Observation of radionuclides in marine biota off the coast of Fukushima prefecture after TEPCO's Fukushima Daiichi Nuclear Power Plant accident

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Monitoring and surveying radioactivity in seawater and biota in the marine environment off the coast of Fukushima prefecture in the Pacific are important for understanding the dispersion of artificial radionuclides after TEPCO's Fukushima Daiichi Nuclear Power Plant (F1NPP) accident. Marine biota were collected in the coastal area of Fukushima prefecture after this accident due to investigate the activities of ^{134}Cs, ^{137}Cs, and ^{110m}Ag in marine biota, including not only fish and shellfish but also benthos. It is well known that ^{108m}Ag, one of the radioactive isotopes of Ag, was observed in some kinds of squid and octopus before this accident. As the results, ^{110m}Ag was observed in many kinds of marine biota off the coastal area of Fukushima. It is suggested that rapid change in the radioactivities in seawater, resuspension of particles from sediments and food chain effects led to high radionuclide activities in marine biota after this accident.

Key Words: *Marine organisms, ^{134}Cs, ^{137}Cs, ^{110m}Ag*

1. Introduction

Monitoring and surveying of radioactivity in seawater, sediments and biota in the marine environment around the eastern Japan in the Pacific are important for understanding the dispersion of artificial radionuclides after Tokyo Electric Power Company (TEPCO)'s Fukushima Daiichi Nuclear Power Plant (F1NPP) accident. The activities of ^{134}Cs + ^{137}Cs in seawater were observed to be over 10 KBq/L around F1NPP at the end of March 2011 and have recently decreased gradually to 1–2 mBq/L, approaching the preaccident levels[1)]. On the other hand, higher activities of ^{134}Cs + ^{137}Cs in sediments have been reported off the coast area of Fukushima prefecture in the Pacific off Fukushima[2)]. It is necessery that radioactivity in marine biota, including not only fish and shellfish but also benthos be monitored continuously; it is well known that marine biota accumulates and concentrates elements and radionuclides in their body[3)]. The activities of radionuclides in marine biota, including plankton and benthos etc., off Fukushima are investigated to determine the variation in radioactivity. The observed artificial gamma emitter radionuclides include not only ^{134}Cs and ^{137}Cs but also ^{110m}Ag in marine biota. It was reported that the artificial radionuclides such as ^{134}Cs, ^{137}Cs, ^{141}Ce, ^{144}Ce, ^{103}Ru, ^{106}Ru and ^{110m}Ag in Mediterranean seagrass after Chernobyl accident[4]. ^{141}Ce, ^{144}Ce, ^{103}Ru, and ^{106}Ru were not observed in fallout after this accident[5]. Most marine biota, aside from benthos, contained no observable ^{110m}Ag a year after the accident, as the activities of the short-half life time radionuclide decrese with time ,rather than the discharge with the metabolism in biota . The ^{134}Cs + ^{137}Cs activity in marine biota was classified into three types, either showing a tendency of gradually decrease with time, considerable variation or less than detection limit of the 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.

2. Materials and Methods

Marine biota samples were collected with a plankton net, dredge sampler, and trawl during the cruise of T/S Umitaka, T/S Shinyo, 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. The radioactivity was determined by gamma ray spectrometry using a HPGe detector (Canberra, GX-2019). The activites of radionuclides 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 ^{110m}Ag were estimated within 1 and 0.5 Bq/kg (wet wt), respectively.

3. Results and Discussion

The activities of radionuclides in marine biota as foodstuff off Onahama (Fukushima) on June 21, 2011 and December 20, 2011 are shown in Table 1 and 2, while the activities of radionuclides in marine biota (plankton and benthos, etc.) off Fukushima in the Pacific are shown in Table 3. The radioactivity of ^{134}Cs + ^{137}Cs in marine products was 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. ^{110m}Ag activity was not detected in fishes collected in December 2011, although the activity was observed in the viscera part of squid, crab, and fish collected in June 2011. It is considered that the activities of ^{110m}Ag in fish and plankton gradually decreased with time, because the half life of ^{110m}Ag is 249.8 days. On the other hand, no Cs radioactivity was observed in squid and crab. It is well known that mollusca and crustacea

Table 1 The activities of radionuclides in manine organisms as foodstuff around off Onahama(Fukushima) in June 21, 2011.

Name of biota Marine products	English name	*species genus*	Measurement parts	Cs-134 (Bq/kg-wet weight)	Cs-137 (Bq/kg-wet weight)	Ag-110m (Bq/kg-wet weight)
Fish	Japanese Anchovy	*Engraulis japonica*	Whole body	10.1 ± 0.1	10.8 ± 0.1	< 0.5
Fish	Pacific cod	*Gadus macrocephalus*	Whole body[1)]	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	*Physiculus maximowiczi*	Whole body[1)]	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	*Hippoglossoides dubius*	Whole body[1)]	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	*Dexistes rikuzenius*	Whole body[1)]	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[1)]	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	*Todarodes pacificus*	Whole body[1)]	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	*Chionoecetes opilio*	Soft tissue[2)]	7.2 ± 0.1	7.8 ± 0.2	3.4 ± 0.1

[1)] The activity in whole body was weighted average with that in all parts.
[2)] The activity was calculated with those in muscle and viscera parts, as that in a shell part of crab was not determined.

