$^{239,240}\text{Pu}$ and ^{137}Cs Concentrations in Some Marine Biota, Mostly from the Seas around Japan

Yutaka Nagaya* and Kiyoshi Nakamura*

(Accepted September 11, 1986)

Recent $^{239,240}\text{Pu}$ and ^{137}Cs concentrations in some marine biota collected mostly from the seas around Japan were determined. Ash samples were digested with hot nitric acid, and the radionuclides were purified by ion exchange processes, then their alpha and beta radioactivities were measured.

$^{239,240}\text{Pu}$ concentrations were in levels of 1/10 to 1/100 of ^{137}Cs concentrations, and showed values of some pCi/100 kg-raw in muscles. No clear difference was observed in the contents of the radionuclides of muscles between shallow waters and deep bottom inhabitants.

Considerable variation in $^{239,240}\text{Pu}$ concentrations were shown among organs and tissues, but ^{137}Cs showed no such discrepancy.

Apparent high concentration factors (CF) for ^{137}Cs of the deep water inhabitants were supposed to be caused by the contribution of the food chain in the deep sea.

Concentrations of the radionuclides in krill and fur seal were also discussed.

Concentrations of artificial radionuclides in marine biota have been measured since late 1950's, as a part of national radioactivity surveillance program in Japan, and the results have been reported (for example, Radioactivity Survey Data in Japan, No. 73¹⁾ and Proceedings of 27th Meeting on Environmental Radioactivity Survey in Japan²⁾). Initial objects of the measurements were mostly bomb-produced radionuclides, ^{90}Sr , ^{137}Cs , ^{144}Ce etc., and in due course, with the development in peaceful uses of atomic energy ^{60}Co , ^{54}Mn , ^{106}Ru , ^{131}I etc. were also involved. But as for the isotopes of plutonium, despite their long half lives (^{239}Pu ; 87.7 y, ^{240}Pu ; 2.41×10^4 y, ^{241}Pu ; 6.57×10^3 y) and their common presence in radioactive fallout and in liquid wastes from nuclear fuel reprocessing plants, measurements have not been done so intensively, except in a case of environmental monitoring around a nuclear reprocessing facility.³⁾

In this report, we present the recent $^{239,240}\text{Pu}$ levels of some marine biological samples collected from the seas around Japan and some reference areas, as a part of a research program on behaviour of radioactive materials in the marine environment.⁴⁾ ^{137}Cs levels are also shown, considering the difference with those of plutonium.

Materials and Methods

Coastal samples were collected from seas around Ibaraki and Hokkaido. Fur seal and krill samples

Fig. 1. Analytical Scheme of Plutonium.

* Division of Marine Radioecology, National Institute of Radiological Sciences, Isozaki 3609, Nakaminato 311-12, Japan (長屋 裕, 中村 清: 放射線医学総合研究所, 海洋放射生態学研究部).

