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Special Report: “Radioactivity in the Marine Environment and in Fisheries Products during the Five Years after the Fukushima Dai-ichi Nuclear Power Plant Accident”

Trend of Radioactivity in Fisheries Products

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Abstract: We measured activities of radioactive cesium (^{134}Cs and ^{137}Cs) in fisheries products (38,241 samples, 355 species) obtained from September 2011 to March 2016. The samples were caught in marine areas off Eastern Honshu Japan, but not including areas off Fukushima Prefecture, and from freshwater areas in Eastern Honshu, but not including Fukushima Prefecture. Activities that exceeded the national standard limit (100 Bq/kg) were detected in some samples of marine organisms from the Pacific Ocean off eastern Japan from Aomori Prefecture to Chiba Prefecture, and of freshwater organisms in areas of eastern Japan likely affected by surface contamination of radioactive substance. Of all the samples, the percentage containing radioactive cesium concentration above the national standard limit has declined over time since the March 2011 disaster at the Fukushima Dai-ichi nuclear power plant. This percentage has been 0% for marine products since September 2014 and less than 1% for freshwater products since October 2015.

Keywords: radioactive cesium, fisheries products, marine areas, freshwater areas, Fukushima, nuclear power plant

Introduction

The March 2011 accident at the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) contaminated marine areas near Japan with airborne radioactive materials and/or radioactive substance-contaminated runoff. The accident led to detection of radioactive cesium (^{134}Cs and ^{137}Cs) in some commercial fish. When radioactive cesium enters an organism’s body, it tends to take in the muscle and other edible parts. This tendency makes monitoring radioactive cesium activities in marine and freshwater organisms an urgent task from the standpoint of food safety. The standard limit of the Japanese Ministry of Health, Labour, and Welfare for radioactive cesium in general food products was set at 100 Bq/kg on 1 April 2012. The Marine Ecology Research Institute (MERI) has been commissioned by the Japanese Fisheries Agency with the task of surveying the effects of radioactive substances in fisheries products collected from marine areas off Eastern Honshu in Japan (not including those off Fukushima Prefecture, where commercial fisheries have voluntary suspended operations since the nuclear disaster) and freshwater areas in Eastern Honshu (but not including those in Fukushima Prefecture). We began these surveys in September 2011 and they continue to the present day. The results of these surveys have been published on the website of the Fisheries Agency (Fisheries Agency, 2016), and trends in radioactive cesium activities in commercial fish up to March 2015 have been reported by Yokota *et al*. (2013, 2014, 2015a, 2015b). In this report, we describe the trends in radioactive cesium activities in fisheries products from marine and freshwater areas up to March 2016.

Methods

Survey

The fisheries products were collected by 14

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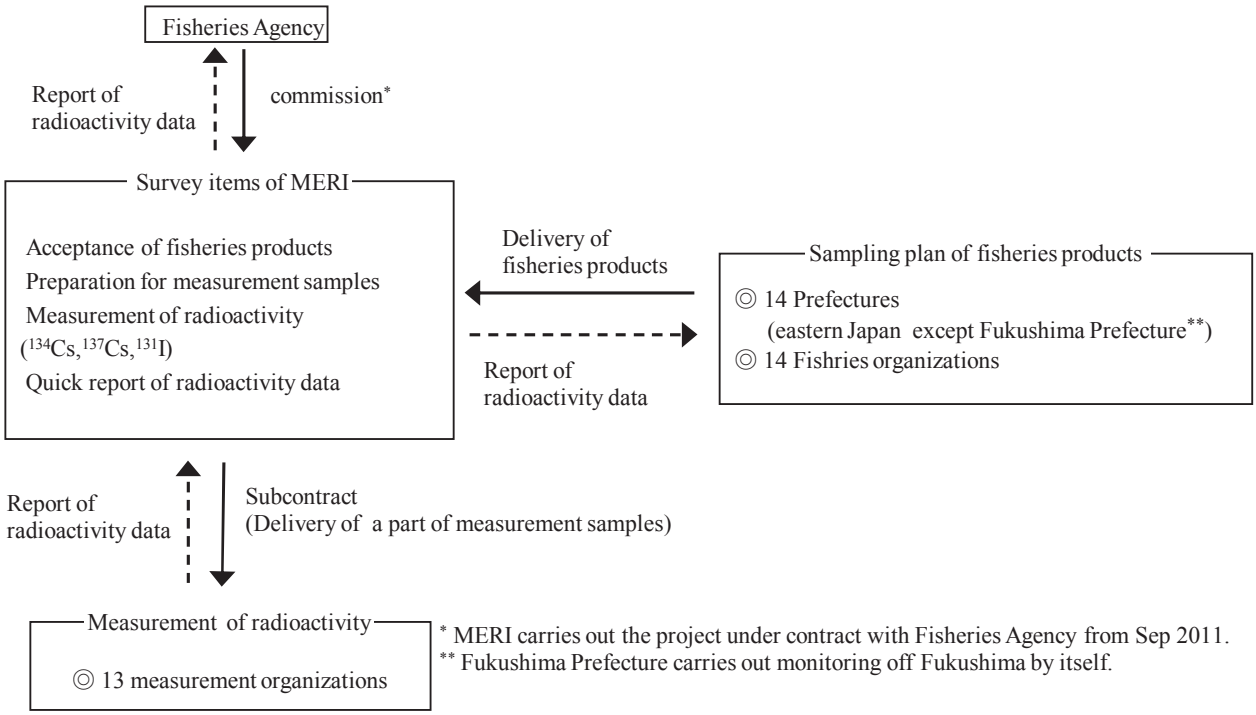


