derived from past global fallout is also included in the r-Cs positive samples.

Strontium-90 concentrations of r-Cs positive samples and r-Cs negative samples are plotted for each food category in Fig. 2. Strontium-90 concentrations in r-Cs positive samples did not significantly exceed those in r-Cs negative samples or the ^{90}Sr concentration ranges in comparable food categories from past surveys occurring before the Fukushima Daiichi NPP accident. Therefore, we could not any clear increase in ^{90}Sr concentrations in r-Cs positive samples as a consequence of the Fukushima Daiichi NPP accident.

As described above, the new standard limits for radionuclide ingestion include the contributions of ^{134}Cs, ^{137}Cs, ^{90}Sr, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{106}Ru. The contributions of target radionuclides other than r-Cs were calculated by estimating radioactive concentration ratios of other radionuclides to ^{137}Cs. The observed radioactive concentration ratios of ^{90}Sr to ^{137}Cs in our analyzed samples are shown in Table 5, along with the estimated ratios from the report concerning the new standard regulatory limits for radionuclides in foods[2]. The observed ratios (^{90}Sr/^{137}Cs ratio) were less than the estimated ratios except in the case for freshwater fish and shellfish. This result supports view that the estimated ^{90}Sr/^{137}Cs ratios used for the new limits were set on the safe side for the food categories considered here. While ^{90}Sr/^{137}Cs ratios in freshwater fish and shellfish were higher than the estimated ratios, this could be because we analyzed the whole body, including bones and shells, which are known to accumulate ^{90}Sr[3]. The ^{90}Sr/^{137}Cs ratio in freshwater fish and shellfish was estimated using the transfer factor ratio (^{90}Sr/^{137}Cs=0.0012) from fresh water to muscular tissues. This transfer factor ratio is much smaller than that from fresh water to the whole body[8]. This is consistent with the observation that the ^{90}Sr/^{137}Cs ratio from the whole body sample was higher than the estimated one. We attempted to calculate the induced limiting value of r-Cs through the consumption of foods using the highest observed ratio (0.025) in freshwater fish and shellfish in the same manner as the standard limit for males between the ages of 13–18. The induced limiting value of r-Cs was 120 Bq/kg (rounding down the third digit), which is the same as the original induced limiting value[2]. Thus, even such a significant increase of ^{90}Sr/^{137}Cs ratio as was seen in freshwater fish and shellfish did not affect the induced standard limit, most likely because of the very low daily consumption of freshwater fish and shellfish in a normal diet.

2. *Food categories*

2.1 *Sea fish and shellfish*

Because sea fish containing relatively high concentrations of r-Cs were difficult to obtain, we were only able to analyze 5 r-Cs positive samples collected in 2012 and 2013. For 2 out of 5 samples, the whole body was analyzed without removal of bones because of the small amount of these samples. Although ^{90}Sr was not detected in 3 samples analyzed using only the edible part (mainly muscle), 0.18 and 0.46 Bq/kg of ^{90}Sr were detected in the 2 samples analyzed using the whole body. It is well known that ^{90}Sr absorbed into the body is highly distributed in bone[3], therefore it was considered that ^{90}Sr was indeed detect in these two samples. According to the environmental radioactivity database, the range of ^{90}Sr concentration in sea fish for a decade before the Fukushima Daiichi NPP accident was ND-0.094 Bq/kg (Table 4). These data were obtained from analysis of edible parts, mainly muscle. Because ^{90}Sr concentration in flatfish was regarded as a reference value due to its low Sr recovery rate, we compared our data on marbled flounder to the previous data on sea fish. The ^{90}Sr concentrations in the whole marbled flounder were about 2-fold higher than the previous data, because bones were included in the analyzed samples.