Table 1. List of Samples

Sample No.	Location of Sample Collection		Year	Biota	Common Name	Scientific Name
1		Ibaraki	1981	Pisces	Suzuki	<i>Lateolabrax japonicus</i>
2		"	"	"	Hirame	<i>Pralichthys olibaceus</i>
3	Central	"	"	"	Hobo	<i>Chelidonichthys spinosus</i>
4	Japan	"	1983	"	Maanago	<i>Conger myriaster</i>
5	—Shallow	Off Boso P.	"	"	Katsuo	<i>Euthynnus pelamis</i>
6	Waters—	Ibaraki	"	Crustacea	Hiratsumegani	<i>Ovalipes punctatus</i>
7		"	1984	"	Okiami	<i>Euphausia pacifica</i>
8		"	1977	Cephalopoda	Iidako	<i>Octopus ocellatus</i>
9	NW-	Off Boso P.	1981	Pisces	Hokakedara	<i>Coryphaenoides</i> sp.
10	Pacific	30°N, 146°E	1982	"	"	<i>Coryphaenoides</i> sp.
11	—Deep	Sagami Bay	1981	"	Kongoanago	<i>Simenchelys parasiticus</i>
12	Bottom—	"	"	"	Irakoanago	<i>Synaphobranchus kaupii</i>
13	(1,500–6,000 m)	"	"	Crustacea	Ezoibaragani	<i>Paralomis multisipina</i>
14	Northern	Off Sanriku	1984	Pisces	Suketodara	<i>Theragra chalcogramma</i>
15	Japan	Toshima P.,	"	"	Madara	<i>Gadus macrocephalus</i>
16	—Cold	Hokkaido	"	"	Madara	<i>Gadus macrocephalus</i>
16	Waters—	"	"	Cephalopoda	Surumeika	<i>Ommastrephes sloanei pacificus</i>
17			1984	Crustacea	Krill	<i>Euphausia superba</i>
18	Southern Ocean		"	"	"	"
19	—Cold Waters—		"	"	"	"
20	(72°–75°E; 62°–68°S)		"	"	"	"
21			"	"	"	"
22	N-North	Off Sanriku	1984	Mammalia	Fur Seal (11y)	<i>Callorhinus ursinus</i>
23	Pacific					
23	—Cold	Bering Sea	1984	"	" (3 y)	"
24	Waters—	"	"	"	" (2 y)	"

Remarks: No. 9–No. 13 were supplied by courtesy of Radioactive Waste Management Center.

No. 17–No. 21 were supplied by courtesy of Dr. Y. Komaki, Far Seas Fisheries Research Laboratory.

No. 22–No. 24 were supplied by courtesy of Dr. K. Yoshida, Far Seas Fisheries Research Laboratory.

were collected on two research cruises conducted by Fisheries Agency—R/V Shunyo Maru's Investigations on North Pacific Fur Seal Stock in 1984 in the Bering Sea, and R/V Kaiyo Maru's SIBEX 83/84 (KY58-2) in the Southern Ocean, Indian Sector. Deep sea samples were collected on three cruises for fish trap experiments by Radioactive Wastes Management Center.^{5,9)} List of samples are shown in Table 1.

Samples were dissected, dried for 48 h at 110°C and ashed at 450 to 550°C for 48 h. 2 dpm of ²⁴²Pu spike (8 M-HNO₃ solution) and 20 mg of Cs⁺ (2 M-HCl solution) were added to each ashed sample (5 to 20 g). The sample was digested by 50 ml of hot 8 M-HNO₃. The solution was filtrated through a glass-fiber filter, and the residue was decomposed by adding 50 ml of H₂O₂ aq. dropwise and by heating on a hot plate, then dissolved with another 50 ml of hot 8 M-HNO₃, and filtered. Both filtrates were combined and evaporated to

nearly dryness on a hot plate, then 100 to 200 ml of 8 M-HNO₃ was added. The solution was boiled, and residue was removed with a glass-fiber filter. Analytical processes after the digestion and the filtration are shown in Fig. 1. Plutonium was purified by three steps of anion-exchange resin column treatment, and finally electrodeposited (500 mA, 2 h) on a stainless steel disc (dia. 25 mm) and its alpha-ray spectrum was measured by a surface-barrier silicon detector (Tennelec) coupled with a 4096 ch. pulse-heights analyzer. The chemical recovery of plutonium isotopes were estimated from ²⁴²Pu activity and showed values of 20 to 80%. The samples with large amounts of residue showed rather poor recovery rates.