Fig.1 Flow of the project commissioned by Fisheries Agency.

Fisheries Organizations and 14 prefectures in eastern Japan. The samples were then sent to the MERI Central Laboratory in accordance with the collection plans of these groups (Fig.1). The fisheries products were first minced in sample preparation rooms and then sent to the analysis office in the Central Laboratory or to outside institutes for analysis of radioactivity. MERI quickly notified the relevant entities of the results of fisheries products analyses, which were then released to the public. MERI analyzed and summarized the effects of radioactive substances on fisheries products based on the data.

Analytical methods

After identifying the species of each fish, its muscle, liver, ovaries, testes, and other edible parts were collected (Fig.2). In principle, the edible parts from several individuals were mixed together and minced to produce a fresh sample for analysis. Some of the sample were sent to outside institutes for radioactivity analysis. In principle, about 2,000 g of a sample was placed in a 2-L Marinelli container. If this was not possible, about 100 g were placed in a 100-mL U-8 container.

Radioactivity analyses were performed by gamma-ray spectrometry using a germanium semiconductor detector (SEIKO EG&G Co. or Canberra Japan KK.) to measure the activities of ^{134}Cs (half-life of about 2 years) and ^{137}Cs (half-life of about 30 years) in the samples. In principle, radioactivities were calculated per kg of wet weight (Bq/kg-wet), except for seaweeds, which were sent in a dried state (Fig.3). The methods of analysis of the Japanese Ministry of Education, Culture, Sports, Science, and Technology (1992) were followed. In principle, analyses were performed within 24 hours after delivery to an analytical institute. The measurements of radioactivity took one hour for both the 2-L Marinelli containers and the 100-mL U-8 containers.

Results and Discussion

Marine organisms

Of the 30,059 samples (313 species) that underwent radioactivity analyses from September 2011 to March 2016, radioactive cesium activities exceeding

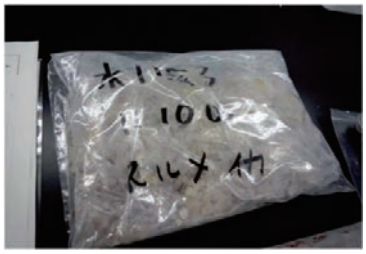
1. Acceptance of fisheries products

4. Extraction of edible part

2. Identification of fish species

5. Mincing of edible part

3. Mesurement of length and weight of fisheries products



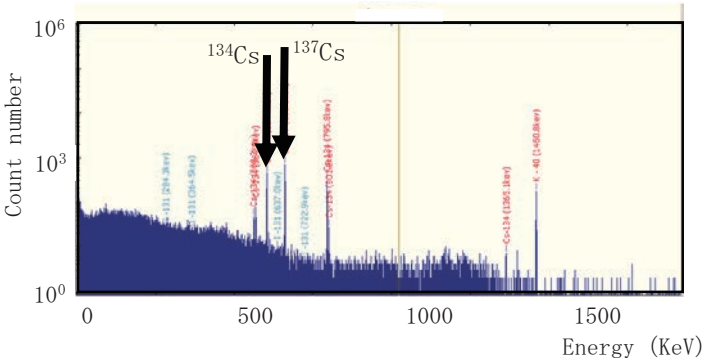
6. Sample for measurement of radioactivity

Fig.2 Preprocessing to analyze the radioactivity of fisheries products.

1. Packing minced sample to a container

2. Setting a sample to γ-ray detector

3. Counting γ activity



4. An example of γ spectrum of a sample

Fig.3 Radioactivity measurement.