In sea fish and shellfish, the ^{90}Sr/^{137}Cs ratio was not established because the amount of outflow of radionuclides from the Fukushima Daiichi NPP accident into the sea remains unclear. Therefore, the standard limit was established based on the hypothesis that the committed effective dose ratio of the target radionuclides other than r-Cs to r-Cs was 0.5[2]. In the case of internal exposure as a result of eating or drinking contaminated foods, the influence on humans is estimated by calculating the committed effective dose, because orally ingested radionuclides cause long-term cumulative effects. The committed effective dose ratio of ^{90}Sr to r-Cs was calculated using the observed concentrations of ^{90}Sr and r-Cs (if ^{90}Sr concentration was less than LOD, the value of LOD was used as the ^{90}Sr concentration). The committed effective dose coefficients for adults (mSv/Bq) used for calculation were 2.8×10^{-5} for ^{90}Sr, 1.9×10^{-5} for ^{134}Cs and 1.3×10^{-5} for ^{137}Cs[9]. As a result of these calculations, the maximum committed effective dose ratio was determined to be 0.063, which is sufficiently lower than 0.5.

2.2 *Freshwater fish and shellfish*

Three pond smelts collected in 2012 and a loach collected in 2013 were analyzed as r-Cs positive samples, while two pond smelts and a small Japanese trout collected in 2013 were analyzed as r-Cs negative samples. These samples were analyzed as the whole body including bone because these small freshwater fishes are commonly eaten whole. Concentration of ^{90}Sr in the r-Cs positive samples (pond smelt A–C) were in the range of 0.073–0.15 Bq/kg (Table 2). Although pond smelt NA, NB and Japanese trout were r-Cs negative samples, ^{90}Sr was detected in the range of 0.028–0.17 Bq/kg. The concentration ranges of the above r-Cs positives and negatives were similar. This fact suggests that some ^{90}Sr was contained in freshwater fish independently of the Fukushima Daiichi NPP accident. Briefly, it is considered that some effects of past fallout still remain. The ^{90}Sr/^{137}Cs ratios in r-Cs positive freshwater fish and shell were 0.0039–0.025, which were higher than the estimated concentration ratio (0.000024) (Table 5). The reason why the observed ^{90}Sr/^{137}Cs ratios in our study were

markedly higher than the estimated ratio was that the observed ratios were calculated using concentrations of ^{90}Sr and ^{137}Cs in whole-body freshwater fish samples including bones, which contain an abundance of Sr, whereas the estimated ratios were derived using the transfer factor ratio from fresh water to muscular tissues. However, as discussed above, even the significant increase in ^{90}Sr/^{137}Cs ratio in freshwater fish and shellfish did not affect the induced standard limit.

2.3 Rice, grains and beans

Four brown rice and two rice brans were analyzed as r-Cs positive samples. These samples were all harvested in 2011. Additionally, one brown rice and two well-milled rice samples were also analyzed as r-Cs negative samples. All three r-Cs negative samples had been harvested before the Fukushima Daiichi NPP accident. Although ^{90}Sr was not detected in any r-Cs negative rice, ^{90}Sr was detected in four out of six r-Cs positive rice samples. Strontium-90 concentrations in the r-Cs positive brown rice and rice bran were 0.036–0.071 and 0.27 Bq/kg, respectively. The ^{90}Sr concentrations in brown rice were within the range of the past data. Strontium-90 concentration in rice bran was similar to the previously reported values[10]. It was considered that ^{90}Sr was not detected in rice bran A because the LOD in rice bran A was 10-fold higher than the lowest LOD due to the small sample size. Tsukada et al. reported that ^{90}Sr concentration in rice bran was 26–29-fold higher than in well-milled rice[10]. Furthermore, from their reported data on ^{90}Sr concentration and weight of rice plant components[10], ^{90}Sr concentration in brown rice was calculated and compared to that in rice bran. The result was that the ^{90}Sr concentration in rice bran was 7.4–7.6-fold higher than that in brown rice. These values were also in a similar range of magnitude to our results (3.8–7.5-fold).