¹³⁷Cs was coprecipitated with ammonium molybdo-phosphate from effluent obtained in the first anion-exchange resin column treatment in plutonium analysis. After purified by a cation-exchange resin column treatment, ¹³⁷Cs was

Table 2-1. $^{239,240}\text{Pu}$ Contents (pCi/100 kg-raw)

Sample No.	Muscle	Viscera and/or Digestive Tract			Gill	Skin/Shell
		Viscera	Digestive Tract	Viscera and Dig. Tract		
1	0.0 ± 0.2	—	—	—	—	—
2	1.0 ± 0.5	—	—	—	—	—
4	0.0 ± 0.6	0.8 ± 0.8	1.9 ± 0.4	—	—	0.0 ± 0.3
5	1.6 ± 0.5	39.9 ± 5.6	3.1 ± 1.5	—	14.7 ± 6.2	2.3 ± 2.3
6	1.1 ± 0.6	—	—	18.2 ± 2.8	27.7 ± 6.8	11.9 ± 3.0
8	0.8 ± 0.6	—	—	—	—	—
9	1.8 ± 0.6	0.0 ± 1.1	15.2 ± 2.8	—	—	8.5 ± 3.6
10	11.5 ± 6.5	—	30.3 ± 3.0	—	—	23.4 ± 5.2
11	0.0 ± 1.0	0.6 ± 0.9	4.9 ± 1.1	—	—	0.0 ± 0.6
12	0.0 ± 0.9	0.0 ± 0.1	4.5 ± 1.1	—	—	0.0 ± 1.3
13	2.4 ± 0.5	—	—	54.6 ± 2.7	—	88.8 ± 13.2
14	0.0 ± 0.4	—	—	7.2 ± 1.8	—	3.6 ± 1.8
15	0.1 ± 0.3	—	—	0.0 ± 0.2	—	—
16	3.7 ± 1.2	—	—	16.6 ± 1.3	—	—

Note: Precision criterion is estimated from counting statistics.

Table 2-2. ^{137}Cs Contents (pCi/kg-raw)

Sample No.	Muscle	Viscera and/or Digestive Tract			Gill	Skin/Shell
		Viscera	Digestive Tract	Viscera and Dig. Tract		
1	15.3 ± 0.6	—	—	—	—	—
2	9.2 ± 0.6	—	—	—	—	—
4	4.5 ± 0.2	2.3 ± 0.6	3.4 ± 0.4	—	—	2.5 ± 0.2
5	13.2 ± 0.3	23.2 ± 1.1	11.2 ± 1.3	—	5.3 ± 0.7	6.7 ± 1.0
6	2.2 ± 0.3	—	—	1.5 ± 0.4	1.4 ± 0.4	0.1 ± 1.3
8	0.1 ± 0.2	—	—	—	—	—
9	8.1 ± 0.3	4.8 ± 0.8	2.7 ± 0.4	—	—	4.0 ± 0.6
10	3.6 ± 0.4	—	3.9 ± 0.6	—	—	1.9 ± 1.7
11	9.1 ± 0.9	4.9 ± 1.1	4.8 ± 2.4	—	—	3.0 ± 0.6
12	8.0 ± 0.9	4.5 ± 1.1	3.2 ± 2.6	—	—	5.7 ± 1.1
13	0.0 ± 0.2	—	—	0.4 ± 0.2	—	1.2 ± 4.0
14	6.2 ± 0.7	—	—	1.3 ± 0.2	—	3.4 ± 0.4
15	7.3 ± 0.4	—	—	1.8 ± 0.2	—	—
16	1.5 ± 0.2	—	—	1.3 ± 0.4	—	—

Note: Precision criterion is estimated from counting statistics.

collected as platinochlorate and its beta-ray radioactivity was measured by a low-background gas-flow counter (Tracer Lab, Omni-Guard type) Recovery of ^{137}Cs were measured gravimetrically and showed values of 50 to 90%.

The analytical procedures were the same for sea water and sediment analyses, except decomposition and pretreatments.⁸⁻¹¹⁾

Results

Results of analyses are shown in Table 2 and 3, and Fig. 2. A part of the results on deep bottom

Table 2-3. $^{239,240}\text{Pu}$ and ^{137}Cs Contents of Krill (whole body)

Sample No.	Location	$^{239,240}\text{Pu}$ (pCi/100 kg-raw)	$^{137}\text{Cs}^*$ (pCi/kg-raw)
7	Ibaraki coast	26.8 ± 3.1	2.6 ± 0.3
17	Southern Ocean	17.2 ± 2.9	4.2 ± 0.4
18	"	16.5 ± 2.7	4.9 ± 0.4
19	"	35.8 ± 3.8	5.2 ± 0.5
20	"	39.5 ± 6.0	3.7 ± 0.4
21	"	43.5 ± 4.6	2.9 ± 0.3

Note: Precision criterion is estimated from counting statistics.

