**$^{239,240}\text{Pu}$ and ^{137}Cs Concentrations in Some Marine Organisms,
Mostly from the Ibaraki and Aomori Coasts,
Japan, 1987-1989**

Yutaka Nagaya,* Yuzuru Suzuki,* and Kiyoshi Nakamura*

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Several marine biological samples were collected from the Pacific coasts of Ibaraki and Aomori Prefecture in 1987 to 1989, and $^{239,240}\text{Pu}$ and ^{137}Cs concentrations were measured. A comparison was also made between some pelagic and benthic fishes for their radionuclide concentration.

$^{239,240}\text{Pu}$ contents of coastal fishes were higher in viscera, and muscle usually showed the lowest value. On the contrary, ^{137}Cs contents were rather higher in muscle.

The differences of radionuclide concentrations were found among species, but no definite regional variation was observed between the Ibaraki and Aomori coasts.

The $^{239,240}\text{Pu}$ concentration level in coastal fishes was higher than that in pelagic ones, and no significant difference was observed between pelagic fishes from warm water masses and cold water masses. On the contrary, ^{137}Cs contents of the pelagic fishes showed clear difference between warm water species and cold water ones.

The surface and benthic fishes seemed to show similar concentration levels of radionuclides.

The concentrations of artificial radionuclides, $^{239,240}\text{Pu}$ and ^{137}Cs in some marine organisms from the seas around Japan, in 1981-1984, had been reported in our previous report,¹⁾ as a part of radioactive pollution study in the marine environment.

The results indicated that the ^{137}Cs concentration was at an approximately similar level, and it seemed that no significant difference was observed among regions but that slight a discrepancy was found between species. On the contrary, $^{239,240}\text{Pu}$ showed considerable variation.

In an effort to clarify the discrepancies in the radionuclide contents among regions, species or tissues of Japanese marine organisms, several biological samples were collected from two coastal areas and $^{239,240}\text{Pu}$ and ^{137}Cs concentrations were measured and reported in this article.

Materials and Methods

Most samples were collected from the Pacific coastal seas of Ibaraki and Aomori Pref., Japan during 1987 to 1989 (Fig. 1). The areas were similar in their geographical and geological features, having monotonous north-south sandy be-

aches dotted with a few rocky shores and rather shallow water depth (less than 50 m at 10 km off shore) with sandy bottom. As for water masses, Aomori coast has both cold water (Oyashio) and warm water (Tsuruga current, a branch of Tsushima Current) from sea of Japan, whereas Ibaraki is also influenced by warm water (Kuroshio) and the cold water (Oyashio).²⁾

The radionuclides of some pelagic and mesopelagic-benthic fishes were also measured to make comparisons with the coastal species.

Analytical and measuring procedures are the same as described in our previous papers.^{1,3-6)}

Results

The analytical results of the coastal samples are shown in Tables 1 and 2, and the data of pelagic and mesopelagic fishes are shown in Tables 3 and 4. The precision criterion in the tables is derived from counting statistics. The mean values for coastal fishes are indicated in Table 5, and Table 6 shows mean values for other organisms.

General levels of the radionuclide concentrations in coastal samples are similar to those observed in 1981 to 1984,¹⁾ showing 10^1 to 10^2 mBq/kg-raw

* Division of Marine Radioecology, National Institute of Radiological Sciences, Isozaki, Nakaminato 311-12, Japan (長屋 裕, 鈴木 謙, 中村 清: 放射線医学総合研究所, 海洋放射生態学研究部).

Fig. 1. Sampling Locations and Currents.

for ^{137}Cs and 10^{-1} to 10^1 mBq/kg-raw for $^{239,240}\text{Pu}$.

As for $^{239,240}\text{Pu}$ in coastal fishes, viscera (including digestive tract) showed the highest value, followed by gill, skin and spine bone, and the concentration in muscle is usually the lowest of all organs and tissues. On the contrary, the ^{137}Cs concentration in muscle seems to be higher than those in other organs and tissues.

Among the edible or soft parts of coastal organisms, viscera and algae showed higher $^{239,240}\text{Pu}$ concentration, whereas the ^{137}Cs content appeared to be a rather similar level and no definite difference was observed.

$^{239,240}\text{Pu}$ concentrations in muscle and viscera of Ibaraki coastal fishes seems to be higher than those of Aomori coast, but the ^{137}Cs content in muscle show a reverse tendency. This inconsistent discrepancy in radionuclide contents of fishes between the regions implies that the differences of the radioisotope contents were derived from the different affinities of species to the radionuclides, and not from the differences in environmental radionuclide concentrations. Moreover, because the standard deviations for mean concentrations were relatively large (Table 5), no significant regional difference could be identified.