Table 1 The number of sample of marine products with $^{134+137}$Cs concentration of >100Bq/kg-wet. Data from the Fukushima offshore were excluded. Measurement samples in Sep 2011 - Mar 2016 were used for data analysis.

Type of fisheries products	Measurement part	The number of samples with >100 Bq/kg-wet	The total number of samples of five years
Fish	Muscle	62	23,586
	Whole	-	1,191
	Liver	-	890
	Testicle	-	205
	Ovary	-	451
	Heart	-	3
	Mixture	-	54
Squid	Muscle	-	917
	Whole	-	5
	Liver	-	81
Octopus	Muscle	-	633
Shrimp	Muscle	-	117
Crab	Whole	-	7
	Mixture	-	187
Squilla	Muscle	-	15
Shellfish	Muscle	-	210
	Soft body part	-	522
Sea cucumber	Muscle	-	29
Urchin	Gonad	-	30
Ascidian	Muscle	-	300
Krill	Whole	-	100
Mysid shrimp	Whole	-	1
Seaweed	Whole	-	431
Whale	Muscle	-	84
Other	Fish meal	-	6
	Fish oil	-	2
	Broth	-	2
Total		62	30,059

"-" represents no samples exceeded the 100 Bq/kg-wet level.

100 Bq/kg-wet were detected in the muscle of 62 fish samples (15 species) (Table 1). Apart from fish, radioactive cesium activities exceeding 100 Bq/kg-wet were not detected in any other marine organism, including species of squid, octopus, shrimp, crab, mantis shrimp, shellfish, sea cucumbers, sea urchins, ascidians, krill, Mysidacea, seaweeds, and whales.

Marine fish with radioactive cesium activities exceeding 100 Bq/kg-wet were collected in coastal areas on the Pacific Ocean side of eastern Japan (not including Fukushima), including Aomori, Iwate, Miyagi, Ibaraki, and Chiba prefectures (Fig.4). Most of the samples (58 of 62) of marine fish with radioactive cesium activities exceeding 100 Bq/kg-wet were collected off Miyagi and Ibaraki prefectures, which border on Fukushima (Table 2). All 15 species with

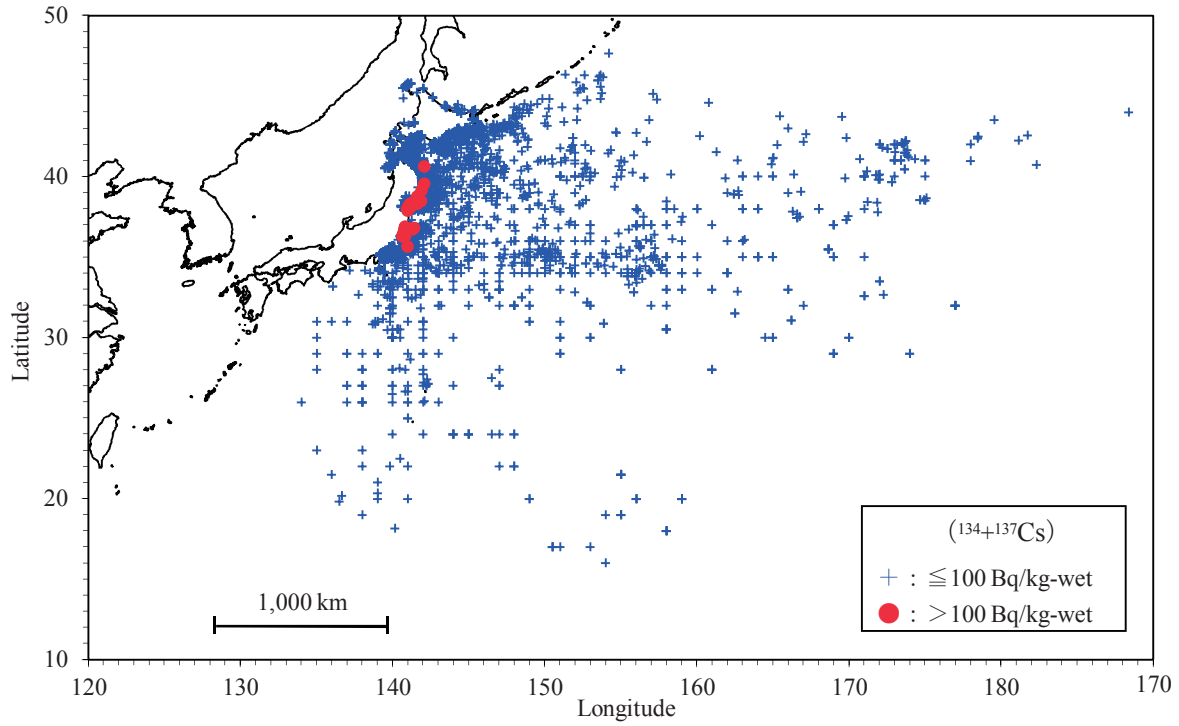


Fig.4 Distribution of radiocesium concentration in marine products (Sep 2011 - Mar 2016). Data from the waters off Fukushima Prefecture were excluded.