Two crude wheat and wheat bran samples harvested in 2011 were analyzed for ^{90}Sr as r-Cs positive grain samples. Strontium-90 was detected in all the samples. As in the case of rice, ^{90}Sr concentration in wheat bran was 3.5 to 10-fold higher than that in crude wheat. The ^{90}Sr concentrations in crude wheat were within the range of previous data reported in 2000–2010. Strontium-90 concentration in wheat bran could not be compared with past data because ^{90}Sr concentration in wheat bran in Japan has not been reported. However, previous data in the United States in 1959 showed that ^{90}Sr concentration in wheat bran and crude wheat were 11 and 1.8 Bq/kg, respectively[11]. That is, ^{90}Sr concentrations in wheat bran were 6.4-fold higher than in crude wheat. Although these previous ^{90}Sr concentrations were markedly higher than our data, they are consistent with our finding that ^{90}Sr concentration in wheat bran was higher than in crude wheat.

Strontium-90 concentration of a r-Cs positive soybean sample harvested in 2011 and a negative soybean sample harvested in 2013 were 0.28 Bq/kg and 0.15 Bq/kg, respectively. There was about a 70-fold difference of r-Cs concentration between the two samples, whereas there was only 2-fold difference of ^{90}Sr concentration between them. The ^{90}Sr concentrations of both samples were within the range of past data reported in 1957–1999 (Table 4).

Our results showed that ^{90}Sr concentration from the edible parts of rice, grains and soybeans were within the range of past data before the Fukushima Daiichi NPP accident, except for exodermises of rice and wheat (rice bran and wheat bran). Observed ^{90}Sr/^{137}Cs ratios in r-Cs positive rice, grains and soybean were 0.00044–0.013, 0.00071–0.0024 and 0.0020, respectively (Table 5). The observed concentration ratios in all the analyzed samples were lower than the estimated concentration ratios.

2.4 Beef

Five r-Cs positive beef samples collected in 2011 and two r-Cs negative beef samples collected in 2013 were analyzed for ^{90}Sr. ^{90}Sr was detected in only two r-Cs positive samples. Their r-Cs concentrations were the top two among five r-Cs positive beef. Strontium-90 concentrations in the two beef samples were 0.028 and 0.081 Bq/kg contributing to a lower concentration than in ^{90}Sr detected samples in other food categories. According to past reports from 1957–2010, there were no instances where ^{90}Sr was detected in beef. However, considering that the detected ^{90}Sr was only slightly higher than the LODs in our samples, it is difficult to determine whether the ^{90}Sr detected in the beef in this study was derived from the Fukushima Daiichi NPP accident. Observed ^{90}Sr/^{137}Cs ratios in beef were 0.000043–0.000067, and were over 100-fold lower than the estimated concentration ratio (0.0096).

2.5 Others

This category includes mushrooms, edible wild plants, wildflowers and tea.

As r-Cs positive samples, eight shiitake mushrooms including two dried shiitake mushrooms, four wild mushrooms including nameko, kuritake, tawny milkcap mushroom and a composite of several kinds of wild mushrooms, were analyzed for ^{90}Sr. Most of these samples were harvested in 2013, except for the tawny milkcap mushroom and wild mushrooms composite sample, which were harvested in 2012. In addition, one r-Cs negative dried shiitake mushroom harvested in 2010 was also analyzed. Strontium-90 was detected in four out of eight r-Cs positive shiitake mushrooms, and one r-Cs negative dried shiitake mushroom. By converting the ^{90}Sr concentrations (Bq/kgdw) in r-Cs positive dried shiitake mushrooms into Bq/kgfw using the ratio of water contents described in standard tables of food composition in Japan[12], the ^{90}Sr concentration in dried shiitake mushroom A and B were calculated to correspond to 0.045 and 0.085 Bq/kgfw, respectively. These values were close to the ^{90}Sr concentration in r-Cs positive fresh shiitake mushrooms. The maximum ^{90}Sr concentrations from the radioactive r-Cs positive dried shiitake mushrooms harvested after the Fukushima Daiichi NPP acci-

