Chapter 11

Observation of Radionuclides in Marine Biota off the Coast of Fukushima Prefecture After TEPCO's Fukushima Daiichi Nuclear Power Station Accident

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Abstract Monitoring and surveying of radioactivity in seawater and biota in the marine environment off the coast of Fukushima Prefecture in the Pacific Ocean are important for understanding the dispersion of artificial radionuclides after the TEPCO Fukushima Daiichi Nuclear Power Station (FD1NPS) accident. Marine biota were collected in the coastal area off Fukushima Prefecture after this accident to investigate the radioactivity of ^{134}Cs , ^{137}Cs , and $^{110\text{m}}\text{Ag}$ in marine biota, including not only fish and shellfish but also benthos. It is well known that $^{108\text{m}}\text{Ag}$, one of the radioactive isotopes of Ag, was observed in some kinds of squid and octopus before this accident. As a result, $^{110\text{m}}\text{Ag}$ was observed in many kinds of marine biota off the coastal area of Fukushima. It is suggested that rapid change in the radioactivity in seawater, resuspension of particles from sediments, and food chain effects led to high radionuclide activities in the marine biota after this accident.

Keywords $^{110\text{m}}\text{Ag}$ • ^{134}Cs • ^{137}Cs • Marine organisms

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11.1 Introduction

Monitoring and surveying of radioactivity in seawater, sediments, and biota in the marine environment in the Pacific Ocean around Eastern Japan are important for understanding the dispersion of artificial radionuclides after the Tokyo Electric Power Company (TEPCO) Fukushima Daiichi Nuclear Power Station (FD1NPS) accident. The activities of $^{134}\text{Cs} + ^{137}\text{Cs}$ in seawater were observed to be more than 10 kBq/L around FD1NPS at the end of March 2011 and have recently decreased gradually to 1–2 mBq/L, approaching the pre-accident levels [1]. On the other hand, higher activities of $^{134}\text{Cs} + ^{137}\text{Cs}$ in sediments have been reported off the coast area of Fukushima Prefecture in the Pacific Ocean [2]. It is necessary that radioactivity in marine biota, including not only fish and shellfish but also the benthos, be monitored continuously as it is well known that marine biota accumulate and concentrate elements and radionuclides in their bodies [3]. The activities of radionuclides in marine biota off Fukushima, including plankton and benthos, are investigated to determine the variation in radioactivity. The observed artificial gamma-emitting radionuclides in marine biota include not only ^{134}Cs and ^{137}Cs but also $^{110\text{m}}\text{Ag}$. Artificial radionuclides such as ^{134}Cs , ^{137}Cs , ^{141}Ce , ^{144}Ce , ^{103}Ru , ^{106}Ru , and $^{110\text{m}}\text{Ag}$ were reported in Mediterranean seagrass after the Chernobyl accident [4]. However, ^{141}Ce , ^{144}Ce , ^{103}Ru , and ^{106}Ru were not observed in the fallout after this accident [5]. Most marine biota, aside from benthos, contained no observable $^{110\text{m}}\text{Ag}$ a year after the accident, as the activities of the short half-life radionuclide decrease with time, rather than being discharged by metabolic activity in the biota. $^{134}\text{Cs} + ^{137}\text{Cs}$ activity in marine biota was classified into three types, either tending to gradually decrease with time, or showing considerable variation, or being less than the detection limit of activity [6]. The aims of the present study were to examine the temporal and spatial changes in radioactivity in marine biota and to survey the contamination after this accident.

11.2 Materials and Methods

Marine biota samples were collected with a plankton net, dredge sampler, and trawl during cruises of T/S *Umitaka-maru*, T/S *Shinyo-maru*, and some research and fishing vessels. After being classified into species and weighed, each sample was dried with a vacuum drying machine, homogenized, and packed into a plastic container (U-8). Radioactivity was determined by gamma-ray spectrometry using a HPGe detector (GX-2019; Canberra). The radionuclide activities of biota in the sampling date were calculated with the correction of the decay and the coincidence-summing of ^{134}Cs . Detection limits of ^{134}Cs , ^{137}Cs , and $^{110\text{m}}\text{Ag}$ were estimated within 1 and 0.5 Bq/kg [wet weight (wet wt)], respectively.

