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Baseline

Concentrations of ^{137}Cs, ^{90}Sr, ^{108m}Ag, $^{239+240}$Pu and atom ratio of ^{240}Pu/^{239}Pu in tanner crabs, *Chionoecetes japonicus* and *Chionoecetes opilio* collected around Japan

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ABSTRACT

The anthropogenic radionuclides, ^{137}Cs, ^{90}Sr, ^{108m}Ag, $^{239+240}$Pu, were measured in two *Chionoecetes* species, red queen crab (*Chionoecetes japonicus*) and snow crab (*Chionoecetes opilio*) collected around Japan during 1996–2007. There was no increase in the concentrations of these radionuclides and no large variation of the atom ratio of ^{240}Pu/^{239}Pu during this research period. These results indicated that the source of the radionuclides was not the radioactive wastes dumped by the former USSR and Russia and originated from past nuclear weapon tests. The higher atom ratio in the crab species than that from global fallout would be contributed by the Pacific Proving Grounds close-in fallout. The variability of the concentration of radionuclides in the crab species would result from the variability of the composition and quantity in the diet. However, the decrease in the concentration of radionuclides with sampling depth would depend on the concentration in the seawater and diet.

The former Union of Soviet Socialist Republics (USSR) and Russia dumped radioactive wastes, which included about 440 TBq of liquid and 140 TBq of solid-state wastes, in the northern part of the Japan Sea, and about 0.01 TBq of liquid and 113 TBq of solid-state wastes in the north-western part of the Pacific Ocean off the Kamchatka Peninsula (IAEA, 1999; Yablokov 2001). In order to assess the radiological impact from the dumped radioactive wastes, two Japanese–Korean–Russian joint expeditions with the IAEA's Marine Environment Laboratory (IAEA-MEL) were carried out in the Japan Sea and its marginal seas in 1994 and 1995 (Hirose et al., 1999; Ikeuchi et al., 1999; Kim et al., 2003; Pettersson et al., 1999). These expeditions and later other surveys have been unable to detect any anthropogenic radionuclides originating from the dumped radioactive wastes (Ito et al., 2003; Morita et al., 2007; Zheng and Yamada, 2005).

The Japan Sea has an area of 1.01×10^6 km^2, a mean depth of 1350 m and a maximum depth of 3796 m. The Japan Sea is separated from the Pacific Ocean, the East China Sea, and the Sea of Okhotsk by four shallow straits. The main exchange of seawater occurs in the surface layer (upper 200–300 m) and there is a low rate exchange in the deeper water. The turnover time of deep water in the Japan Sea is estimated to be ca. 100 years (Watanabe et al., 1991). These characteristics of the Japan Sea would make the radioactive pollution from the dumped radioactive wastes remain in its deep layer for a long time.

Two tanner crab species, red queen crab *Chionoecetes japonicus* and snow crab *Chionoecetes opilio* occur around Japan. These species have different habit depths to each other, from ca. 500 to ca. 2700 m for red queen crab and shallower than ca. 500 m for snow crab, and distribute overlapping only at the boundary zone (Fujikura et al., 2000; Yosho and Hayashi, 1994). These crab species would potentially be initially polluted by radionuclides released from the dumped radioactive wastes as they feed in deep benthic habitats. These two crab species are also commercially important species in Japan, especially supporting fisheries in the Japan Sea. There has been high concern about radioactive pollutants in marine organisms in Japan, because those are frequently used as food sources (Kasamatsu and Ishikawa, 1997; Morita et al., 2007, 2010a,b; Nagaya et al., 1990; Suzuki et al., 1973). Therefore, much attention has been paid to the concentrations of radionuclides in the two crab species collected in the Japan Sea.

An anthropogenic radionuclides detected in marine organisms collected around Japan usually have a long physical half-life, e.g. ^{137}Cs (physical half-life: 30.1 years), ^{90}Sr (28.8 years), ^{108m}Ag (418 years), ^{239}Pu (24,110 years) and ^{240}Pu (6564 years). Although these radionuclides were introduced into the marine environment from nuclear weapon production, nuclear power stations, nuclear fuel reprocessing plants, dumping of radioactive wastes, accidents involving nuclear materials and so on (UNSCEAR, 2000; Hamilton,

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2004), the main source is the stratospheric (global) fallout from atmospheric nuclear weapon tests conducted from 1945 to 1980, which peaked in 1963 (Bowen et al., 1980; UNSCEAR, 2000). The atom ratio of $^{240}Pu/^{239}Pu$ is frequently used to identify the source of Pu isotopes in the environment (Kenna and Sayles, 2002; Olivier et al., 2004; Yamada and Zheng, 2008; Zheng and Yamada, 2004, 2005) since the atom ratio depends upon the specific weapon design and test yields (Cooper et al., 2000).

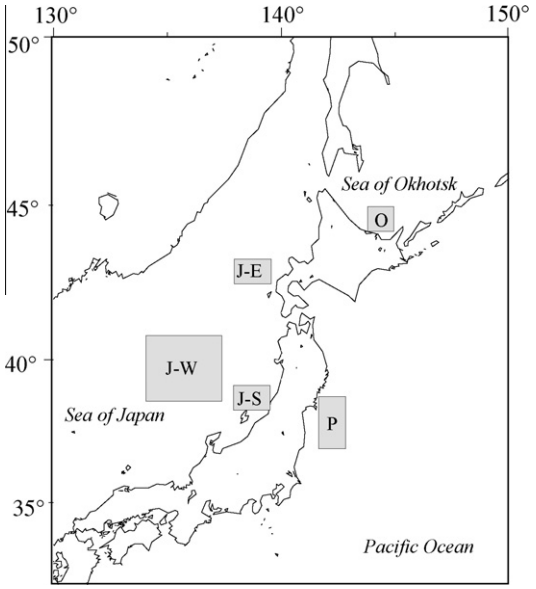

Fig. 1. Map of the sampling sites. Red queen crab (*Chionoecetes japonicus*) samples were collected in the region J-W, J-E, J-S and P. Snow crab (*C. opilio*) samples were in J-W and O.

Here we report on the temporal variation of ^{137}Cs, ^{90}Sr, ^{108m}Ag, $^{239+240}Pu$ concentrations and the $^{240}Pu/^{239}Pu$ atom ratio in two crab species, red queen crab and snow crab collected around Japan during 1996–2007. Our results show that the sources of the radionuclides detected in two crab species did not originate from the radioactive wastes dumped by the former USSR and Russia, but mainly from the past nuclear weapon tests. The radioactive concentration data obtained in this study show that the concentration in the two crab species is in the normal situation, *i.e.* what can be called 'the back-ground concentration'. The data are valuable as baseline data for evaluating potential leakages from the dumped radioactive material and also for assessing the radiation dose from human consumption of marine food products.

Samples were collected using large crab pots by the R/V Soyo–maru of the National Research Institute of Fisheries Science Fisheries Agency and using trawl nets by the R/V Wakataka–maru of Tohoku National Fisheries Research Institute Fisheries Agency. Sampling sites and information are shown in Fig. 1 and Table 1. We divided the samples into shell, muscle and hepatopancreas parts in the laboratory. As each part from one individual is only a small amount and has low Pu concentrations, we cannot significantly detect them. Therefore, the samples for measurement were prepared by combining each part from many individuals. The muscle, hepatopancreas and shell samples consisted of tissues from ca. 130 individuals, respectively. All divided samples were dried in an oven at 105 °C for 72–120 h, carbonized in a gas furnace at 350–400 °C for about 6 h, and ashed in an electric furnace at 450 °C for 48–72 h.

A 40 g of an ashed sample was decomposed with nitric acid and hydrogen peroxide after an addition of a known amount of ^{242}Pu as

Table 1
Sample information.

Species	Sample code	Sampling data	Region	Latitude	Longitude	Depth (m)
Chionoecetes japonicus red queen crab	R-J-1	1997.07.14	J-W	40-56.0N	136-06.0E	1360
	R-J-2	1998.07.23	J-W	39-56.0N	136-06.0E	1355
	R-J-3	1999.07.23	J-W	39-59.0N	135-60.0E	1430
	R-J-4	2000.07.21	J-W	39-13.0N	135-30.0E	1260
	R-J-5	2002.07.25	J-W	39-55.0N	136-14.0E	1461
	R-J-6	2003.07.27	J-W	39-35.0N	135-55.0E	535
	R-J-7	2003.07.27	J-W	39-56.0N	136-06.0E	1382
	R-J-8	2005.07.28	J-W	40-01.0N	137-05.9E	1750
	R-J-9	2006.08.02	J-W	39-24.9N	135-15.7E	1400
	R-J-10	2007.07.21	J-W	40-03.2N	137-06.1E	1800
	R-J-11	2007.07.22	J-W	39-56.1N	136-06.1E	1290
	R-J-12	2001.07.28	J-W	38-48.0N	134-02.0E	1510
	R-J-13	2004.07.30	J-W	38-48.0N	134-01.0E	1467
	R-J-14	2005.07.29	J-W	39-55.9N	136-06.2E	1320
	R-J-15	2001.07.25	J-E	43-59.9N	139-58.0E	1516
	R-J-16	2005.07.26	J-E	43-01.0N	139-59.9E	1934
	R-J-17	2007.07.26	J-E	42-59.1N	139-59.9E	1341
	R-J-18	1996.07.20	J-S	38-53.0N	138-00.0E	1350
	R-P-1	2000.06.10	P	37-31.4N	141-58.1E	310
	R-P-2	2000.10.16	P	39-37.1N	142-33.7E	878
	R-P-3	2001.06.12	P	35-50.9N	141-35.3E	650
	R-P-4	2007.08.06	P	39-36.0N	142-33.9E	946
Chionoecetes opilio snow crab	S-J-1	2000.07.20	J-W	39-28.0N	135-23.0E	425
	S-J-2	2002.07.26	J-W	39-26.0N	135-41.0E	355
	S-J-3	2005.07.31	J-W	39-24.9N	135-15.7E	256
	S-J-4	2006.08.02	J-W	39-24.4N	135-15.7E	360
	S-J-5	2007.07.23	J-W	39-24.0N	135-12.3E	301
	S-J-6	2001.07.27	J-W	39-00.1N	134-20.0E	329
	S-J-7	2004.07.26	J-W	39-00.0N	134-19.0E	344
	S-O-1	1999.07.31	O	44-27.5N	144-06.0E	350
	S-O-2	2001.07.19	O	44-27.8N	144-05.6E	214
	S-O-3	2003.08.03	O	44-26.0N	144-06.0E	220
	S-O-4	2005.07.20	O	44-27.1N	144-06.6E	213
	S-O-5	2007.07.30	O	44-20.7N	144-05.6E	218

Table 2
Activities of ^{239}Pu and ^{240}Pu and the ^{240}Pu/^{239}Pu atom ratio in the crab species.