Table 2 Marine products with $^{134+137}$Cs concentration of >100Bq/kg-wet. Data from the Fukushima offshore were excluded. Measurement samples in Sep 2011 - Mar 2016 were used for data analysis.

Fish species	Offshore area					The total number of >100 Bq/kg-wet (The total number of samples)	Maximum value (Bq/kg-wet)
	Aomri	Iwate	Miyagi	Ibaraki	Chiba		
Acanthopagrus schlegelii	-	-	16	-	-	16 (188)	3,300
Lateolabrax japonicus	-	-	7	7	1	15 (1,667)	1,000
Okamejei kenojei	-	-	-	3	-	3 (379)	520
Paralichthys olivaceus	-	-	5	2	-	7 (2,034)	400
Sebastes schlegelii Hilgendorf	-	1	-	-	-	1 (209)	400
Microstomus achne	-	-	-	1	-	1 (612)	260
Pseudopleuronectes yokohamae	-	-	-	2	-	2 (835)	180
Hexagrammos otakii	-	-	-	1	-	1 (642)	170
Sebastes cheni	-	-	-	1	-	1 (147)	170
Gadus macrocephalus	1	-	4	3	-	8 (5,367)	160
Takifugu pardalis	-	-	1	-	-	1 (183)	140
Nibea mitsukurii	-	-	-	3	-	3 (129)	130
Sebastes thompsoni	-	-	-	1	-	1 (147)	120
Seriola quinqueradiata	-	1	-	-	-	1 (655)	110
Oncorhynchus kisutch	-	-	1	-	-	1 (37)	110
Total	1	2	34	24	1	62 (13,231)	

"-" represents no samples exceeded the 100 Bq/kg-wet level.

radioactive cesium activities exceeding 100 Bq/kg-wet appear in coastal areas shallower than 30 m. Moreover, the two species that exhibited radioactive cesium activities of 1,000 Bq/kg-wet or higher, *Acanthopagrus schlegelii* and *Lateolabrax japonicas*, can migrate to the brackish waters of rivers. The presence of high radioactive cesium activities in *A. schlegelii* and *L. japonicus* was likely influenced by the fact that the concentration factor of cesium is about 25 times higher in freshwater fish than in marine fish (IAEA, 2010). It may therefore be more difficult for fish in brackish areas to eliminate radioactive cesium.

The radioactive cesium activities detected in marine organisms collected off the Pacific Ocean side of Eastern Honshu (not including Fukushima) from April 2011 to March 2016 decreased over time. None of the samples has exhibited radioactive cesium activities exceeding 100 Bq/kg-wet since September 2014, and the radioactive cesium activities of most samples have fallen to 50 Bq/kg-wet or lower (Fig.5). Furthermore, the monthly percentage of samples with radioactive cesium activities exceeding 100 Bq/kg-wet (mean number of samples tested per month: 500) decreased from about 10% soon after the FDNPP accident to 0% in September 2014 (Fig.6).

Freshwater organisms

Of the 8,182 samples (42 species) that underwent radioactivity analysis from September 2011 to March 2016, radioactive cesium activities exceeding 100 Bq/kg-wet were detected in the muscle or whole bodies of 209 fish samples (15 species) (Table 3). Apart from fish, radioactive cesium activities exceeding 100 Bq/kg-wet were not detected in any other freshwater organisms, including species of shrimp, crab, and shellfish.