11.3 Results and Discussion

Radionuclide activity in the marine biota off Onahama (Fukushima) that are used as foodstuffs, measured on June 21, 2011 and December 20, 2011, is shown in Tables 11.1 and 11.2. Radionuclide activity in marine biota (plankton and benthos, etc.) off Fukushima in the Pacific Ocean is shown in Table 11.3. The radioactivity of $^{134}\text{Cs} + ^{137}\text{Cs}$ in marine products ranged from 15 to 132 Bq/kg (wet wt) in June 2011 and from less than 1 to 135 Bq/kg (wet wt) in December 2011. $^{110\text{m}}\text{Ag}$ activity was not detected in fishes collected in December 2011, although this activity was observed in the viscera of squid, crab, and fish collected in June 2011. It is considered that the activity of $^{110\text{m}}\text{Ag}$ in fish and plankton gradually decreased with time because the half-life of $^{110\text{m}}\text{Ag}$ is 249.8 days. However, no cesium radioactivity was observed in squid and crab. It is well known that Mollusca and Crustacea concentrate silver in their visceral parts. After the accident, $^{110\text{m}}\text{Ag}$ was observed from fallout in the eastern area of Japan in April and May 2011 [2]. $^{110\text{m}}\text{Ag}$ added to seawater tends to be rapidly scavenged to the sediments because the solubility of silver in seawater is extremely low. The bottom-dwelling biota such as shellfish and benthos had high activities of Cs and Ag 1 year after the accident. Comparison of weight and radioactivity in each part of the marine organisms is shown in Fig. 11.1. The ratios of cesium activity in the various parts of marine organisms are similar to their weight ratios. The average cesium activity in seawater was 0.20 Bq/L in June 2011, which gradually decreased to 0.03 Bq/L in December 2011 around the sampling area [7]. Cesium activity in seawater decreased to a low ten times during 6 months, and then activity in marine organisms was also reduced. On the other hand, the decrease of $^{110\text{m}}\text{Ag}$ activity in mollusks could be observed to be almost that of the physical half-life time. It is thought the decrease of activity of these radionuclides in marine organisms depends on differences in the mechanisms of accumulation and metabolism.

The CR [concentration ratio = activity in biota (Bq/kg (wet wt))/activity in seawater (Bq/kg or Bq/L)] is usually represented in term of the concentration in biota relative to that of seawater for modeling purposes. This value of cesium in marine organisms was calculated with the activity of cesium in seawater (Fig. 11.2). CR of cesium (CR-Cs) in marine organisms ranged from $2.6\text{E}+1$ in the muscle part of squid to $1.0\text{E}+4$ in clam viscera. Large differences in CR-Cs in different parts of marine organisms were not observed. These values were higher than the reported CR of fish, crustaceans, mollusks, and macroalgae in TRS-422, $1.0\text{E}+2$, $5.0\text{E}+1$, $6.0\text{E}+1$, and $5.0\text{E}+1$, respectively [8]. CR in plankton was also calculated with the activity of cesium in seawater collected around the sampling area during this monitoring period. These resulting values ranged from $5.8\text{E}+1$ to $7.8\text{E}+2$, and were higher than the Cs-CR values ($2.0\text{E}+1$ – $4.0\text{E}+1$), but also similar to the Cs- K_d value in TRS-422 [8]. It was suggested that the rapid change in radioactivity in seawater and the resuspension of particles from the sediments led to high CRs of Cs after the accident.

Table 11.1 Activities of radionuclides in marine organisms used as foodstuffs off Onahama (Fukushima) on June 21, 2011