Sample code	Shell			Muscle			Hepatopancreas		
	^{239}Pu mBq/kg-wet	^{240}Pu mBq/kg-wet	^{240}Pu/^{239}Pu	^{239}Pu mBq/kg-wet	^{240}Pu mBq/kg-wet	^{240}Pu/^{239}Pu	^{239}Pu mBq/kg-wet	^{240}Pu mBq/kg-wet	^{240}Pu/^{239}Pu
R-J-1	2.38 ± 0.41[a]	<8.68[b]		2.37 ± 0.41	<8.69		7.87 ± 1.66	7.87 ± 2.55	0.27 ± 0.11
R-J-2	28.42 ± 2.43	25.75 ± 5.03	0.25 ± 0.053	<1.90	<2.84		7.2 ± 0.72	6.93 ± 0.64	0.26 ± 0.036
R-J-3	22.79 ± 3.86	21.04 ± 4.38	0.25 ± 0.068	1.70 ± 0.23	2.12 ± 0.27	0.34 ± 0.062	4.63 ± 0.75	4.37 ± 0.69	0.26 ± 0.058
R-J-4	<12.50	<35.80		<2.08	<6.94		6.67 ± 0.84	4.06 ± 1.13	0.17 ± 0.051
R-J-5	10.26 ± 2.32	<19.40		1.97 ± 0.20	<5.98		8.68 ± 0.73	5.32 ± 0.45	0.17 ± 0.02
R-J-6	6.48 ± ± 1.71	<8.36		2.31 ± 0.25	2.01 ± 0.55	0.24 ± 0.07	15.7 ± 1.31	12.47 ± 1.91	0.22 ± 0.038
R-J-7	13.59 ± 1.43	<1.43		2.32 ± 0.17	<2.80		9.77 ± 1.04	8.58 ± 0.83	0.24 ± 0.034
R-J-8	24.08 ± 0.2	22.08 ± 0.80	0.25 ± 0.009	1.66 ± 0.04	1.43 ± 0.07	0.23 ± 0.013	6.22 ± 0.44	5.44 ± 0.39	0.24 ± 0.024
R-J-9	9.57 ± 2.09	8.97 ± 0.27	0.26 ± 0.056	1.39 ± 0.02	1.20 ± 0.04	0.24 ± 0.008	8.38 ± 0.16	7.34 ± 0.18	0.24 ± 0.0075
R-J-10	14.28 ± 0.45	12.98 ± 0.55	0.25 ± 0.013	0.928 ± 0.02	0.82 ± 0.04	0.24 ± 0.012	7.81 ± 0.03	6.7 ± 0.06	0.23 ± 0.0021
R-J-11	13.89 ± 0.24	12.68 ± 0.27	0.25 ± 0.007	0.921 ± 0.03	0.79 ± 0.02	0.23 ± 0.009	5.68 ± 0.11	4.54 ± 0.09	0.22 ± 0.006
R-J-12	8.5 ± 0.93	<12.83		1.71 ± 0.99	1.29 ± 0.42	0.21 ± 0.136	6.38 ± 0.9	7.45 ± 0.8	0.32 ± 0.056
R-J-13	3.68 ± 0.5	2.89 ± 0.42	0.21 ± 0.043	1.42 ± 0.06	1.17 ± 0.04	0.23 ± 0.013	9.66 ± 0.08	8.28 ± 0.11	0.23 ± 0.0037
R-J-14	8.12 ± 0.39	7.14 ± 0.79	0.24 ± 0.029	1.16 ± 0.05	1.02 ± 0.03	0.24 ± 0.011	6.28 ± 0.08	5.46 ± 0.08	0.24 ± 0.0047
R-J-15	11.07 ± 3.31	<2.544		2.04 ± 0.21	2.20 ± 0.71	0.29 ± 0.1	5.3 ± 0.32	5.57 ± 0.64	0.29 ± 0.037
R-J-16	17.26 ± 0.31	16.08 ± 0.59	0.25 ± 0.01	1.90 ± 0.04	1.62 ± 0.12	0.23 ± 0.017	5.82 ± 0.08	5.06 ± 0.03	0.24 ± 0.0035
R-J-17	6.69 ± 0.04	6.10 ± 0.16	0.25 ± 0.007	0.963 ± 0.01	0.86 ± 0.01	0.24 ± 0.006	5.4 ± 0.02	4.59 ± 0.05	0.23 ± 0.0028
R-J-18	8.04 ± 1.47	<22.64		<2.04	<5.84		8.32 ± 0.55	7.28 ± 1.35	0.24 ± 0.047
R-P-1	16.27 ± 2.89	15.37 ± 2.53	0.26 ± 0.062	<5.24	<5.24		10.41 ± 1.35	<5.08	
R-P-2	11.75 ± 0.51	10.90 ± 1.55	0.25 ± 0.038	1.36 ± 0.14	<1.46		17.09 ± 4.01	12.82 ± 0.37	0.2 ± 0.048
R-P-3	7.64 ± 1.21	7.32 ± 1.31	0.26 ± 0.062	1.17 ± 0.19	1.02 ± 0.20	0.24 ± 0.06	13.36 ± 1.75	12.08 ± 1.26	0.25 ± 0.041
R-P-4	7.74 ± 0.22	6.82 ± 0.18	0.24 ± 0.009	0.77 ± 0.02	0.66 ± 0.02	0.23 ± 0.008	13.56 ± 0.11	11.3 ± 0.11	0.23 ± 0.003
S-J-1	<4.01	<40.00		<0.786	<5.24		5.41 ± 0.39	4.43 ± 1.08	0.22 ± 0.057
S-J-2	21.71 ± 2.53	16.82 ± 2.35	0.21 ± 0.038	2.90 ± 0.35	2.09 ± 0.32	0.2 ± 0.038	25.48 ± 2.6	18.2 ± 1.98	0.19 ± 0.029
S-J-3	27.32 ± 0.25	24.09 ± 0.35	0.24 ± 0.004	5.82 ± 0.05	5.06 ± 0.18	0.24 ± 0.009	15.18 ± 0.2	13.16 ± 0.15	0.24 ± 0.0042
S-J-4	10.49 ± 0.22	9.15 ± 0.22	0.24 ± 0.008	1.64 ± 0.02	1.52 ± 0.04	0.25 ± 0.007	17.49 ± 0.21	15.37 ± 0.29	0.24 ± 0.0054
S-J-5	17.69 ± 0.22	15.30 ± 0.22	0.24 ± 0.004	2.39 ± 0.03	1.98 ± 0.05	0.23 ± 0.007	16.56 ± 0.17	14.35 ± 0.11	0.24 ± 0.003
S-J-6	9.62 ± 1.69	8.44 ± 2.36	0.24 ± 0.079	2.13 ± 0.55	1.71 ± 0.50	0.22 ± 0.085	16.17 ± 1.8	14.58 ± 0.9	0.25 ± 0.031
S-J-7	6.16 ± 0.43	5.21 ± 0.40	0.23 ± 0.024	1.17 ± 0.04	0.84 ± 0.07	0.2 ± 0.018	10.45 ± 0.19	9.23 ± 0.24	0.24 ± 0.0078
S-O-1	16.64 ± 3.61	<20.80		<9.66	<6.44		4.46 ± 1.37	2.68 ± 0.81	0.16 ± 0.07
S-O-2	10.79 ± 1.99	<8.14		2.88 ± 0.48	2.14 ± 0.57	0.2 ± 0.063	4.68 ± 0.41	4.13 ± 0.96	0.24 ± 0.06
S-O-3	<1.54	<4.41		0.45 ± 0.11	<0.852		6.47 ± 0.37	4.93 ± 0.34	0.21 ± 0.019
S-O-4	2.08 ± 0.04	1.56 ± 0.41	0.21 ± 0.054	0.31 ± 0.01	0.25 ± 0.03	0.22 ± 0.031	4.46 ± 0.03	3.27 ± 0.06	0.2 ± 0.0039
S-O-5	4.13 ± ± 0.25	3.31 ± 0.33	0.22 ± 0.025	0.27 ± 0.01	0.23 ± 0.05	0.23 ± 0.056	3.28 ± 0.08	2.59 ± 0.07	0.22 ± 0.008

[a] 1σ values derived from three replicates acquisition (total 300 passes scans).
[b] 3σ values derived from three replicates acquisition (total 300 passes scans).