* Results of duplicate analyses by this work and by Ueda *et al.*¹³⁾

Table 2-4. $^{239,240}\text{Pu}$ and ^{137}Cs Contents in Fur Seal

Sample No.	Nuclide	Muscle	Liver	Kidney	Lung	Heart
22	$^{239,240}\text{Pu}$ (pCi/100 kg-raw)	0.8 ± 0.3	14.3 ± 3.1	3.9 ± 0.8	0.0 ± 0.9	5.6 ± 2.2
23		0.2 ± 0.2	—	—	—	—
24		0.0 ± 0.3	—	—	—	—
22	^{137}Cs (pCi/kg-raw)	$10.1 \pm 0.3^{*2}$	$3.8 \pm 0.4^{*1}$	$0.6 \pm 0.1^{*1}$	$0.3 \pm 0.1^{*1}$	$12.4 \pm 1.5^{*1}$
23		$6.8 \pm 0.3^{*2}$	—	—	—	—
24		10.1 ± 0.3	—	—	—	—

Note: Precision criterion is estimated from counting statistics.

*¹ Data obtained by Ueda *et al.*¹⁴⁾

*² Results of duplicate analyses by this work and by Ueda *et al.*¹⁴⁾

Table 3. Mean Contents of $^{239,240}\text{Pu}$ and ^{137}Cs of Samples No. 1–No. 16

Tissue & Organ	Sample	$^{239,240}\text{Pu}$ (pCi/100 kg-raw)				^{137}Cs (pCi/kg-raw)			
		Nos.	Mean Value	Standard Deviation	Counting Error	Nos.	Mean Value	Standard Deviation	Counting Error
Muscle	Pisces	10	1.6	± 3.6	± 0.7	10	8.5	± 3.6	± 0.2
	Crustacea	3	1.5	± 0.8	± 0.3	3	1.5	± 1.3	± 0.2
	Cephalopoda	2	2.3	± 2.1	± 0.7	2	0.8	± 1.0	± 0.1
Viscera	Pisces	8	9.9	± 16.0	± 0.9	8	5.8	± 7.2	± 0.3
	Crustacea*	2	36.4	± 25.7	± 1.9	3	1.2	± 1.1	± 0.2
	Cephalopoda*	1	16.6	—	± 1.3	1	1.3	—	± 0.4
Digestive Tract	Pisces	5	9.8	± 6.8	± 1.3	5	5.1	± 3.5	± 0.8
Gill	Pisces	2	14.6	± 3.8	± 6.3	2	5.3	± 1.3	± 0.7
	Crustacea	1	27.7	—	± 6.8	1	1.4	—	± 0.4
Skin or Shell	Pisces	7	5.4	± 8.5	± 1.0	7	3.9	± 1.7	± 0.4
	Crustacea	2	50.4	± 54.3	± 6.8	2	0.7	± 0.8	± 2.1

Note: * Including digestive tract.

samples were reported by Shimizu *et al.*¹²⁾ The present levels of the radionuclides in sea waters and sediments are also shown in Table 4.

In general, $^{239,240}\text{Pu}$ concentrations in marine biological samples were about 1/10 to 1/100 of those for ^{137}Cs , and seemed to coincide roughly with the difference of their levels in sea water.