Name of biota						
Marine products	English name	Genus, species	Measured parts	^{134}Cs (Bq/kg-wet wt)	^{137}Cs (Bq/kg-wet wt)	$^{110\text{m}}\text{Ag}$ (Bq/kg-wet wt)
Fish	Japanese anchovy	<i>Engraulis japonica</i>	Whole body	10.1 ± 0.1	10.8 ± 0.1	<0.5
Fish	Pacific cod	<i>Gadus macrocephalus</i>	Whole body ^a	27.5 ± 0.3	30.2 ± 0.5	<0.5
			Muscle (edible portion)	37.9 ± 0.2	41.8 ± 0.3	<0.5
			Viscera	15.1 ± 0.1	16.3 ± 0.2	1.2 ± 0.1
			Bony parts	24.9 ± 0.2	27.2 ± 0.4	<0.5
Fish	Fat greenling	<i>Physiculus maximowiczii</i>	Whole body ^a	30.5 ± 0.4	33.4 ± 0.5	<0.5
			Muscle (edible portion)	42.3 ± 0.2	46.2 ± 0.3	<0.5
			Viscera	11.0 ± 0.1	11.8 ± 0.1	0.6 ± 0.03
			Bony parts	35.3 ± 0.3	39.0 ± 0.4	<0.5
Fish	Pointhead flounder	<i>Hippoglossoides dubius</i>	Whole body ^a	63.3 ± 0.9	68.8 ± 1.3	<0.5
			Muscle (edible portion)	137.0 ± 0.9	148.9 ± 1.2	<0.5
			Viscera	22.5 ± 0.2	24.3 ± 0.3	<0.5
			Bony parts	18.4 ± 0.1	20.2 ± 0.2	<0.5
Fish	Rikuzen sole	<i>Dexistes rikuzenius</i>	Whole body ^a	13.2 ± 0.2	14.1 ± 0.3	0.8 ± 0.1
			Muscle (edible portion)	13.5 ± 0.1	14.7 ± 0.2	<0.5
			Viscera	17.6 ± 0.2	18.8 ± 0.2	5.3 ± 0.1
			Bony parts	11.6 ± 0.1	12.2 ± 0.2	<0.5
Squid	Bobtail squid	Sepioida	Whole body ^a	5.4 ± 0.1	5.7 ± 0.1	37.6 ± 0.2
			Muscle (edible portion)	2.5 ± 0.1	2.7 ± 0.1	2.9 ± 0.1
			Viscera	8.0 ± 0.1	8.6 ± 0.1	70.7 ± 0.2
Squid	Japanese common squid	<i>Todarodes pacificus</i>	Whole body ^a	10.6 ± 0.1	11.5 ± 0.2	24.9 ± 0.3
			Muscle (edible portion)	10.6 ± 0.1	11.5 ± 0.1	1.5 ± 0.03
			Viscera	10.6 ± 0.1	11.3 ± 0.1	92.8 ± 0.3
Crab	Snow crab	<i>Chionoecetes opilio</i>	Soft tissue ^b	7.2 ± 0.1	7.8 ± 0.2	3.4 ± 0.1

^aWhole-body activity was weighted as average with that in all parts^bActivity was calculated with those in muscle and viscera parts because that in the shell part of crab was not determined

Table 11.2 Radionuclide activity in marine organisms used as foodstuffs off Onahama (Fukushima) on December 20, 2011

Name of biota						^{110m} Ag
Marine products	English name	Genus, species	Measured parts	¹³⁴ Cs (Bq/kg-wet wt)	¹³⁷ Cs (Bq/kg-wet wt)	(Bq/kg-wet wt)
Algae	Arame	<i>Eisenia bicyclis</i>	Whole body	16.4±0.3	20.9±0.4	1.9±0.1
Fish	Greeneyes	<i>Chlorophthalmus albatrossis</i>	Whole body ^a	11.9±0.3	15.1±0.4	<0.5
			Muscle (edible portion)	12.1±0.3	15.2±0.5	<0.5
			Viscera	14.0±0.4	18.2±0.7	<0.5
			Bony parts	10.9±0.2	14.5±0.3	<0.5
Fish	Slime flounder	<i>Microstomus achne</i>	Whole body ^a	1.7±0.1	2.3±0.2	<0.5
			Muscle (edible portion)	1.5±0.1	2.1±0.2	<0.5
			Viscera	2.9±0.1	3.8±0.2	<0.5
			Bony parts	1.4±0.1	1.7±0.1	<0.5
Squid	Japanese squid	<i>Loliolus (Nipponololigo) japonica</i>	Whole body ^a	<1	<1	4.2±0.1
			Muscle (edible portion)	<1	<1	4.6±0.1
			Viscera	<1	<1	43.3±0.7
			Eyeball	<1	<1	6.9±0.1
			Cartilage	<1	<1	15.1±0.9
Squid	Spear squid	<i>Loligo bleekeri</i>	Whole body ^a	<1	<1	2.4±0.2
			Muscle (edible portion)	<1	<1	<0.5
			Viscera	<1	<1	12.4±0.4
			Eyeball	<1	<1	1.7±0.1
			Cartilage	<1	1.1±0.1	<0.5
Crab	Snow crab	<i>Chionoecetes opilio</i>	Whole body ^b	<1	<1	3.1±0.1
			Muscle (edible portion)	<1	<1	2.2±0.1
			Viscera	<1	<1	11.1±0.2
Shellfish	Sakhalin surf clam	<i>Pseudocardium sachalinense</i>	Whole body ^c	58.9±1.1	76.3±1.8	19.0±0.7
			Muscle (edible portion)	16.8±0.3	20.5±0.5	4.8±0.2
			Mantle	108.4±1.6	142.1±2.6	29.7±1.0
			Viscera	23.7±0.5	29.5±0.8	31.7±0.6