Table 1. Radionuclide Concentrations in Ibaraki Organisms (1988–1989)

Biota	Name	Organ & Tissue	Ash (%)	$^{239,240}\text{Pu}$ (mBq/kg-raw)	^{137}Cs
Pisces	Konoshiro	muscle	2.6	2.8 ± 0.3	131 ± 7
	<i>Konosirus punctatus</i>	viscera	2.3	21.4 ± 1.7	64 ± 4
	Maanago	muscle	1.6	0.08 ± 0.08	42 ± 3
	<i>Conger myriaster</i>				
	Makogarei	muscle	1.8	0.05 ± 0.07	72 ± 4
	<i>Limanda yokohamae</i>	viscera	2.4	4.8 ± 0.4	128 ± 7
	Mebaru	muscle	2.5	0.2 ± 0.2	108 ± 6
	<i>Sebastes inermis</i>	viscera	2.3	3.0 ± 1.0	187 ± 14
Bivalve	Chosen hamaguri	soft part	1.9	1.2 ± 0.2	133 ± 7
	<i>Meretrix lamarckii</i>				
	Kotamagai	soft part	1.9	1.3 ± 0.2	134 ± 7
	<i>Gomphina melanaegis</i>				
	Saragai	soft part	2.7	3.3 ± 0.5	246 ± 16
Cephalopod	<i>Peronidia venulosa</i>				
	Madako	muscle	2.3	0.8 ± 0.2	54 ± 4
	<i>Octopus vulgaris</i>				
	Iidako	whole body	2.4	2.4 ± 0.3	46 ± 4
Algae	<i>Octopus ocellatus</i>				
	Hiziki*	whole body	5.1	4.0 ± 0.5	181 ± 8
	<i>Hizikia fusiformis</i>				
	Arame*	whole body	4.5	2.8 ± 0.3	146 ± 5
	<i>Eisenia bicyclis</i>				
	Umitoranoo*	whole body	5.0	27.2 ± 1.3	234 ± 11
	<i>Sargassum thunbergii</i>				
	Tsunomata	whole body	5.6	4.5 ± 0.6	233 ± 15
	<i>Chondrus ocellatus</i>				
	Harigane	whole body	8.9	19.2 ± 2.1	214 ± 14
	<i>Ahnfeltia paradoxa</i>				
	Fukurofunori	whole body	4.9	5.1 ± 0.6	192 ± 13
	<i>Gloiopeltis furcata</i>				

* Mean value of two samples.

In comparison with pelagic fishes, the $^{239,240}\text{Pu}$ contents of the coastal ones tend to be higher in muscle and viscera, and similar in bone, skin and gill. On the other hand, the ^{137}Cs concentrations in muscle of the coastal fishes are either lower or similar to those of pelagic fishes, and for skin and gill either higher than or similar to the pelagic ones. These controversial ^{137}Cs discrepancies were caused by significant differences in the radionuclide concentrations between cold-water inhabitants (*Oncorhynchus keta*) and warm-water inhabitants (*Katsuwonus pelamis* and *Parathunnus sibi*), as shown in Table 3.

Other pelagic cold-water fishes, *Theragra chalcogramma* and *Gadus macrocephalus*, also showed lower ^{137}Cs contents than pelagic warm-water fishes, *Katsuwonus pelamis*.¹⁾ Since these pelagic cold-water fishes were known as benthic species, comparison with some warm-water benthic fishes would be desirable, so several mesopelagic benthic

fishes were investigated, as shown in Table 4. The results show similar concentration levels of both $^{239,240}\text{Pu}$ and ^{137}Cs between warm-water benthic and surface fishes. On the other hand, the cold water benthic fishes seem to show rather lower ^{137}Cs contents than warm-water benthic ones.

Discussion

Concentration of radionuclides in marine organisms is controlled by both the biological affinity and the physicochemical state of the isotopes in sea water.

Most of ^{137}Cs is supposed to be in a dissolved state in sea water and transported with water movement,^{4,5)} whereas considerable portions of plutonium isotopes are reported to be in particulate form.^{7,8)} Therefore, the general tendencies of the radionuclide distribution in fishes, that is, higher $^{239,240}\text{Pu}$ contents in viscera, gill or skin and higher

Table 2. Radionuclide Concentrations in Aomori Organisms (1987-1989)