As shown in Table 3, levels of $^{239,240}\text{Pu}$ in muscle were about the same among pisces, crustacea and cephalopoda, and showed values of some pCi/100 kg-raw. On the other hand, ^{137}Cs concentrations in pisces muscle tended to be rather higher than those of crustacea and cephalopoda, as observed in other investigations.⁴⁾

Digestive tracts and other viscera showed higher $^{239,240}\text{Pu}$ concentrations, nearly one order of magnitude higher than concentrations in muscles, and the variations among samples were considerably large. On the contrary, the ^{137}Cs levels showed no significant differences from those of muscles, and slightly higher values for pisces in

comparison with crustacea and cephalopoda, as similar to the differences in muscles.

$^{239,240}\text{Pu}$ concentrations in gill were nearly 10 times of those in muscles, but ^{137}Cs levels showed no such discrepancy.

As for the skin, $^{239,240}\text{Pu}$ concentrations of pisces were in the same level with those of muscles, but shells of crustacea showed remarkably higher levels than those of muscles. On the other hand, ^{137}Cs contents in shell or skin of the samples were not so different from those in other organs or tissues.

As shown in Fig. 2, among the pisces, *Theragra chalcogramma* showed the highest $^{239,240}\text{Pu}$ content in spine bone, almost 10 times of those for other pisces, which showed rather similar or slightly higher $^{239,240}\text{Pu}$ contents in spine bone than in muscles.

With a exception of *Euthynnus pelamis*, which seems to accumulate the radionuclides intensively in its viscera, digestive tracts of pisces showed

Fig. 2. $^{239,240}\text{Pu}$ Contents in Spine Bones.

rather higher $^{239,240}\text{Pu}$ concentrations than in their other viscera, and the tendency was remarkable in a case of deep bottom inhabitant *Corphaenoides* sp. But ^{137}Cs contents of viscera among pisces hardly varied, except a higher value for *Euthynnus pelamis*.

It was very interesting that a crustacea *Euphausia superba* from the Southern Ocean showed about the same $^{239,240}\text{Pu}$ and ^{137}Cs levels to the North Pacific *Euphausia pacifica*.

$^{239,240}\text{Pu}$ and ^{137}Cs contents in fur seal muscle were of nearly the same levels as those of pisces muscle collected from shallow waters. $^{239,240}\text{Pu}$ content in spine bone of adult fur seals is higher than that of juveniles.

Discussion

It is reported that plutonium isotopes accumulated in marine biota are supplied mainly from sea water, and the food chain showed less contribution for accumulation.^{16,17)} Besides, considerable portions of plutonium in seas and oceans were known to be in particulate form, because of their involvement in the biological processes within the surface layer,^{18,19)} and significant amounts of the particulate plutonium are reported to sink rather rapidly²⁰⁾. Their vertical distribution shows a subsurface maximum, increase of concentration in near-bottom waters and similar concentration levels between surface and deep layers.¹⁰⁾ The concentration level of $^{239,240}\text{Pu}$ in deep water often exceeds that of surface water.¹⁰⁾ A part of particulate plutonium is to be removed from water and deposited in bottom sediments.^{11,15)}

Table 4. Recent Levels of $^{239,240}\text{Pu}$ and ^{137}Cs Contents in Waters and Sediments of Western Northwest Pacific

	$^{239,240}\text{Pu}$	^{137}Cs
	(pCi/10000 l)	(pCi/100 l)
Surface Water	8-11	13-20
(<200 m)		
Sea Water*1 Subsurface Water	5-16	0.1-20
(200-1500 m)		
Deep Water	2-12	0.1-1.2
(>1500 m)		
	(pCi/kg-dry)	(pCi/kg-dry)
Sediment*2 Surface (0-5 cm)	4-30	30-340

Note: *1 Data from Nagaya and Nakamura.^{9,10)}

*2 Data from Nakamura and Nagaya¹¹⁾ and Yang *et al.*¹⁵⁾

So, the general tendencies in $^{239,240}\text{Pu}$ accumulation by biota observed in Table 2-1, that meant that (1) levels in gill, skin or shell are higher than the level in muscle, and that (2) the concentration in digestive tracts shows higher value in comparison with the other viscera, and also that (3) no significant difference in concentrations is observed between the contents in muscles of shallow or surface water inhabitants and in deep bottom ones, are believed to be caused by the particle formation and rapid sinking of plutonium in the sea water.