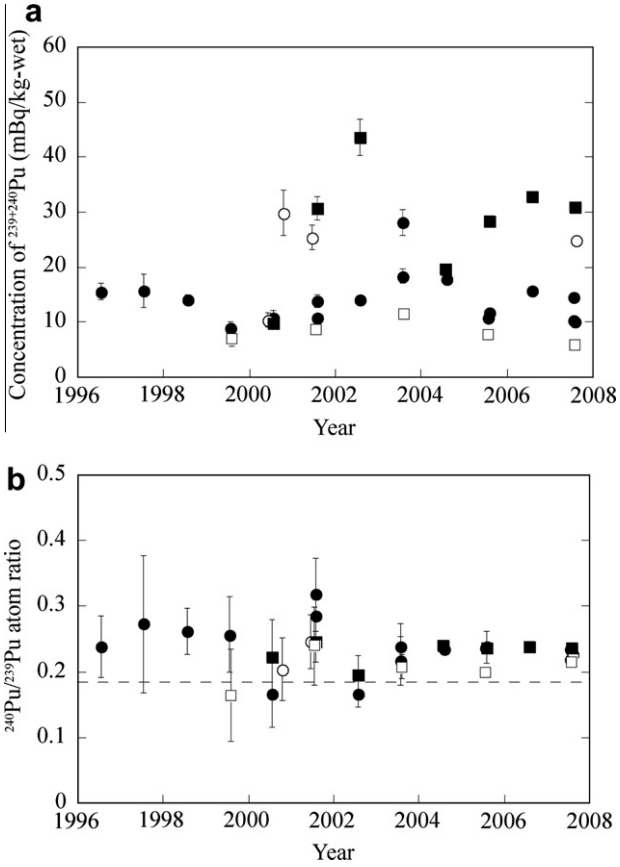

Fig. 2. Temporal variation in concentrations of $^{239+240}$Pu (A) and ^{240}Pu/^{239}Pu atom ratios (B) in the hepatopancreas of the two crab species. Closed and open circles present the data for the red queen crabs collected in the sampling region J and P, respectively. Closed and open squares show the data for the snow crabs collected in the sampling region J and O, respectively. Error bars in (a) show 1σ value from three replicates acquisition (total 300 passes scans). Error bar in (b) was calculated from 1σ value of each Pu isotope. The dotted line (b) shows the atom ratio of ^{240}Pu/^{239}Pu, ca. 1.8, from the global fallout (Kelley et al. 1999; Krey et al. 1976).

a yield monitor. Plutonium in the sample was measured by a sector field ICP-MS (JEOL JMS-PLASMAX2) with an ultrasonic nebulizer after radiochemical separation of Pu using an anion exchange resin. Data of the concentrations for Pu isotopes were expressed in terms of units of radioactivity to allow comparison with the data with those obtained by using α-spectrometry in other studies.

The concentration of ^{90}Sr was determined using well-established radiochemical analytical methods (STA, 1983). ^{90}Sr was purified using ion exchange chromatography. The concentration of ^{90}Sr was determined by measuring beta rays emitted from ^{90}Y in radioactive equilibrium with ^{90}Sr using a low background gas flow GM beta-ray counter. The concentrations of ^{137}Cs and ^{108m}Ag were measured using a high purity germanium (HPGe) semiconductor detector with a multichannel analyzer on the ground. This HPGe semiconductor detector has a resolution of 1.44 keV at a peak of 662.15 keV (^{137}Cs). The counting time was 320,000–560,000 s for one sample. The energy dependent efficiency calibration for HPGe semiconductor detector was conducted with five gamma ray reference sources purchased from the Japan Radioisotope Association. These reference sources contained quantified concentrations of gamma ray radionuclides, ^{54}Mn, ^{57}Co, ^{60}Co, ^{88}Y, ^{109}Cd, ^{137}Cs and ^{139}Ce, and had different radioactive concentrations and heights from each other. The concentrations of three standard deviation (σ) from counting error was defined as the detection limit concentration. The concentrations of all radionuclides were corrected for decay from the sampling date.

A 50–60 mg of an ashed sample was used for measuring stable elements, of P, K, Ca, V, Mn, Fe, Ni, Co, Cu, Zn, Rb, Sr, Ag, Cd, Cs, Ba, Pb, Th, and U. The sample was decomposed completely using a mixed solution of HNO_3, $HClO_4$ and HF in a Teflon-sealed vessel at 150 °C for 5 h. These reagents used were EL grade. Those elements in the sample were analyzed by using a quadrupole ICP-MS (Seiko SPQ9900).

One-way analysis of variance (ANOVA) was used for comparison of more than two groups. If the one-way ANOVA was significant, multiple comparisons between groups were conducted using Tukey–Kramer test. The relations among radioisotopes and between radioisotopes and stable elements were investigated by Pearsons's correlation test.

^{239}Pu and ^{240}Pu could be detected in shell, muscle and hepatopancreas samples of both crab species, while the concentration in some shell and muscle samples was below the detection limit (Table 2). The maximum concentration of $^{239+240}$Pu in shell, muscle and hepatopancreas samples was 54 ± 5.57 mBq/kg-wet in sample

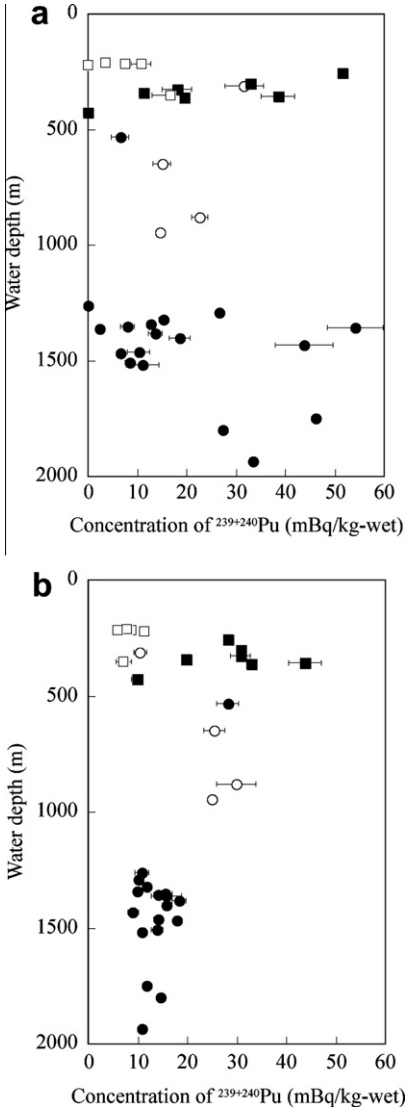

Fig. 3. Relationship between sampling depth and Pu isotopes concentrations in the shell sample (a) and hepatopancreas sample (b). Closed and open circles indicate the data for the red queen crabs collected in the sampling region J and P, respectively. Closed and open squares did the data for the snow crabs collected in the sampling region J and O, respectively. Error bars show 1σ value from three replicate acquisition (total 300 passes scans).

code R-J-2, 11 ± 0.18 mBq/kg-wet in sample S-J-3, 44 ± 2.0 mBq/kg-wet in sample S-J-2, respectively. The previous report showed that that maximum concentration of $^{239+240}$Pu in liver samples of common squid *Todarodes pacificus* collected in the Japan Sea was 52 ± 2.4 mBq/kg-wet (Oikawa and Yamamoto, 2007). There was no statistical difference in concentration of all radionuclides detected in this study among the sampling sites J-W, J-E and J-N. Therefore, these sites were grouped as a sampling region J below. In comparison of each Pu isotope concentration among the three tissues, the concentrations of both Pu isotopes in the muscle samples were statistically lower than those in the other tissue samples ($P < 0.01$). Furthermore, the concentrations of both Pu isotopes in the hepatopancreas of snow crabs in sampling region J were significantly higher than those in sampling region O ($P < 0.05$), whereas there was no difference in the concentration of both Pu isotopes in each tissue of red queen crab between sampling region P and J. On the other hand, there was no significant difference in the atom ratio among the three tissues and among sampling regions (Table 2). Fig. 2 shows the temporal variation of $^{239+240}$Pu concentrations and the atom ratio of ^{240}Pu/^{239}Pu in the hepatopancreas samples. No increase in the concentration and the minimal variability of the atom ratio indicated that the pollution source of the Pu isotopes was the same for all sampling regions during this investigation period, 1996–2007. Fig. 3 shows the relationship between the sampling depth and the $^{239+240}$Pu concentration and the atom ratios in the shell and hepatopancreas samples. The variability of Pu concentration in the hepatopancreas samples decreased with the sampling depth, while such a tendency to decrease was not observed in shell sample. In addition, the Pu concentration in the hepatopancreas samples except for sampling region O also decreased as sampling depth become deeper, whereas that in shell sample did not decrease. On the contrary, the atom ratio in the shell and hepatopancreas samples was similar to each other and almost constant regardless of the sampling depth (Fig. 4).