Table 2 The activities of radionuclides in manine organisms as foodstuff around off Onahama(Fukushima) in December 20, 2011.

Name of biota Marine products	English name	*species genus*	Measurement parts	Cs-134 (Bq/kg-wet weight)	Cs-137 (Bq/kg-wet weight)	Ag-110m (Bq/kg-wet weight)
Algae	Arame	*Eisenia bicyclis*	Whole body	16.4 ± 0.3	20.9 ± 0.4	1.9 ± 0.1
Fish	Greeneyes	*Chlorophthalmus albatrossis*	Whole body[1)]	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	*Microstomus achne*	Whole body[1)]	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	*Loliolus (Nipponololig) japonica*	Whole body[1)]	< 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	*Loligo bleekeri*	Whole body[1)]	< 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	*Chionoecetes opilio*	Whole body[2)]	< 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	*Pseudocardium sachalinense*	Whole body[3)]	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

1) The activity in whole body was weighted average with that in all parts.
2) The activity in whole body was calculated with those in muscle and viscera parts, as that in a shell part of crab was not determined.
3) The activity in whole body was calculated with those in muscle, mantle and viscera parts, as that in a shell part of shellfish was not determined.

concentrate silver in their visceral parts. After the accident, ^{110m}Ag was observed from fallout in the eastern area of Japan in April and May 2011[2]. ^{110m}Ag added to seawater tends to be rapidly scavenged to sediments, as the solubility of silver is extremely low in seawater. The bottom-dwelling biota such as shellfish and benthos had high activities of Cs and Ag a year after the accident.

The CR (Concentration ratio = Activity in biota (Bq/kg (wet wt))/Activity in seawater (Bq/kg or Bq/L)) of Cs in plankton was calculated with the activities ceasium in seawater, which were collected around sampling area during this monitoring period. These resulting values ranged from 5.8E+1 to 7.8E+2, were higher than the Cs-CR values (2.0E+1~4.0E+1), but also similar with the Cs-Kd value (Sediment distribution coefficients) reported by International Atomic Energy Agency (IAEA)-Technical Reports Series No.422[7]. 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.

4. Conclusion

The activities of ^{134}Cs, ^{137}Cs, and ^{110m}Ag in marine biota off the coast of Fukushima prefecture in the Pacific were investigated a year after the F1NPP accident. ^{110m}Ag could be observed in many marine biota after this accident, although it is well-known that mollusca and crustacea concentrate silver in visceral parts. Finally, it was suggested that the CR fluctuations in plankton is a result of both radioactivities in seawater as well as sediment resuspension.

Acknowledgements

Sampling would not have been possible without the field support obtained from the T/S Umitaka, T/S Shinyo,

Table 3 The activities of radionuclides in marine biota(plankton and benthos) around off Fukushima in the Pacific.

Date	Location Latitude				Longitute				Depth m	Name of biota Marine products	English name	*species genus*	Measurement part	Cs-134 (Bq/kg-wet weight)	Cs-137 (Bq/kg-wet weight)	Ag-110m (Bq/kg-wet weight)
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	7	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	7	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	10	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	26	Plankton (mesh size of net: 330mm)			whole	32.2 ± 0.8	37.1 ± 1.1	< 1
2012.4.25	37	50	0	N	141	6	0	E	28	Plankton (mesh size of net: 330mm)			whole	22.2 ± 1.1	31.5 ± 1.3	< 1
2011.10.22	36	55	2	N	141	0	0	E	40	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	40	Sea Urchin		*Echinocardium cordatum*	whole	271.0 ± 5.6	311.4 ± 8.5	< 1
2011.10.22	36	55	2	N	141	0	0	E	40	Starfish		*Distolasterias nipon*	whole	5.6 ± 0.3	7.0 ± 0.4	8.3 ± 0.3
2011.10.22	36	55	2	N	141	0	0	E	40	Starfish	Northern Pacific seastar	*Asterias amurensis*	whole	3.4 ± 0.3	4.7 ± 0.4	16.0 ± 0.4
2011.10.22	36	55	2	N	141	0	0	E	40	Sea slug	Opisthobranchia Spengel	*Philine argentata*	whole	17.2 ± 0.4	20.6 ± 0.5	21.8 ± 0.4

and some research and fishing vessels. A part of sample treatment supports were provided by staff members of National Research Institute of Fisheries Science, Fisheries Research Agency. This work was partly supported by Health Labour Sciences Research Grant and MEXT Grant-in-Aid for Scientific Research on Innovative Areas (24110004).

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