Biota	Name	Organ & Tissue	Ash (%)	$^{239,240}\text{Pu}$ (mBq/kg-raw)	^{137}Cs
Pisces	Hirame (adult)	skin	2.4	0.4 $\pm$ 0.3	148 $\pm$ 11
		<i>Pralichthys olivaceus</i> muscle	1.4	0.1 $\pm$ 0.3	300 $\pm$ 11
		viscera	1.2	0.1 $\pm$ 0.2	155 $\pm$ 15
		spine	13.8	0.6 $\pm$ 0.3	—
		gill	4.5	0.6 $\pm$ 1.0	44 $\pm$ 30
	Hirame (juvenile)	skin	2.4	0.2 $\pm$ 0.2	74 $\pm$ 15
		muscle	1.5	0.19 $\pm$ 0.07	266 $\pm$ 11
		viscera	1.4	1.0 $\pm$ 1.3	159 $\pm$ 26
		spine	9.8	0.9 $\pm$ 0.1	—
		gill	5.0	0.4 $\pm$ 0.6	96 $\pm$ 89
	Ainame (adult)	skin	2.6	0.0 $\pm$ 0.1	218 $\pm$ 22
		<i>Hexagrammos otakii</i> muscle	1.4	0.0 $\pm$ 0.1	522 $\pm$ 18
		viscera	2.1	4.6 $\pm$ 0.4	92 $\pm$ 7
		spine	11.1	2.5 $\pm$ 4.1	—
		gill	4.2	0.3 $\pm$ 0.3	221 $\pm$ 48
	Ainame (juvenile)	muscle	1.6	0.19 $\pm$ 0.07	166 $\pm$ 11
		viscera	2.7	5.0 $\pm$ 0.6	33 $\pm$ 30
		spine	8.5	0.2 $\pm$ 0.3	—
Echinoderm	Kitamurasakiuni	ovary	2.2	4.2 $\pm$ 0.8	129 $\pm$ 15
		<i>Strongylocentrotus nudus</i> shell	61.4	18.1 $\pm$ 9.3	—
Bivalve	Hotategai	muscle	2.4	1.5 $\pm$ 0.3	67 $\pm$ 27
	<i>Patinopecten yessoensis</i>	viscera	2.7	5.1 $\pm$ 0.4	11 $\pm$ 11
	Ubagai	soft part	4.1	5.7 $\pm$ 0.3	42 $\pm$ 13
Algae	Makonbu	whole body	5.4	10.8 $\pm$ 1.2	155 $\pm$ 33
	<i>Laminaria japonica</i>				

Table 3. Radionuclide Concentrations in Pelagic Fishes (1989)

Name and Location		Organ & Tissue	Ash (%)	$^{239,240}\text{Pu}$ (mBq/kg-raw)	^{137}Cs
Sake	<i>Oncorhynchus keta</i> (off Sanriku)	skin	1.5	0.11±0.05	56±11
		muscle	1.3	0.05±0.06	133±12
		viscera*1	1.5	0.7 ±0.2	67± 9
		spine	7.0	0.2 ±0.2	—
		gill	1.1	0.03±0.03	17± 4
Katsuo	<i>Katsuwonus pelamis</i> (off Sanriku)	skin	4.3	0.5 ±0.1	116±24
		muscle	1.3	0.17±0.09	438±16
		viscera	1.8	0.6 ±0.3	370±29
		d.t.*2	2.7	1.9 ±0.6	472±24
		spine	11.3	0.3 ±0.3	—
		gill	7.7	0.6 ±0.3	175±32
Mebachi	<i>Parathunnus sibi</i> (Kumano-Nada)	skin	4.3	0.1 ±0.1	226±31
		muscle	1.0	0.06±0.06	458±14
		viscera	2.3	0.2 ±0.1	488±56
		d.t.*2	1.6	0.2 ±0.1	471±34
		spine	12.4	0.9 ±0.9	—
		gill	6.9	0.3 ±0.3	239±32

*¹ Including digestive tract.*² Digestive tract.

Table 4. Mean Radionuclide Concentrations in Mesopelagic-Benthic Fishes from Izu Region in 1979-1986 (mBq/kg-raw)

Name	Organ & Tissue	Nos.	$^{239,240}\text{Pu}$			^{137}Cs		
			Mean Value	Standard Deviation	Counting Error	Mean Value	Standard Deviation	Counting Error
Kinmedai	Muscle	2	0.04	± 0.03	± 0.04	391	± 49	± 18
<i>Beryx splendens</i>	Spine	2	0.63	± 0.89	± 1.25	—	—	—
Medai	Muscle	3	0.02	± 0.02	± 0.03	218	± 55	± 11
<i>Hyperogyphe japonica</i>	Spine	3	1.37	± 1.02	± 0.33	—	—	—
Kuromutsu	Muscle	1	0.03	—	± 0.09	466	—	± 30
<i>Scombrops gilberti</i>	Spine	1	0.78	—	± 0.58	—	—	—
Akoudai	Muscle	1	0.17	—	± 0.11	331	—	± 41
<i>Sebastes matsubarae</i>	Spine	3	0.67	± 0.35	± 0.63	—	—	—
Aodai	Muscle	1	0.04	—	± 0.10	282	—	± 15
<i>Paracaesio caeruleus</i>	Spine	1	0.22	—	± 0.54	—	—	—
Total Mean	Muscle	8	0.05	± 0.05	± 0.03	314	± 102	± 9
	Spine	10	0.84	± 0.71	± 0.34	—	—	—