After radioactive substances emitted into the atmosphere from the FDNPP were deposited on the

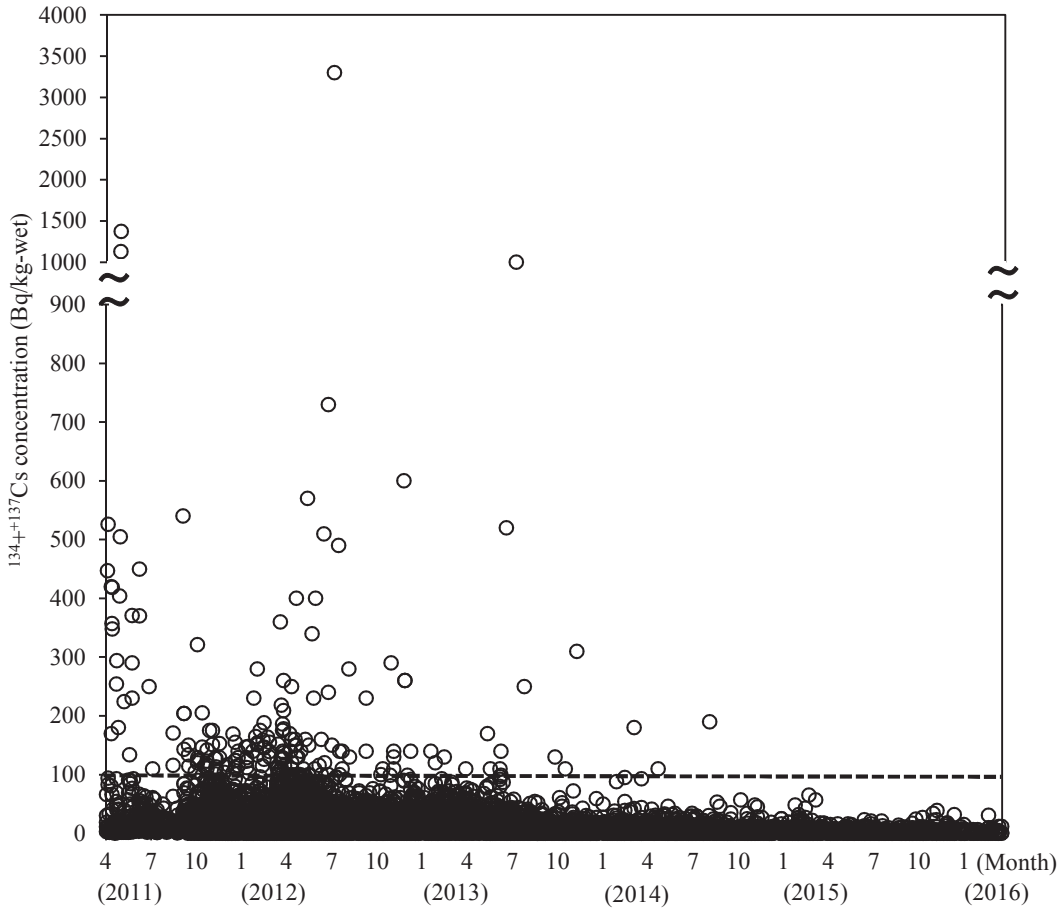


Fig.5 Temporal change of $^{134+137}$Cs concentration in marine products caught in the Pacific area of eastern Japan. Data adapted from Fisheries Agency (2016).