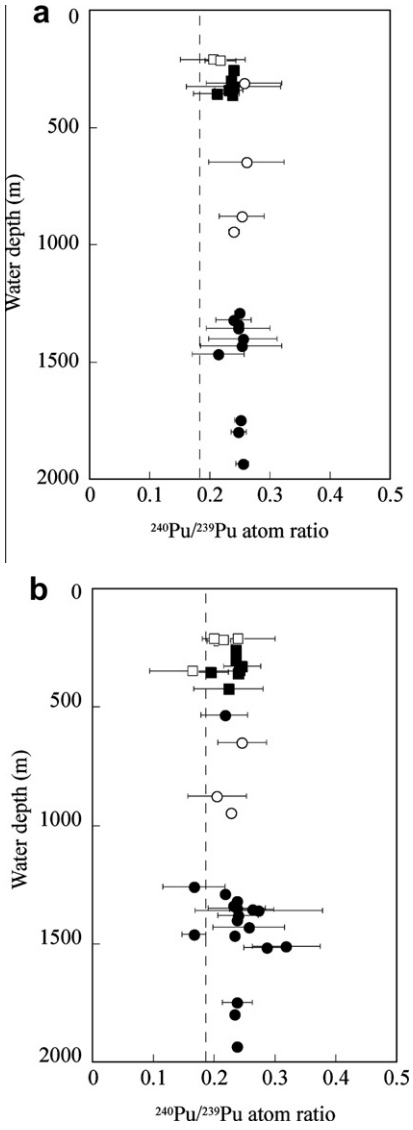

Fig. 4. Relationship between sampling depth and Pu atom ratio in the shell sample (a) and hepatopancreas sample (b). Closed and open circles present the data for the red queen crabs collected in the sampling region J and P, respectively. Closed and open squares indicate the data for the snow crabs collected in the sampling region J and O, respectively. Error bars were calculated from 1σ value of each Pu isotope. The dotted line shows the atom ratio of ^{240}Pu/^{239}Pu, ca. 1.8, from the global fallout (Kelley et al. 1999; Krey et al. 1976).

Table 3 shows the concentrations of anthropogenic gamma emitting radionuclides detected in the muscle and hepatopancreas samples. ^{137}Cs could be detected in the muscle and hepatopancreas samples and ^{108m}Ag was detected in the hepatopancreas samples, whereas anthropogenic gamma emitting radionuclides were not detected in all shell samples. Other anthropogenic gamma emitting radionuclides e.g., ^{54}Mn, ^{55}Fe, ^{60}Co, ^{65}Zn, ^{115}Cd, ^{115m}Cd and ^{207}Bi were detected in marine organisms caught at Bikini and Eniwetok Atolls (Noshkin et al., 1997). ^{103}Ru, ^{110m}Ag, ^{134}Cs and ^{131}I were also detected in marine organisms just after the Chernobyl reactor accident (Molero et al., 1999; Morita et al., 2010b). Such anthropogenic gamma emitting radionuclides, however, were not detected in this study.

^{137}Cs is well known to accumulate in muscle tissues (Young and Folsom, 1979). The concentration of ^{137}Cs in some muscle samples of both crab species was, however, below the detection limit (Table 3). The maximum concentration of ^{137}Cs detected in muscle and hepatopancreas samples was 0.046 ± 0.010 Bq/kg-wet in sample S-O-2 and 0.024 ± 0.0048 Bq/kg-wet in sample S-J-4, respectively. The concentration of ^{137}Cs in muscle samples of both crab species was considerably lower than that in muscle samples of walleye pollock *Theragra chalcogramma* collected in the Japan Sea, ca. 0.25 Bq/kg-wet (Morita et al., 2007). On the contrary the concentrations of ^{137}Cs in the muscle and hepatopancreas samples of common octopus *Octopus vulgaris* collected in the East China Sea, ca. 0.30 Bq/kg-wet for both tissues (Morita et al., 2010b), were the same level as those in the crab species in this study. An increase in the concentration of ^{137}Cs detected in muscle and hepatopancreas samples of both crab species was not observed during this investigation period (Fig. 5a and b). This result indicates that ^{137}Cs has not been newly released into these sampling regions during this period. The concentration of ^{137}Cs detectable in muscle and hepatopancreas samples slightly decreased with the sampling depth (Fig. 6).

The maximum concentration of ^{108m}Ag detected in hepatopancreas sample was 0.21 ± 0.0093 Bq/kg-wet in sample code R-P-1 (Table 3). The concentration was the similar level as that in liver samples of common octopus collected in the East China sea, ca. 0.20 Bq/kg-wet (Morita et al., 2010b). In comparison of the concentration of ^{108m}Ag in hepatopancreas sample among four groups which were red queen crab in sampling regions J and P and snow crab in regions J and O, the concentration in snow crab in sampling region O was statistically lower than those in the other groups ($P < 0.01$). Fig. 7a shows the temporal variation of ^{108m}Ag concentration in the hepatopancreas samples. The low variability of the concentration shows that the source of ^{108m}Ag has been constant in these sampling regions during this investigation period. Fig. 8a shows the relationship between the sampling depth and ^{108m}Ag concentration in the hepatopancreas samples. The variability of

Table 3
Activities of ^{90}Sr, ^{137}Cs and ^{108m}Ag in‘ the crab species.

Sample code	Shell	Muscle	Hepatopancreas	
	^{90}SrBq/Kg-Wet	^{137}CsBq/Kg-Wet	^{137}CsBq/Kg-Wet	^{108m}AgBq/Kg-Wet
R-J-1	N.A.[a]	<0.031[b]	<0.020	0.079 ± 0.0040[c]
R-J-2	N.A.	<0.017	<0.022	0.11 ± 0.0076
R-J-3	0.067 ± 0.017	<0.021	<0.013	0.13 ± 0.0066
R-J-4	0.045 ± 0.014	0.032 ± 0.0093	<0.026	0.11 ± 0.0070
R-J-5	0.11 ± 0.019	<0.023	<0.021	0.068 ± 0.0045
R-J-6	0.096 ± 0.031	0.015 ± 0.0049	0.023 ± 0.0049	0.06 ± 0.0036
R-J-7	0.097 ± 0.019	<0.017	<0.012	0.12 ± 0.0037
R-J-8	0.11 ± 0.024	0.018 ± 0.0048	0.013 ± 0.0035	0.11 ± 0.0039
R-J-9	0.15 ± 0.014	<0.017	0.014 ± 0.0032	0.07 ± 0.0036
R-J-10	0.16 ± 0.014	0.015 ± 0.0036	0.011 ± 0.0036	0.084 ± 0.0038
R-J-11	0.12 ± 0.014	0.015 ± 0.0036	0.011 ± 0.0036	0.083 ± 0.0039
R-J-12	0.047 ± 0.014	<0.022	<0.020	0.070 ± 0.0070
R-J-13	0.087 ± 0.021	<0.017	0.017 ± 0.0037	0.096 ± 0.0030
R-J-14	0.19 ± 0.032	<0.012	0.017 ± 0.0040	0.069 ± 0.0098
R-J-15	0.055 ± 0.013	<0.023	<0.019	0.070 ± 0.0070
R-J-16	0.14 ± 0.028	0.019 ± 0.0047	0.0012 ± 0.0035	0.11 ± 0.0038
R-J-17	0.12 ± 0.0085	0.022 ± 0.0032	0.012 ± 0.0037	0.0062 ± 0.0036
R-J-18	N.A.	<0.019	<0.032	0.081 ± 0.0090
R-P-1	0.030 ± 0.0089	<0.021	<0.016	0.210 ± 0.0093
R-P-2	0.043 ± 0.014	<0.021	<0.021	0.11 ± 0.0087
R-P-3	0.064 ± 0.014	<0.019	<0.021	0.063 ± 0.0076
R-P-4	0.050 ± 0.007	<0.011	<0.0095	0.050 ± 0.0035
S-J-1	0.22 ± 0.028	0.023 ± 0.0069	<0.026	0.021 ± 0.0059
S-J-2	0.31 ± 0.038	0.032 ± 0.0072	<0.032	0.14 ± 0.0085
S-J-3	0.32 ± 0.042	0.026 ± 0.0049	<0.027	0.14 ± 0.0098
S-J-4	0.22 ± 0.011	0.030 ± 0.0066	0.024 ± 0.0048	0.10 ± 0.0047
S-J-5	0.26 ± 0.012	0.022 ± 0.0033	0.020 ± 0.0043	0.10 ± 0.0047
S-J-6	0.17 ± 0.020	0.043 ± 0.0073	<0.020	0.072 ± 0.0070
S-J-7	0.28 ± 0.026	<0.014	0.023 ± 0.0057	0.075 ± 0.0042
S-O-1	0.12 ± 0.018	0.016 ± 0.0053	<0.013	0.015 ± 0.0043
S-O-2	0.17 ± 0.022	0.046 ± 0.010	<0.018	0.034 ± 0.0052
S-O-3	0.26 ± 0.035	0.029 ± 0.0067	0.024 ± 0.0051	0.019 ± 0.0030
S-O-4	0.24 ± 0.028	0.015 ± 0.0042	0.014 ± 0.0042	0.014 ± 0.0035
S-O-5	0.17 ± 0.010	0.018 ± 0.0043	0.018 ± 0.0027	0.016 ± 0.0023

[a] Not analyzed.
[b] 3σ values derived from counting statistics.
[c] 1σ values derived from counting statistics.

the concentration decreased as the sampling depth became deeper. On the other hand, the decrease in concentration with the sampling depth was not observed.

^{90}Sr is also an important radionuclide for radioactive monitoring research in the marine environment (Hirose et al., 1999; Ikeuchi et al., 1999; Ito et al., 2003; Pettersson et al., 1999; Povinec et al., 2003). It is well known that the behavior of Sr is similar to that of Ca. The main component of crab shell is chitin which bonds Ca strongly. Subsequently, the measurement of ^{90}Sr was carried out in shell sample of both crab species. The maximum concentration was 0.32 ± 0.042 Bq/kg-wet in sample S-J-4 (Table 3). Although marine alga has been used as bioindicator for ^{90}Sr in marine environment (Morita et al., 2010a; Yang et al., 2002), the concentration in shell sample was considerable higher than that in a marine alga species *Undaria pinnatifida* collected around Japan, ca. 0.030 Bq/kg-wet (Morita et al., 2010a). The concentration of ^{90}Sr in the shell samples of snow crabs was statistically higher than that of red queen crabs ($P < 0.01$). It was not found that the concentration of ^{90}Sr in both crab species increased during this investigation period (Fig. 7b). Consequently, new pollution source of ^{90}Sr did not also occur in these sampling regions during this period. Fig. 8b showed the relationship between the sampling depth and ^{90}Sr concentration in the shell samples. The concentration of ^{90}Sr in the sampling region J decreased with the sampling depth.