Higher $^{239,240}\text{Pu}$ concentrations in viscera and shell of deep bottom crab *Paralomis multispina* in comparison with those in shallow water crab *Ovalipes punctata* might be caused by higher concentrations in fine deep sea sediments than in the rather coarse sand of the coastal sea off Ibaraki.

As shown in Table 3, ^{137}Cs concentrations of marine biota were generally at similar levels, and this tendency is clearly distinct from that of $^{239,240}\text{Pu}$ contents. No higher accumulation in skin, shell and viscera is observed, and also no clear difference of the concentrations is indicated between shallow water inhabitants and deep bottom ones. Unlike plutonium, most of ^{137}Cs is supposed to be dissolved in sea water and transported with its movement.^{9,10)} As shown in Table 4, the concentration in surface water decreases sharply with increasing depth and reaches a value of 1/10 to 1/100 of that in the top of the water column. The concentration in the deep layer is rather uniform, and sometimes shows an increase toward the bottom.^{9,10)} A small portion of ^{137}Cs is believed to become particulate and accumulated in sediment surface after rather rapid sinking

through the water column.^{11,15)}

In the assessment of radioactive pollution of marine biota, concentration factor (a ratio of radionuclide contents in biota and in sea water) is commonly used to compare affinity of biota to radionuclides. The concept of CF is based on the assumption that all radionuclides accumulated by biota are derived from sea water as dissolved materials. As the contents of ¹³⁷Cs are in similar levels among marine biota (Table 3) and the concentrations in sea water are different (1 to 2 orders of magnitude, between surface and deep waters as shown in Table 4), the concentration factors (CF) of ¹³⁷Cs for deep bottom biota could be 100 times of those in shallow water biota, but this remarkable discrepancy is unlikely within the same class of pisces, cephalopoda or crustacea.

Suzuki *et al.*²¹⁾ reported that ¹³⁷Cs in pisces are accumulated through both environmental sea water and their foods, in nearly equal rates when food biota have been reared in the sea water containing radioactivity of the same level as that for the predator. Then, the higher the ¹³⁷Cs concentration in sea water for food biota, the larger the contribution of the food for ¹³⁷Cs uptake by predators living in sea water of lower radioactivity, and deep bottom biota are estimated to feed on detritus from surface water, or prey on the biota, which feed on the detritus. So, the apparent high CFs of ¹³⁷Cs in deep bottom biota, that meant similar ¹³⁷Cs concentration levels between shallow water biota and deep bottom ones, were probably caused by the contribution of the food chain in the deep sea.

Higher radionuclide contents in viscera of *Euthynnus pelamis* imply intensive accumulation of minor elements in viscera by pelagic migratory fishes. As for *Theragra chalcogramma*, we have no explanation for the high plutonium content in spine bone, in comparison with other fishes.

Similar concentration levels of radionuclides in *Euphausia superba* and *Euphausia pacifica* might imply similar levels of radionuclides in the surface waters of the Southern Ocean and the North Pacific, but more investigations are required to clarify this suggestion.

In comparison with human adults,^{22,23)} ^{239,240}Pu contents of an 11 year old fur seal was considerably lower in muscle and lung, rather lower in liver, nearly equal in kidney, and somewhat lower in spine. Plutonium contained in human (a mammalian) body is reported to be taken mostly by inhalation. So, lower content in the lung

suggests that either fur seal might accumulate plutonium rather through its food chain or plutonium content in the recent North Pacific air is considerably lower than before 1975.

Acknowledgement

We wish to express our hearty thanks to the staff of Radioactive Wastes Management Center and to Drs. Y. Komaki and K. Yoshida of Far Seas Fisheries Research Laboratory, Fisheries Agency, for supplying the precious samples.