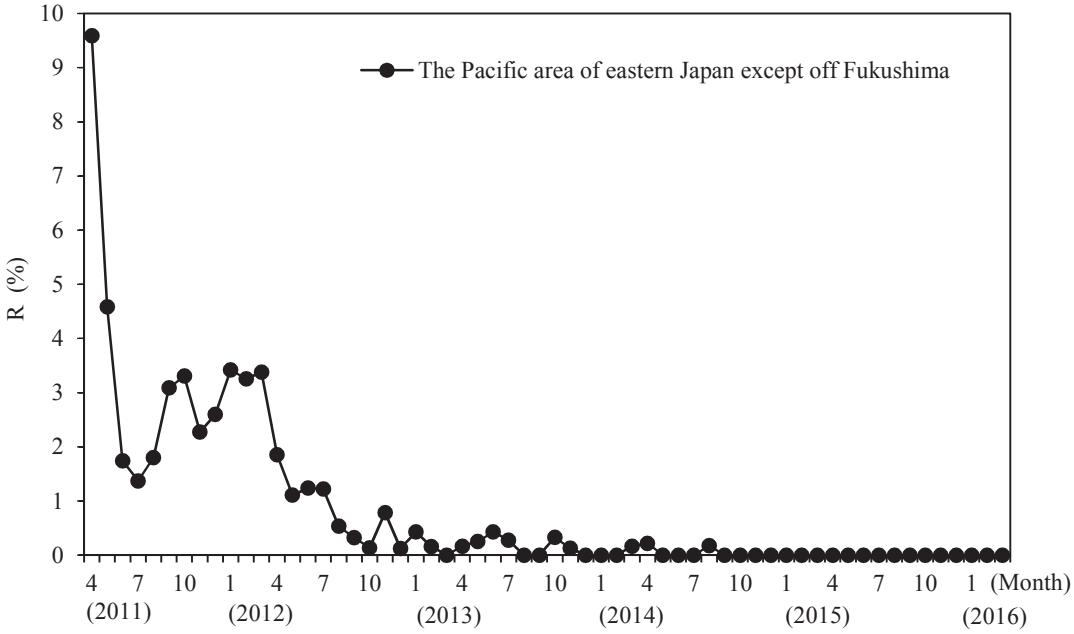


Fig.6 Temporal change of ratio (R) of samples with >100 Bq/kg-wet. The ratio were calculated based on the following equation using data adapted from Fisheries Agency (2016); R (%) = [The number of samples with concentration of >100 Bq/kg-wet] / [The number of all samples] x 100.

Table 3 The number of sample of freshwater products with $^{134+137}$Cs concentration of >100Bq/kg-wet. Data from the Fukushima offshore were excluded. Measurement samples in Sep 2011 - Mar 2016 were used for data analysis.

Type of fisheries products	Measurement part	The number of samples with >100 Bq/kg-wet	The total number of samples of five years
Fish	Muscle	180	5,400
	Whole	29	2,236
Shrimp	Whole	-	163
Crab	Whole	-	35
	Mixture	-	6
Shellfish	Soft body part	-	341
Softshell turtle	Whole	-	1
Total		209	8,182

"-" represents no samples exceeded the 100 Bq/kg-wet level.

ground, spatial radiation dose rates measured at a height of 1 m that exceeded 0.1 μSv/h were found over an extensive area (Japan Map Center et al., 2016). Freshwater fishes with radioactive cesium activities exceeding 100 Bq/kg-wet were collected from freshwater areas in Iwate, Miyagi, Ibaraki, Tochigi, Gunma, Chiba, Saitama, and Kanagawa prefectures of eastern Japan (Table 4). Samples with radioactivities exceeding 100 Bq/kg-wet were not collected from freshwater areas in prefectures on the Sea of Japan side, where there was little fallout of the airborne radioactive substances after the nuclear disaster. The distribution of radioactive cesium activities exceeding 100 Bq/kg-wet in freshwater fish was largely consistent with spatial radiation dose rates near the surface (Fig.7).

Sixty-six of the 209 freshwater fish samples with

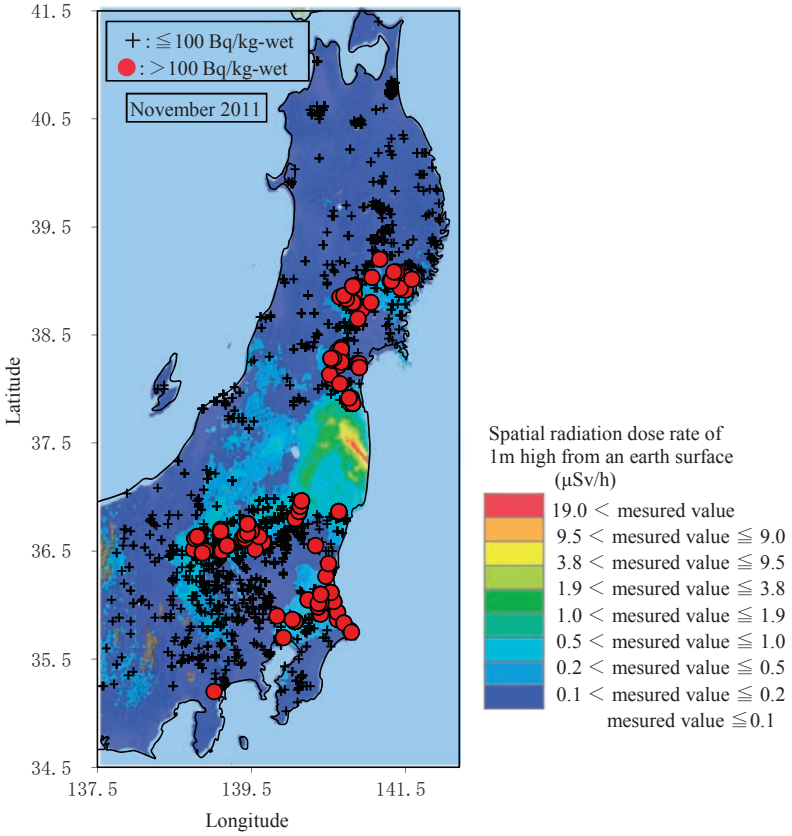


Fig.7 Distribution of radiocesium concentration in freshwater products (Apr 2012 - Mar 2016). Data from Fukushima Prefecture were excluded. Spatial radiation dose rate (Japan Map Center et al., 2016) were measured at 1m high from an earth surface at Nov 2011.