Table 5. Mean Concentration of Radionuclides in Coastal Fishes (mBq/kg-raw)

Organ & Tissue	Region	Nos.	$^{239,240}\text{Pu}$			Nos.	^{137}Cs		
			Mean Value	Standard Deviation	Counting Error		Mean Value	Standard Deviation	Counting Error
Muscle	Aomori	4	0.13	± 0.09	± 0.08	4	314	± 150	± 7
	Ibaraki	4	0.77	± 1.34	± 0.09	4	88	± 39	± 3
	Total	8	0.45	± 0.95	± 0.06	8	201	± 158	± 4
Viscera*	Aomori	4	2.68	± 2.49	± 0.38	4	110	± 60	± 11
	Ibaraki	3	9.70	± 10.16	± 0.67	3	126	± 62	± 5
	Total	7	5.70	± 7.18	± 0.36	7	117	± 56	± 7
Spine	Aomori	4	1.05	± 1.00	± 1.03	—	—	—	—
Skin	Aomori	3	0.20	± 0.20	± 0.12	3	147	± 72	± 10
Gill	Aomori	3	0.43	± 0.15	± 0.40	3	120	± 91	± 35

* Including digestive tract.

Table 6. Mean Concentration of Radionuclides in Mollusca and Algae (mBq/kg-raw)

Biota	Organ & Tissue	Nos.	$^{239,240}\text{Pu}$			^{137}Cs		
			Mean Value	Standard Deviation	Counting Error	Mean Value	Standard Deviation	Counting Error
Bivalve	soft part	6	3.0	± 2.0	± 0.1	106	± 85	± 5
Cephalopod	soft part	2	1.6	± 1.1	± 0.2	50	± 6	± 3
Algae	whole body	7	10.5	± 9.3	± 0.4	194	± 35	± 6

^{137}Cs contents in muscle, were probably caused by the difference in the physicochemical form of the radionuclides.

The fact that no significant regional difference in the radionuclide contents was observed between Aomori and Ibaraki biota suggests a similar radioactive pollution level along the Pacific coastal sea of northern Japan.

The lower ^{137}Cs concentrations of pelagic cold-water inhabitants than those of warm-water ones seem to be due to the lower contents of the radionuclide in northern Northwest Pacific water in comparison with the southern water,⁵⁾ but we have no explanation for the apparently similar levels of $^{239,240}\text{Pu}$ contents between warm- and cold-water species, despite the difference of the

radioisotope contents in sea water,* except the relatively higher counting errors due to the very low plutonium contents in the biological samples, which have made precise comparison of data rather difficult.

Considerable portions of artificial radionuclides entered into a coastal marine environment are known to be retained in sediment,^{8,10)} then the sediments are "enriched" with flow-in from land,^{8,11)} and could cause a delayed contamination of coastal biota.¹²⁾ The differences of the radionuclide contents between coastal and pelagic fishes could be, at least partly, attributed to either this enrichment or the regional discrepancy of the radionuclide contents in water masses,⁵⁾ or both. The enrichment is supposed to have made ^{239,240}Pu contents in the coastal fishes higher than those in the pelagic ones, but not sufficient for ¹³⁷Cs to make the contents in the coastal fishes higher than those of the pelagic fishes from southern waters.

As mentioned earlier, considerable portions of plutonium in sea water are in particulate form^{7,8)} and significant amounts of particulate plutonium sink rather rapidly.¹³⁾ Therefore, the concentration level of ^{239,240}Pu in deep water is rather similar to or often exceeds that in surface layer.⁵⁾ Besides, plutonium isotopes in marine organisms are supplied mainly from sea water,^{14,15)} so the similar levels of ^{239,240}Pu contents in the benthic fishes and the surface fishes might be attributed to the particle formation and the rapid sinking of plutonium in sea water, as suggested previously.¹⁾

Unlike plutonium, most of the ¹³⁷Cs is in dissolved form in sea water and transported with its movement, and the concentration in deep water is usually 1/10 to 1/100 of that in surface water.^{4,5)} Despite the remarkable discrepancy in the concentrations in sea water, the ¹³⁷Cs contents in surface fishes and benthic ones are at similar levels. The marine fishes are known to accumulate ¹³⁷Cs through both environmental sea water and their foods,¹⁶⁾ so the higher the ¹³⁷Cs concentration in sea water for food organisms, the larger the contribution of the food for ¹³⁷Cs uptake by predators living in lower radioactivity sea water. Besides, benthic animals are estimated to feed on detritus from surface water or prey on the organisms which feed on the detritus. Therefore, the similar ¹³⁷Cs concentration levels be-

tween surface fishes and benthic ones were probably caused by the contribution of the food chain in benthic environment, as suggested in our previous report.¹⁾

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