References

- 1) Japan Chemical Analysis Center: Radioactivity Survey Data in Japan, No. 73, Science and Technology Agency, Tokyo, 1985, pp. 25-30.
- 2) K. Yoshida and K. Tabata: Proceedings of 27th Meeting on Environmental Radioactivity Survey in Japan, Science and Technology Agency, Tokyo, 1985, pp. 94-96. (in Japanese)
- 3) N. Miyakawa, N. Hayashi, J. Ishida, and M. Iwai: Proceedings of 27th Meeting on Environmental Radioactivity Survey in Japan, Science and Technology Agency, Tokyo, 1985, pp. 119-120. (in Japanese).
- 4) Y. Nagaya, Y. Suzuki, K. Nakamura, R. Nakamura, N. Ishii, and T. Ueda: Proceedings of 27th Meeting on Environmental Radioactivity Survey in Japan, Science and Technology Agency, Tokyo, 1985, pp. 86-89. (in Japanese)
- 5) Radioactive Wastes Management Center: Report on Development of a Fish Trap and Its Test (I), 1981, pp. 1-20. (in Japanese)
- 6) Radioactive Wastes Management Center: Report on Development of a Fish Trap and Its Test (II), 1982, pp. 1-20. (in Japanese)
- 7) Radioactive Wastes Management Center: Report on Development of a Fish Trap and Its Test (III), 1982, pp. 1-24. (in Japanese)
- 8) D. C. Sutton, G. Calderon, and W. Rosa: HASL-307, ERDA, New York, 1976, pp. 1-43.
- 9) Y. Nagaya and K. Nakamura: *J. Oceanogr. Soc. Japan*, **37**, 135-14 (1981).
- 10) Y. Nagaya and K. Nakamura: *J. Oceanogr. Soc. Japan*, **40**, 416-424 (1984).
- 11) K. Nakamura and Y. Nagaya: in "Marine and Estuarine Geochemistry" (ed. by A. C. Sigleo and A. Hattori), Lewis Pub. Inc., Chelsea, 1985, pp. 171-179.
- 12) S. Shimizu, Y. Kurabayashi, K. Fukuda, Y. Horibe, Y. Kodama, T. Gamo, Y. Nagaya, and M. Michizima: Proceedings of Radioactive Wastes Forum '84, Science and Technology Agency, Tokyo, 1984, Chapter 2, pp. 19-28.
- 13) T. Ueda, N. Ishii, Y. Suzuki, R. Nakamura, and

- M. Nakahara: *Nippon Suisan Gakkaishi*, (to be published)
- 14) T. Ueda, N. Ishii, Y. Suzuki, M. Nakahara, and R. Nakamura: *J. Radiat. Res.*, (to be published).
- 15) H. Yang, Y. Nozaki, H. Sakai, Y. Nagaya, and K. Nakamura: *Geochim. J.*, **20**, 29–40 (1986).
- 16) J. C. Guary and A. Fraizer: *Health Phys.*, **32**, 21–28 (1977).
- 17) J. C. Guary, S. W. Fowler, and T. M. Beaseley: *Mar. Poll. Bull.*, **13**, 99–102 (1982).
- 18) S. W. Fowler, S. Vallesstra, J. LaRosa, and R. Fukai: *Deep-Sea Res.*, **30**, 1221–1233 (1983).
- 19) E. R. Sholkovitz: *Earth Sci. Rev.*, **19**, 95–161 (1983).
- 20) H. D. Livingston and R. F. Anderson: *Nature*, **303**, 228–231 (1983).
- 21) Y. Suzuki, M. Nakahara, R. Nakamura, and T. Ueda: *Nippon Suisan Gakkaishi*, **45**, 1409–1516 (1979).
- 22) H. Kawamura and G. Tanaka: *Health Phys.*, **44**, suppl. 1, 451–461 (1983).
- 23) Y. Igarashi, R. Seki, and N. Ikeda: *J. Radiat. Res.*, **27**, 213–218 (1986).