There was no correlation observed among radionuclides detected in the crab species, and even in the same radionuclide among the three tissues (Data not shown). These results indicate that radionuclides had different incorporation pathway and accumulation ability into each tissues form each other radionuclide. Stable element analyses were carried out on the same samples, and the concentrations of P, K, Ca, V, Mn, Fe, Ni, Co, Cu, Zn, Rb, Sr, Ag, Cd, Cs, Ba, Pb, Th, and U were determined (a part of data is shown in Table 4 and the other data not shown). In the hepatopancreas samples, Pu isotopes had a significant correlation with that of Pb ($P < 0.001$, $r = 0.907$ for $^{239+240}$Pu) (Fig. 9). In the shell samples, Pu isotopes showed a good correlation with U ($P < 0.01$, $r = 0.772$ for $^{239+240}$Pu). These results would also indicate that Pu isotopes in shell and hepatopancreas are incorporated into each tissue through different pathways.

It was supposed that the behavior of the radioisotopes and the stable elements was similar to each other. The concentration of ^{108m}Ag in the hepatopancreas samples showed a significant correlation with the stable Ag concentration in hepatopancreas ($P < 0.001$, $r = 0.809$) (Fig. 10a). The ^{137}Cs in the muscle and hepatopancreas samples indicated no correlation with the stable Cs concentration the respective samples. It was expected that the concentration of ^{90}Sr in the shell samples was proportional to Ca concentration in shell samples because Sr behaved as Ca did as described above. Nonetheless, there was no correlation between ^{90}Sr and Ca concentrations in the shell samples. The Ca concentration in the shell samples, however, had a significant correlation with stable Sr concentration ($P < 0.001$, $r = 0.905$) (Fig. 10b), while curiously there was no correlation between ^{90}Sr and stable Sr concentration in shell sample (Fig. 10c).

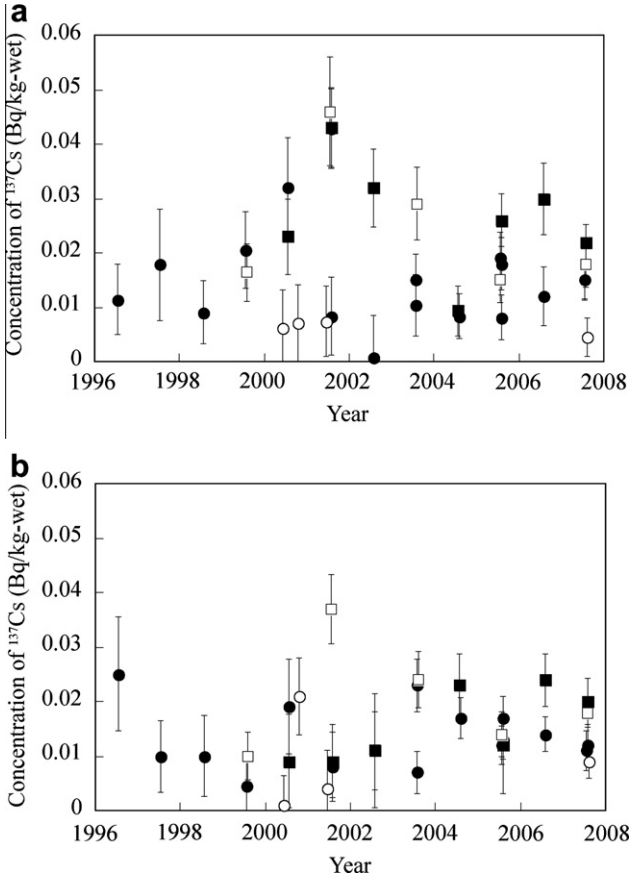

Fig. 5. Temporal variation in concentrations of ^{137}Cs in muscle (a) and in hepatopancreas (b) samples. Closed and open circles presented the data for the red queen crabs collected in the sampling region J and P, respectively. Closed and open squares did the data for the snow crabs collected in the sampling region J and O, respectively. Error bar shows 1σ value derived from counting statistics.

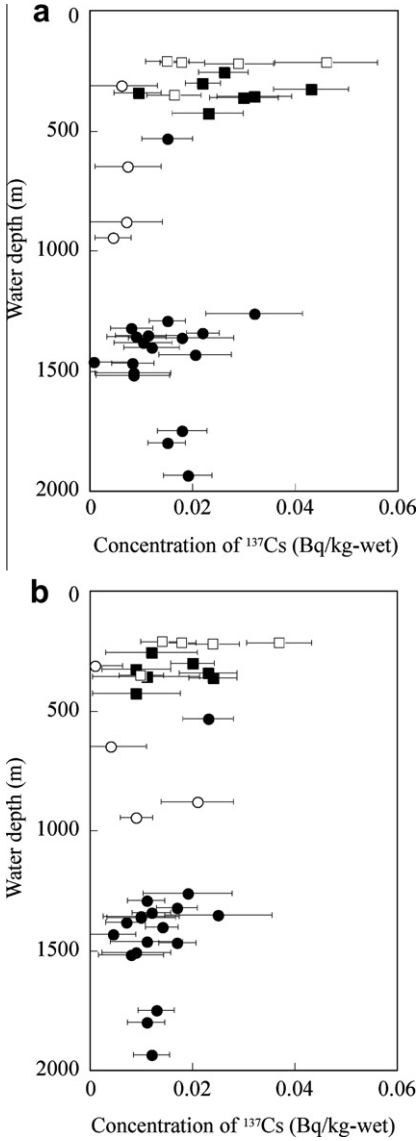

Fig. 6. Relationship between sampling depth and ^{137}Cs concentrations in the muscle samples (a) and hepatopancreas samples (b). Closed and open circles present the data for the red queen crabs collected in the sampling region J and P, respectively. Closed and open squares show the data for the snow crabs collected in the sampling region J and O, respectively. Error bars show 1σ value derived from counting statistics.

A large amount of radioactive wastes had been dumped in the northern part of the Japan Sea and in the north-western part of the Pacific Ocean off the Kamchatka Peninsula by the former USSR and Russia (IAEA, 1999; Yablokov, 2001). Although the contents of radionuclides in the dumped radioactive wastes are unclear, any radionuclides released from the dumped containers would increase the concentration of the radionuclides detected in this study. But there was no evidence of an elevated concentration of any of the radionuclides during this research period (Table 2 and 3). Additionally, radionuclides with a short half-life, e.g. ^{60}Co, ^{131}I, ^{134}Cs and ^{110m}Ag were not detected in this research. A change of the atom ratio of ^{240}Pu/^{239}Pu was also not observed during this period (Table 2, Fig. 2b). These results indicate a lack of the release of radionuclides from the radioactive wastes dumped by the former USSR and Russia and also the lack of any introduction of new other sources of radioactive pollution into the sampling regions in the period of this investigation. The radionuclides released from the dumped wastes might, however, have a serious impact on the marine environment in the future, as such; the current data will allow comparison as a baseline level. The concentrations of ^{137}Cs, ^{108m}Ag, ^{90}Sr, $^{239+240}$Pu detected in the two crab species were low (Table 2 and 3). As described above, other radionuclides with a short half-life and the increase in the concentration of radionuclides detected in this study were not observed during this research period. Therefore, the main pollution source of these radionuclides cam be considered to be the stratospheric (global) fall-out from the past nuclear weapon test conducted from 1945 to 1980 (Bowen et al., 1980; UNSCEAR, 2000). Other sources, e.g., the fallout from the Chernobyl reactor accident and the radionuclides released from nuclear power plants and nuclear fuel reprocessing plants have previously been shown to contribute little to the inventory of these radionuclides in the marine environment (Aoyama and Hirose 1995; Hirose et al., 1999; Ikeuchi et al. 1999; Pettersson et al., 1999).

The atom ratio of ^{240}Pu/^{239}Pu is a powerful tool to identify the Pu source in the environment (Kenna and Sayles, 2002; Olivier et al., 2004; Yamada and Zheng, 2008; Zheng and Yamada, 2004, 2005). The atom ratio in shell and hepatopancreas samples of the crab species in this study showed a wide range 0.21–0.26 with the average 0.24 ± 0.0085 (n = 22) and 0.16–0.32 with the average 0.23 ± 0.0066 (n = 33), respectively. These ratios were similar to those observed in common squid, seawater and sediments collected around the sampling region of the crab species in this study (Oikawa and Yamamoto, 2007; Yamada and Zheng, 2008; Zheng and Yamada, 2004, 2005). Most of these ratios were clearly higher

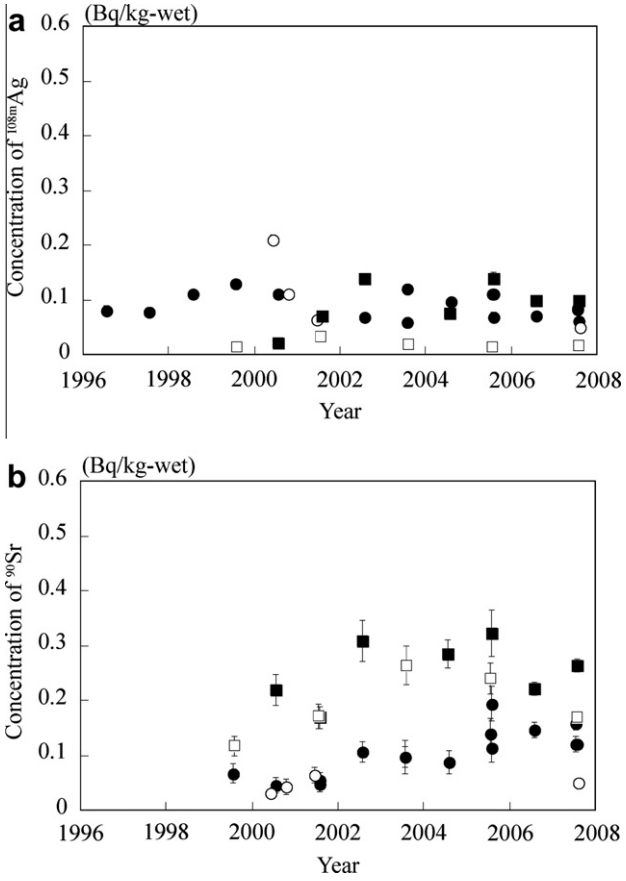

Fig. 7. Temporal variation in concentrations of ^{108m}Ag in hepatopancreas (a) and ^{90}Sr in shell (b) samples. Closed and open circles present the data for the red queen crabs collected in the sampling region J and P, respectively. Closed and open squares show the data for the snow crabs collected in the sampling region J and O, respectively. Error bars show 1σ value derived from counting statistics.