Table 4 Freshwater products with $^{134+137}$Cs concentration of >100Bq/kg-wet. Data from the Fukushima Perfecture were excluded. Measurement samples in Sep 2011 - Mar 2016 were used for data analysis.

Fish species	Prefecture								The total number of >100 Bq/kg-wet (The total number of sumples)	Maximum value (Bq/kg-wet)
	Iwate	Miyagi	Ibaraki	Tochigi	Gunma	Chiba	Saitama	Kanagawa		
Oncorhynchus masou masou	2	-	2	2	12	-	-	-	18 (1,536)	490
Salvelinus leucomaenis	7	18	2	14	18	-	-	-	59 (1,044)	460
Carassius auratus langsdorfii	-	-	5	-	-	17	-	-	22 (315)	400
Hypomesus nipponensis	-	-	-	-	19	-	-	-	19 (482)	370
Cyprinus carpio	-	-	-	-	-	10	-	-	10 (137)	330
Ictalurus punctatus	-	-	20	-	-	-	-	-	20 (46)	320
Tribolodon hakonensis	12	1	-	4	-	-	-	-	17 (970)	310
Salmo trutta	-	-	-	11	-	-	-	-	11 (12)	260
Anguilla japonica	-	1	13	-	-	6	-	-	20 (1,255)	200
Pseudorasbora parva	-	-	-	-	-	2	-	-	2 (143)	170
Plecoglossus altivelis	-	4	-	-	-	-	-	-	4 (894)	140
Oncorhynchus nerka	-	-	-	4	-	-	-	-	4 (50)	140
Silurus asotus	-	-	-	-	-	-	1	-	1 (13)	130
Oncorhynchus mykiss	-	-	-	1	-	-	-	-	1 (245)	120
Micropterus salmoides	-	-	-	-	-	-	-	1	1 (8)	110
Total	21	24	42	36	49	35	1	1	209 (7,150)	

"-" represents no samples exceeded the 100 Bq/kg-wet level.

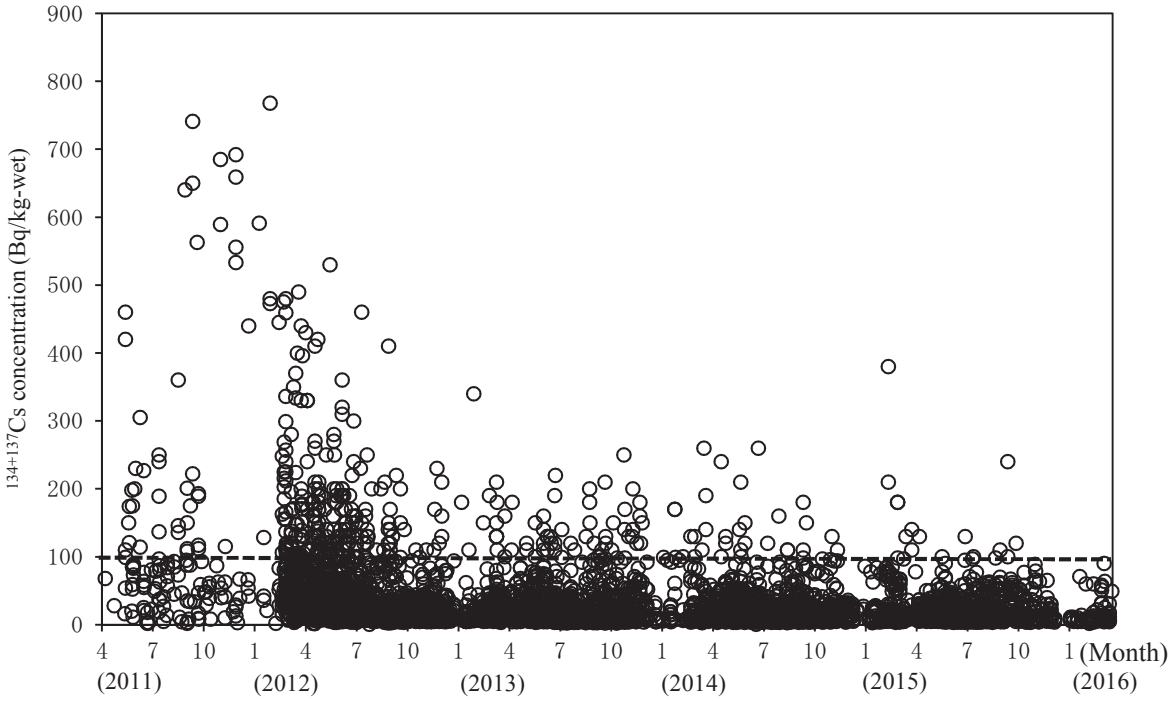


Fig.8 Temporal change of $^{134+137}$Cs concentration of freshwater products caught in eastern Japan. Data adapted from Fisheries Agency (2016).