^aWhole-body activity was weighted as average with that in all parts^bWhole-body activity was calculated with those in muscle and viscera parts, as that in the shell part of crab was not determined^cWhole-body activity was calculated with those in muscle, mantle, and visceral parts, as that in the shell parts of shellfish was not determined

Table 11.3 Radionuclide activity in marine biota (plankton and benthos) off Fukushima in the Pacific Ocean

Date	Location		Depth (m)	Name of biota		English name	Genus, species	Measured part	¹³⁴ Cs (Bq/ kg-wet wt)	¹³⁷ Cs (Bq/ kg-wet wt)	^{110m} Ag (Bq/ kg-wet wt)
	Latitude	Longitude		Marine products							
2011/7/6	36 55	2 N 141 0	0 E	Plankton				Whole	2.7±0.2	2.9±0.2	1.6±0.1
2011/7/6	36 55	2 N 141 25	92 E	Plankton				Whole	2.2±0.9	2.4±1.2	0.9±0.1
2011/7/14	37 5	0 N 140 59	10 E	Crustacea	Mysidacea			Whole	24.1±0.4	26.4±0.6	15.5±0.3
2011/8/17	37 4	58 N 140 59	19 E	Crustacea	Mysidacea			Whole	41.3±0.5	46.7±0.7	7.0±0.2
2011/9/5	37 5	8 N 140 59	56 E	Crustacea	Mysidacea			Whole	31.0±0.5	34.8±0.7	15.0±0.3
2011/11/1	37 4	30 N 141 9	18 E	Plankton (mesh size of net, 330 µm)				Whole	32.2±0.8	37.1±1.1	<1
2012/4/25	37 50	0 N 141 6	0 E	Plankton (mesh size of net, 330 µm)				Whole	22.2±1.1	31.5±1.3	<1
2011/10/22	36 55	2 N 141 0	0 E	Polychaeta	Polychaetes			Whole	146.8±3.8	181.5±5.7	11.6±1.5
2011/10/22	36 55	2 N 141 0	0 E	Sea urchin		<i>Echinocardium cordatum</i>		Whole	271.0±5.6	311.4±8.5	<1
2011/10/22	36 55	2 N 141 0	0 E	Starfish		<i>Distolasterias nipon</i>		Whole	5.6±0.3	7.0±0.4	8.3±0.3
2011/10/22	36 55	2 N 141 0	0 E	Starfish	Northern Pacific seastar	<i>Asterias amurensis</i>		Whole	3.4±0.3	4.7±0.4	16.0±0.4
2011/10/22	36 55	2 N 141 0	0 E	Sea slug	Opisthobranchia Spengel	<i>Philine argentata</i>		Whole	17.2±0.4	20.6±0.5	21.8±0.4

Fig. 11.1 Comparison of weight and radioactivity in each part of the marine organisms. *Black bars*, whole body; *gray bars*, muscle (edible parts); *open bars*, viscera; *light gray bars*, bony parts. Samples were collected in June (a) and December (b), 2011

11.4 Conclusion

The activities of ^{134}Cs , ^{137}Cs , and $^{110\text{m}}\text{Ag}$ in marine biota off the coast of Fukushima Prefecture in the Pacific Ocean were investigated a year after the FD1NPS accident. $^{110\text{m}}\text{Ag}$ could be observed in many marine biota after this accident, although it is

Fig. 11.2 Concentration ratio of cesium ($CR\text{-}Cs$) in each part of the marine organisms. *Black squares*, whole body; *gray squares*, muscle (edible parts); *open squares*, viscera; *open circles*, bony parts

well known that Mollusca and Crustacea concentrate silver in the visceral parts. Finally, it was suggested that the CR fluctuations in plankton is a result of both radioactivity in seawater and sediment resuspension.

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