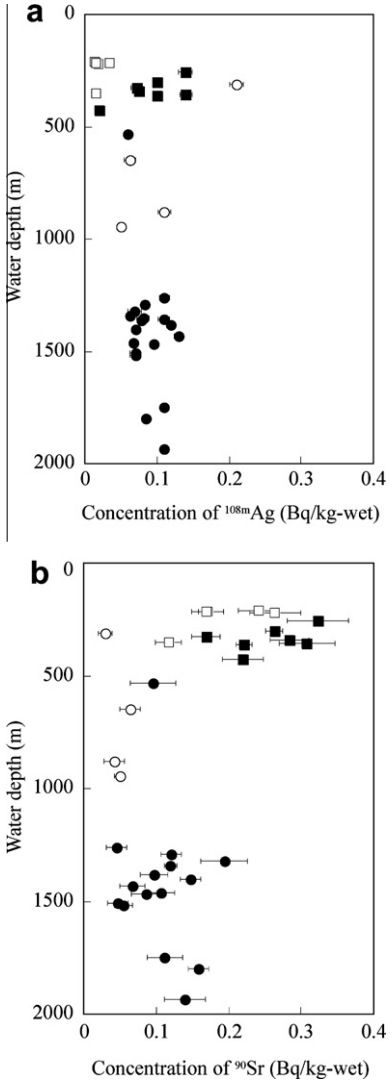

Fig. 8. Relationships between sampling depth and ^{108m}Ag concentrations in the hepatopancreas samples (a), and between the sampling depth and ^{90}Sr concentrations in the shell samples (b). Closed and open circles show the data for the red queen crabs collected in the sampling region J and P, respectively. Closed and open squares did the data for the snow crabs collected in the sampling region J and O, respectively. Error bars show 1σ value derived from counting statistics.

than the global fallout level, 0.176 ± 0.014 (Krey et al., 1976) and 0.180 ± 0.014 (Kelley et al., 1999) based on soil samples. A candidate for the source elevating the ^{240}Pu/^{239}Pu atom ratio in crab species was supposed to be the Chernobyl accident fallout, in which the atom ratio was ca. 0.41 (Muramatsu et al., 2000). However, the amount of Pu isotopes released into the sea around Japan from the Chernobyl accident fallout was small (Hirose et al., 2003) and therefore would contribute little to the increase in the ^{240}Pu/^{239}Pu atom ratio in the crab species. It has been proposed that Pu isotopes in seawater and marine sediment around Japan originated from a mixture of two sources of Pu isotopes, the global fallout and the Pacific Proving Grounds (PPG) tropospheric (closed-in) fall out (Zheng and Yamada, 2004). The ^{240}Pu/^{239}Pu atom ratio in airborne debris from the atmospheric nuclear weapon test, the IVY-Mike shot at Eniwetok Atoll in 1952 was 0.363 ± 0.004 (Diamond et al., 1960). Bikini Island soil also had an atom ratio of 0.338 ± 0.033 (Komura et al., 1984) and 0.306 ± 0.01 (Muramatsu et al., 2001). By using the two end-member mixing model based on the ^{240}Pu/^{239}Pu atom ratio of 0.18 for the global fallout and 0.30 or 0.363 for the PPG close-in fall out, the contribution of the PPG closed-in fallout to Pu isotopes in surface seawaters from the western North Pacific Ocean and Japan Sea was calculated to average 59% or 44% (Yamada and Zheng, 2008). Atmospheric weapon tests went through a moratorium between November 1958 and September 1961. Coral in the Bikini lagoon presenting that the atom ratio of the three years, 1954, 1956 and 1958 of intensive weapon tests at Bikini Island were 0.215, 0.225 and 0.238, respectively (Noshkin et al., 1975). The polar ice cores, which have information about the historical record of the isotopic composition of Pu fallout, also showed that the atom ratio decreased with time from 0.34 to 0.21 in the pre-moratorium period, in which nuclear weapon tests were dominated by the United States (Koide et al., 1985). After the moratorium period, the atom ratio in the nuclear weapon tests dominated by the USSR had a range, from 0.09 to 0.22. Therefore, it was difficult to confirm the atom ratio in the close-in fallout. The atom ratio, 0.272 ± 0.004, originated from the PPG closed-in fallout during the pre-moratorium period was observed in sediment core from the western Northwest Pacific margin (Zheng and Yamada, 2004). In conclusion, the Pu isotopes in the close-in fallout evidently contributed to the high atom ratio in the crab species though its proportion in the Pu isotopes in the crab species was unclear.

The sediment core from the western Northwest Pacific margin showing a high atom ratio, 0.272 ± 0.004, recorded that the atom ratio had already rapidly decreased to ca. 0.23 by 1963 from the release of global fallout, of which the peak deposition occurred in the mid-1960s (Hirose et al., 2003; Zheng and Yamada, 2004). This rapid decrease of the atom ratio in sediments would reflect the

Table 4
Concentrations of trace metals in the crab species.

Sample code	Shell				Muscle	Hepatopancreas			
	Ca (mg/g-wet)	Sr (mg/g-wet)	Pb (mg/kg-wet)	U (mg/kg-wet)	Cs (mg/kg-wet)	Ag (mg/kg-wet)	Cs (mg/kg-wet)	Pb (mg/kg-wet)	U (mg/kg-wet)
R-J-3	54.5	0.89	0.138	0.041	0.0042	5.33	0.0089	0.049	0.029
R-J-5	67.5	0.99	0.039	0.021	0.0035	4.43	0.0066	0.052	0.08
R-J-6	76.7	1.04	0.072	0.028	0.0029	3.54	0.0067	0.109	0.114
R-J-8	67.4	1.19	0.061	0.04	0.0037	6.7	0.0055	0.04	0.022
R-J-12	57.3	0.81	0.032	0.0058	0.0058	4.25	0.0057	0.086	0.035
R-J-13	90.7	1.39	0.043	0.0088	0.0034	5.18	0.0062	0.087	0.029
R-J-14	85.7	1.29	0.041	0.025	0.0026	4.21	0.0061	0.036	0.063
R-J-15	40.1	0.6	0.044	0.011	0.0031	7.51	0.0042	0.051	0.03
R-J-16	77.6	1.29	0.059	0.019	0.003	7.59	0.0031	0.033	0.041
R-P-2	55.7	0.92	0.094	0.022	0.0037	3.29	0.0045	0.095	0.168
R-P-3	53.8	0.77	0.058	0.0066	0.0028	2.78	0.0039	0.077	0.054
S-J-1	73.1	0.95	0.062	0.006	0.0028	0.52	0.004	0.063	0.08
S-J-2	57.2	0.97	0.187	0.047	0.0033	6.49	0.0035	0.234	0.078
S-J-6	60.1	0.88	0.065	0.0084	0.004	2.76	0.0041	0.135	0.041
S-O-1	41.5	0.59	0.057	0.011	0.0026	0.35	0.0066	0.044	0.046
S-O-2	71.4	0.93	0.107	0.01	0.0047	0.45	0.0045	0.058	0.067
S-O-4	91.0	1.20	0.079	0.016	0.0025	0.47	0.0057	0.043	0.057

decrease of that in seawater samples at the site. Also, the atom ratio in seawater samples at sampling site adjacent to Bikini Atoll had already showed the range from 0.20 to 0.25 in 1978 (Bertine et al., 1986). Pu isotopes with a higher atom ratio in the PPG close-in fallout had a different chemical form from Pu isotopes in the global fallout, and therefore was more rapidly removed from seawater samples than Pu isotopes in the global fallout (Buesseler, 1987). Consequently, the rapid decreases of the atom ratio observed in sediment and seawater would be caused by a large increase in the amount of global fallout and the removal of the Pu isotopes with higher atom ratio as over 0.30 from seawater. It was pointed out that the incorporation of Pu isotopes into marine organisms differed with their chemical forms (Noshkin and Gatrousis, 1974). Variability of the atom ratio was found in marine organism, seawater and sediment samples even near the sampling location. The influence on the incorporation of Pu isotopes into marine organisms, the behavior of Pu isotopes in seawater and the adsorption of Pu isotopes into marine organisms and sediments due to differences in chemical forms of Pu isotopes might produce the variability of the atom ratio in each sample.