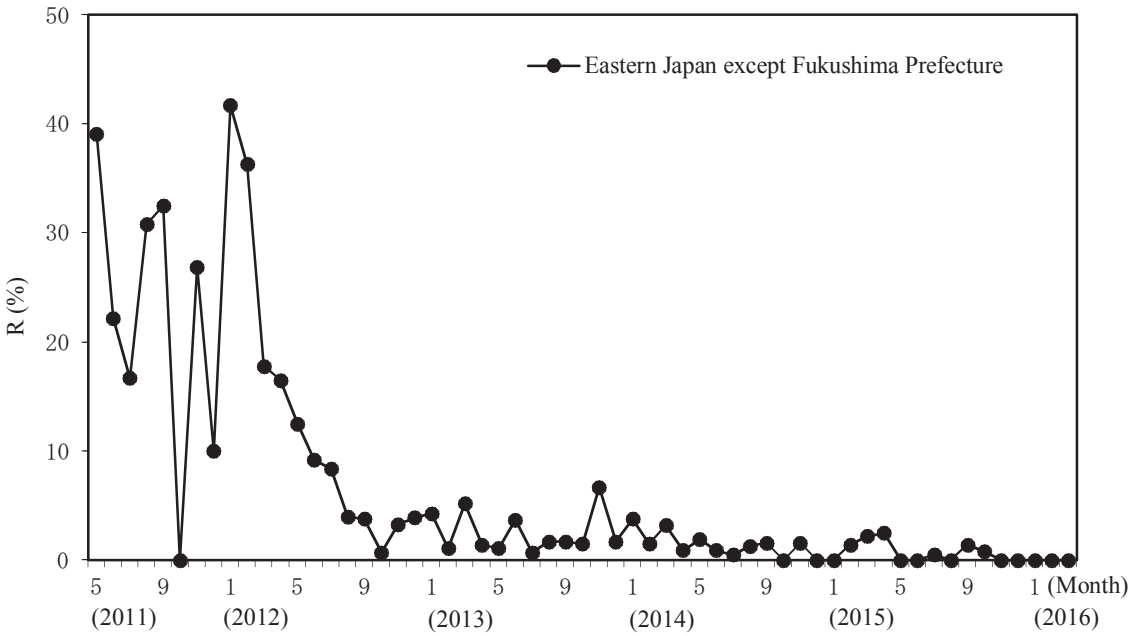


Fig.9 Temporal change of ratio (R) of samples with >100 Bq/kg-wet. The ratio were calculated based on the following equation using data adapted from Fisheries Agency (2016); R (%) = [The number of samples with concentration of >100 Bq/kg-wet] / [The number of all samples] x 100.

radioactive cesium activities exceeding 100 Bq/kg-wet were collected from Miyagi and Ibaraki prefectures, which border on Fukushima; the other 143 samples were obtained from prefectures that do not border Fukushima (Table 4). Of the 15 freshwater fish species with radioactive cesium activities exceeding 100 Bq/kg-wet, 9 species were collected from enclosed waters such as lakes and marshes; 5 species were collected from rivers; and 1 species was collected from both lake/marsh and river environments. The highest radioactive cesium activity measured during the survey was 490 Bq/kg-wet in a specimen of *Oncorhynchus masou* collected from a river.

Radioactive cesium activities detected in freshwater organisms collected in the freshwaters of Eastern Honshu (not including Fukushima) from April 2011 to March 2016 decreased over time. None of the samples has exhibited a radioactive cesium activity exceeding 100 Bq/kg-wet since October 2015, and most of the samples have fallen to 50 Bq/kg-wet or lower (Fig.8). Furthermore, the monthly percentage of samples with radioactive cesium activities exceeding 100 Bq/kg-wet (mean number of samples tested per month: 150) decreased from about 40% soon after the FDNPP accident to below 1% in October 2015 (Fig.9).

Acknowledgements

Data collected from the project commissioned by the Fisheries Agency to survey radioactive substances in fisheries products from 2011 to 2015 were used in this report. We thank the Fisheries Agency, which commissioned the project, and prefectural governments and fisheries organizations for providing the fisheries products used to analyze radioactive substances.

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