dent were similar to that from the r-Cs negative dried shiitake mushroom harvested before the accident. However, because we have only one result for r-Cs negative dried shiitake mushroom, it would be necessary to gather more data to reach any conclusion. On the other hand, ^{90}Sr concentrations in r-Cs positive fresh shiitake mushrooms were over 10-fold lower than the maximum value of past data covering 1957–1999. Among the wild mushrooms, ^{90}Sr was detected in one out of four samples. Strontium-90 concentration in the wild mushrooms tended to be slightly higher than that in cultivated shiitake mushrooms. Because a ^{90}Sr/^{137}Cs ratio was not established in the category of others[2], we calculated the reference ^{90}Sr/^{137}Cs ratios according to the weighted average of ^{90}Sr/^{137}Cs ratios in each food category based on the daily food consumptions in various age groups. Finally, the lowest ratio (0.033) among the age groups was used as the reference value in the food category. Observed ^{90}Sr/^{137}Cs ratios in the wild mushrooms were sufficiently lower than the reference value, because the wild mushrooms contained relatively high-concentration of r-Cs.

Concentrations of ^{90}Sr in edible wild plants and wildflowers harvested in 2013, i.e., horsetail and bracken, were 0.30 and 0.87 Bq/kg, respectively. Because there were no available data regarding ^{90}Sr concentrations in edible wild plants and wildflowers before the Fukushima Daiichi NPP accident in Japan, it is unclear whether ^{90}Sr concentrations in these categories had increased or not after the accident. Therefore, it would be necessary to gather further data regarding ^{90}Sr concentration in r-Cs negative edible wild plants and wildflowers that have not been affected by the accident.

Green tea and mulberry leaf tea were analyzed using dried leaves and not tea infusions. R-Cs positive green tea harvested in 2011 and mulberry leaf tea harvested in 2012 contained 0.60 and 3.7 Bq/kg of ^{90}Sr, respectively. Strontium-90 concentrations in the mulberry leaf tea were over 5-fold higher than that in the green tea, whereas r-Cs concentration in the mulberry leaf tea was one-tenth of that in the green tea. Strontium-90 was also detected in all three r-Cs negative mulberry leaf tea samples harvested in 2013 in the range of 2.0–5.5 Bq/kg. Relatively high concentrations of ^{90}Sr were found in mulberry leaf tea regardless of r-Cs contamination. Our result also showed that mulberry leaf tea contained high concentrations of stable Sr compared to other samples. For example, stable Sr concentrations of all four mulberry leaf tea samples harvested in different areas were similar to each other and 5-fold higher than that in green tea (data not shown). These results suggest that the mulberry leaf tea had a relatively high concentrations of ^{90}Sr compared to other foods due to not impact from Fukushima Daiichi NPP accident but an intrinsic propensity to accumulate Sr in its leaves.

Observed ^{90}Sr/^{137}Cs ratios were sufficiently lower than the estimated ratio (0.033 as reference value) in all the r-Cs positive samples categorized as others.

Conclusion

We detected ^{90}Sr in 25 out of 40 r-Cs positive samples and 8 out of 13 r-Cs negative samples. The ^{90}Sr concentrations in sea fish, freshwater fish, rice, grains, beans, beef, and other foods did not significantly exceed reported concentrations before the Fukushima Daiichi NPP accident or those from r-Cs negative samples. However, the concentrations in some samples such as edible wild plants and wildflowers could not be compared to data acquired before the Fukushima Daiichi NPP accident or from those of r-Cs negative samples. Thus, it could not be determined whether there was a significant increase in ^{90}Sr concentrations from r-Cs positive samples after the accident.

Our results also showed that the observed ^{90}Sr/^{137}Cs ratios in r-Cs positive samples were sufficiently lower than the estimated ^{90}Sr/^{137}Cs ratios except in freshwater fish and shellfish. However, this increase of ^{90}Sr/^{137}Cs ratio observed in freshwater fish and shellfish did not affect the limit set to define the standard regulatory limit. The results indicate that the current standard limit was established safely and conservatively. However, we were only able to analyze limited samples harvested relatively far from the Fukushima Daiichi NPP in limited food categories for this study. We will continue to analyze the ^{90}Sr in various food samples including samples harvested from neighboring areas of the Fukushima Daiichi NPP in order to collect more data for future reference.

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