Variability was found in the concentration of radionuclides detected in the crab species. In particular, the variability in the concentration of ^{90}Sr in the shell samples,^{137}Cs in the muscle samples, and ^{137}Cs, ^{108m}Ag and $^{239+240}$Pu in the hepatopancreas samples decreased with the sampling depth (Fig. 3b, 6a, 6b, 8a, 8b). Marine organisms uptake the radionuclides directly from the surrounding seawaters and through the food chain (Osterberg et al., 1964; Suzuki et al., 1979), in which the radioactive pollutant initially comes from the seawater. Thus, the concentration of radionuclides in marine organisms depends on the concentration in the surrounding seawater and the diet and on the quantity of diet. There was a little variability in concentrations of ^{137}Cs, ^{90}Sr and $^{239+240}$Pu in seawater at the sampling depths for the crab species (Hirose et al., 2002; Ito et al., 2003). The concentration of radionuclides in the dietary organisms was generally determined by their position in the food web. Therefore, the variability in the concentration of these radionuclides detected in the crab species would describe the variability of the composition and quantity of the diet. The large variability in the concentration of these radionuclides, ^{90}Sr in the shell samples,^{137}Cs in the muscle samples, and ^{137}Cs, ^{108m}Ag and $^{239+240}$Pu in the hepatopancreas samples, at shallower sampling depth would indicate the diversity of the diet sorts and the deflection of the supplied diet quantity, while the little variability in the crab species at deeper sampling depth would show the homogenization of the diet quantity and quality by the decomposition of the dietary organisms on the way of transportation from surface layer to the deeper layer. Variability was not found in the concentration of $^{239+240}$Pu in the shell samples with the increase of sampling depth (Fig. 3a). Stable element analyses indicated that the Pu isotope in the shell was incorporated through different pathway from that for the hepatopancreas. The Pu isotopes in the sea-bottom sediment might directly be adsorbed onto shell surface since the shell is in contact with the sea-bottom sediment.

The concentration of ^{90}Sr in the shell, ^{137}Cs in the muscle and ^{137}Cs and $^{239+240}$Pu in the hepatopancreas samples decreased as the sampling depth became deeper (Fig. 3b, 6a, 6b, 8b). The concentration of ^{90}Sr, ^{137}Cs and $^{239+240}$Pu in the seawater at depth below the sampling depth of the crab species generally decreased with water depth (Ito et al., 2003; Povinec et al., 2003). The concentration of radionuclides in marine organisms was determined by the concentration in the surrounding seawater and in the diet and on the quantity of diet as described above. In addition, the concentration of radionuclides in the diet would become constant at a low level with depth owing to the decomposition of diet on the way of transportation from surface layer to the deeper layer. As a

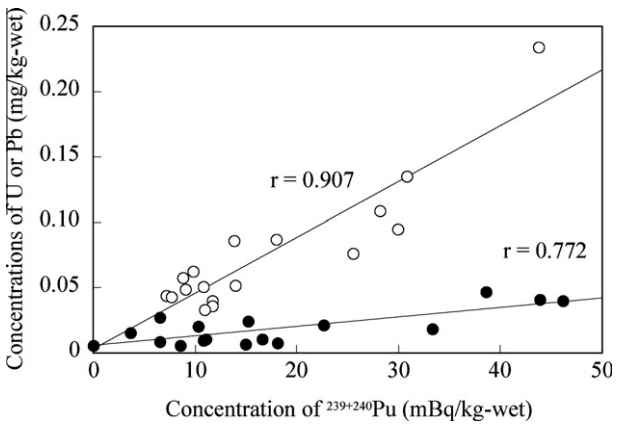

Fig. 9. Relationships between the concentrations of $^{239+240}$Pu and U in shell samples (closed circle) or Pb in hepatopancreas samples (open circle).

result, the concentration of ^{90}Sr in the shell, ^{137}Cs in the muscle and ^{137}Cs and $^{239+240}$Pu in the hepatopancreas samples decreased with the sampling depth.

There was no relationship between the sampling depth and $^{239+240}$Pu concentration in the shell and also ^{108m}Ag concentration in the hepatopancreas samples (Fig. 3b, 8a). As described above, the Pu isotopes in the shell might directly be taken in from the sea-bottom sediment. The sea-bottom sediment around the sampling region for the crab species includes the high concentration of $^{239+240}$Pu (Zheng and Yamada, 2005). Our results show that the vertical profile of ^{108m}Ag concentration in seawater below the sampling depth of the crab species was constant, although, as far as we know, there is no information about the concentrations of ^{108m}Ag and stable Ag in seawater around the sampling regions for the crab species. Stable element analysis showed that the concentration of ^{108m}Ag in the hepatopancreas samples had a significant correlation with the stable Ag concentration in the hepatopancreas samples (Fig. 10a). That indicated that ^{108m}Ag was well mixed with stable Ag in seawaters. Subsequently, the vertical profile of stable Ag concentration in seawater around the sampling regions for the crab species might be almost constant.

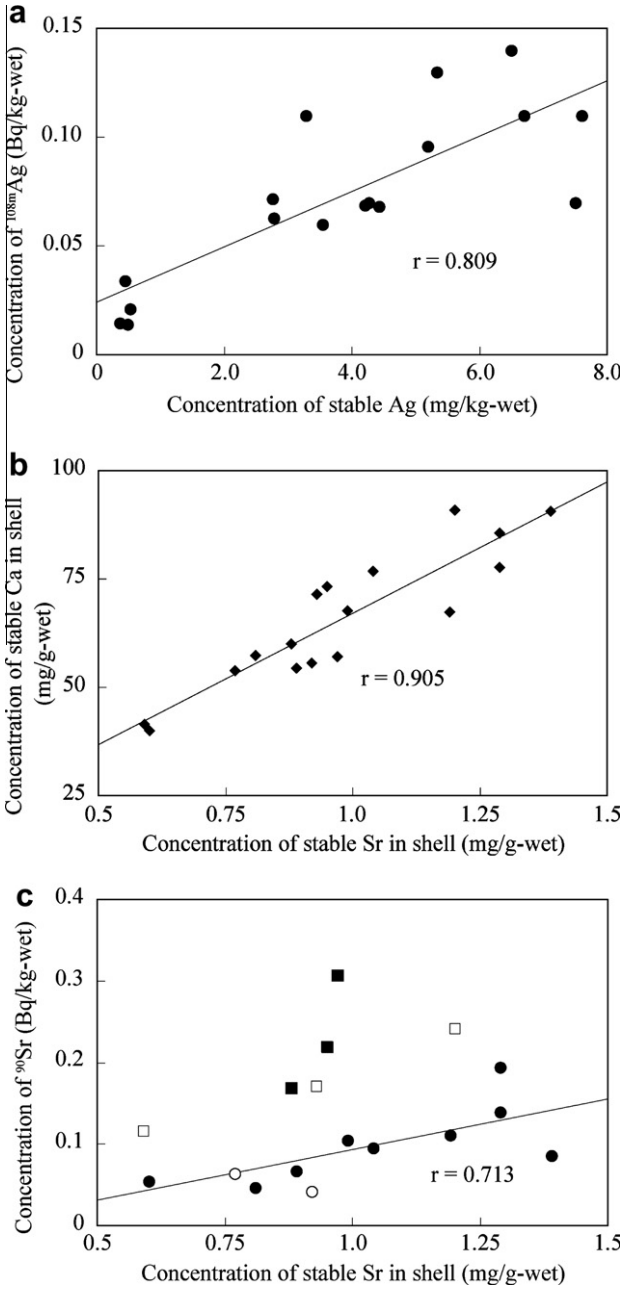

Fig. 10. Relationships between the concentrations of ^{108m}Ag and stable Ag in hepatopancreas samples (a), between the concentrations of stable Ca and stable Sr in shell samples (b) and between the concentrations of ^{90}Sr and stable Sr in shell samples (c). The regression line in (c) was for the concentrations of ^{90}Sr and stable Sr in samples collected in region J (closed circles).

^{137}Cs was not detected in all samples of red queen crab collected in the region P. There was no considerable difference in the diet condition between the sampling region J and P. For example, the concentration of ^{137}Cs in the mantle muscle of common squid collected in the sampling region P, 0.034 ± 0.0046 Bq/kg-wet, was similar to that in common squid collected in the sampling region J, 0.027 ± 0.0038 Bq/kg-wet, (NRIFS, 2009). Subsequently, no detection of ^{137}Cs in red queen crab collected in the region P is considered to have resulted from the low concentration of ^{137}Cs in the surrounding seawater (Ito et al., 2003; Povinec et al., 2003). The concentration of ^{137}Cs at deeper layer in the Japan Sea was slightly higher than that in Pacific Ocean because the formation of deep water in the Japan Sea occurs in the northwestern area of the Japan Sea by wintertime convection and leads to a flux of surface water, which generally has a slightly higher concentration of radionuclides than the deep water, into the deep layer (Ito et al., 2003; Miyao et al., 2000). On the other hand, the rattail deep-sea fish *Coryphaenoides yaquinae* collected at ca. 5600 m depth had the almost the same concentration of ^{137}Cs as the shallow fish did collected at the same site (Yoshida, 1999). This difference between fish and crab species could be brought about from the difference in their physiological differences.

Stable element analysis strangely indicated that no statistical correlation between ^{137}Cs and stable Cs concentrations in the muscle and hepatopancreas samples and no statistical correlation between ^{90}Sr and stable Sr concentrations in the shell sample (Fig. 10c). These would result from the differences in the vertical profiles between ^{137}Cs and stable Cs concentration and between ^{90}Sr and stable Sr concentration in seawater. The concentrations of the stable Cs and stable Sr in seawater were constant whereas the concentrations of ^{137}Cs and ^{90}Sr in seawater decreases with depth (Nozaki, 2001). The tendency of decrease was, however, different among the sampling region and the concentration of ^{90}Sr at mid and deep layer in the Japan Sea was slightly higher than that in the Pacific Ocean (Ito et al., 2003). Therefore, the comparison of ^{90}Sr with stable Sr concentrations in the shell samples of red queen crabs collected in the sampling region J showed a statistical correlation ($P < 0.05$, $r = 0.713$) (Fig. 10c).

Some kinds of marine organisms have been used as bioindicators for marine radioactive pollution. Fish species have been used as bioindicators for ^{137}Cs pollution because ^{137}Cs is accumulated in the muscle tissue (Morita et al., 2007; Young and Folsom, 1979). Although ^{90}Sr is known to accumulate in bone tissues, it is difficult to remove and collect only bones from fish sample. Subsequently, marine alga has been used as bioindicator for ^{90}Sr (Yang et al., 2002; Morita et al., 2010b) owing its ability of accumulation of ^{90}Sr and ease of collection. The concentration of ^{90}Sr in the marine alga *Undaria pinnatifida* was much higher than that in a fish species *Limanda herzensteini* and a mussel species *Mytilus coruscus* collected in the same area (Yang et al., 2002). Cephalopods including squid and octopus species have been frequently used as bioindicators for ^{60}Co, ^{108m}Ag and Pu isotopes (Morita et al., 2010a; Oikawa and Yamamoto, 2007; Umezu, 1994). These species, however, are not suitable for monitoring research for dumped radioactive wastes, which usually are discarded at the deep water. Crabs are benthic organisms, and the two crab species used in this study are deep water species. In particular, red queen crab distributes from ca. 500 to ca. 2700 m depth (Fujikura et al., 2000; Yosho

and Hayashi, 1994). Therefore, these crabs are expected to be rapidly influenced by the release of radionuclides from the dumped radioactive wastes. Additionally, the main radionuclides, ^{137}Cs, ^{90}Sr, ^{108m}Ag, and Pu isotopes, for the present monitoring research for radioactive pollution in the marine environment are able to be measured in these crab species. In addition, the concentration of ^{90}Sr in the shell sample of these crab species is considerably higher than that in marine algae, which are frequently used as bio-indicators for ^{90}Sr in the marine environment. Shell samples of crab species are also easily collected the same as marine alga. Consequently, these crabs are good indicator species for dumped radioactive wastes in the marine environment, and therefore will be increasingly utilized in the future.

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Reference

Aoyama, M., Hirose, K., 1995. The temporal and spatial variation of ^{137}Cs concentration in the Western North Pacific and its marginal seas during the period from 1979 to 1988. Journal of Environmental Radioactivity 29, 57–74.

Bertine, K.K., Chow, T.J., Koide, M., Goldberg, E.D., 1986. Plutonium isotopes in the environment: some existing problems and some new ocean results. Journal of Environmental Radioactivity 3, 189–201.

Bowen, V.T., Noshkin, V.E., Livingston, H.D., Volchok, H.L., 1980. Fallout radionuclides in the Pacific Ocean: vertical and horizontal distributions, largely from GEOSECS stations. Earth and Planetary Science Letters 49, 411–434.

Buesseler, K.O., 1987. The isotopic signature of fallout plutonium in the North Pacific. Journal of Environmental Radioactivity 36, 69–83.

Cooper, L.W., Kelley, J.M., Bond, L.A., Orlandini, K.A., Grebmeier, J.M., 2000. Sources of the transuranic elements plutonium and neptunium in arctic marine sediments. Marine Chemistry 69, 253–276.

Diamond, H., Fields, P.R., Stevens, C.S., Studier, M.H., Fried, S.M., Inghram, M.G., Hess, D.C., Pyle, G.L., Mech, J.F., Manning, W.M., Ghiorso, A., Thompson, S.G., Higgins, G.H., Seanorg, G.T., Browne, C.I., Smith, H.L., Spence, R.W., 1960. Heavy isotope abundances in 'Mike' thermonuclear device. Physical Review 119, 2000–2004.

Fujikura, K., Tsuchida, S., Hashimoto, J., 2000. Density estimate of the beni-zuwai crab *Chionoecetes japonicus*, by an in situ observation method. Fisheries Science 66, 1183–1185.

Hamilton, T.F., 2004. Linking legacies of the Cold War to arrival of anthropogenic radionuclides in the oceans through the 20th century. In: Livingston, H.D. (Ed.), Radioactivity in the Environment. Marine Radioactivity, 6. Elsevier Ltd, Oxford, pp. 23–78.

Hirose, K., Amano, H., Baxter, M.S., Chaykovskaya, E., Chumichev, V.B., Hong, G.H., Isogai, K., Kim, C.K., Kim, S.H., Miyao, T., Morimoto, T., Nikitin, A., Oda, K., Pettersson, H.B.L., Povinec, P.P., Seto, Y., Tkalin, A., Togawa, O., Veletova, N.K., 1999. Anthropogenic radionuclides in seawater in the East Sea/Japan Sea: results of the first-stage Japanese–Korean–Russian expedition. Journal of Environmental Radioactivity 43, 1–13.

Hirose, K., Miyao, T., Aoyama, M., Igarashi, Y., 2002. Plutonium isotopes in the Sea of Japan. Journal of Radioanalytical and Nuclear Chemistry 252, 293–299.

Hirose, K., Igarashi, Y., Aoyama, M., Kim, C.K., Kim, C.S., Chang, B.W., 2003. Recent trends of plutonium fallout observed in Japan: plutonium as a proxy for desertification. Journal of Environmental Monitoring 5, 302–307.

IAEA., 1999. Inventory of Radioactive Waste Disposal at Sea. IAEA-TECDOC No. 1105, IAEA, Viena.

Ikeuchi, Y., Amano, H., Aoyama, M., Berezhnov, V.I., Chaykovskaya, E., Chumichev, V.B., Chung, C.S., Gastaud, J., Hirose, K., Hong, G.H., Kim, S.H., Miyao, T., Morimoto, T., Nikitin, A., Oda, K., Pettersson, H.B.L., Povinec, P.P., Tkalin, A., Togawa, O., Veletova, N.K., 1999. Anthropogenic radionuclides in seawater of the Far Eastern Seas. Science of Total Environment 237 (238), 203–212.

Ito, T., Aramaki, T., Kitamura, T., Otosaka, S., Suzuki, T., Togawa, O., Kobayashi, T., Senjyu, T., Chaykovskaya, E.L., Karasev, E.V., Lishavskaya, T.S., Novichkov, V.P., Tkalin, A.V., Shcherbinin, A.F., Volkov, Y.N., 2003. Anthropogenic radionuclides in the Japan Sea: their distributions and transport processes. Journal of Environmental Radioactivity 68, 249–267.

Kasamatsu, F., Ishikawa, Y., 1997. Natural variation of radionuclide ^{137}Cs concentration in marine organisms with special reference to the effect of food habits and trophic level. Marine Ecology Progress Series 160, 109–120.

Kelley, J.M., Bond, L.A., Beasley, T.M., 1999. Global distribution of Pu isotopes and ^{237}Np. Science of total environment 237 (238), 483–500.

Kenna, T.C., Sayles, F.L., 2002. The distribution and history of nuclear weapons related contamination in sediments from the Ob River Siberia as determined by isotopic ratios of plutonium and neptunium. Journal of Environmental Radioactivity 60, 105–137.

Kim, C.K., Kim, C.S., Chang, B.U., Choi, S.W., Chung, C.S., Hong, G.H., Hirose, K., Pettersson, H.B.L., 2003. ^{240}Pu/^{239}Pu atom ratios in the bottom sediments of the NW Pacific Ocean. Journal of Radioanalytical and Nuclear Chemistry 258, 265–268.

Koide, M., Bertine, K.K., Chow, T.J., Goldberg, E.D., 1985. The ^{240}Pu/^{239}Pu ratio, a potential geochronometer. Earth and Planetary Science Letters 72, 1–8.

Komura, K., Sakanoue, M., Yamamoto, M., 1984. Determination of ^{240}Pu/^{239}Pu ratio in environment samples based on the measurement of Lx/α−ray activity ratio. Health Physics 46, 1213–1219.

Krey, P.W., Hardy, E.P., Pachucki, C., Rourke, F., Coluzza, J., Benson, W.K., 1976. Mass isotopic composition of global fallout plutonium in soil. In: Transuranium Nuclides in the environment. IAEA-SM-199–39. International Atomic Energy Agency, Vienna. pp. 671–678.

Miyao, T., Hirose, K., Aoyama, M., Igarashi, Y., 2000. Trace of the recent deep water formation in the Japan Sea deduced from historical ^{137}Cs data. Geophysical Research Letters 27, 3731–3734.

Molero, J., Sanchez-Cabeza, J.A., Merino, J., Mitchell, P.I., Vidal-Quadras, A., 1999. Impact of ^{134}Cs and ^{137}Cs from the Chernobyl reactor accident on the Spanish Mediterranean marine environment. Journal of Environmental Radioactivity 43, 357–370.

Morita, T., Fujimoto, K., Minamisako, Y., Yoshida, K., 2007. Detection of high concentrations of ^{137}Cs in walleye pollock collected in the Japan Sea. Marine Pollution Bulletin 54, 1287–1306.

Morita, T., Fujimoto, K., Kasai, H., Yamada, H., Nishiuchi, K., 2010a. Temporal variations of^{90} Sr and^{137} Cs concentrations and the^{137} Cs/^{90} Sr activity ratio in marine brown algae, *Undaria pinnatifida and Laminaria longissima*, collected in coastal areas of Japan. Journal of Environmental Monitoring 12, 1179–1186.

Morita, T., Otosaka, S., Fujimoto, K., Nishiuchi, K., Kimoto, K., Yamada, H., Kasai, H., Minakawa, M., Yoshida, K., 2010b. Detection and temporal variation of^{60} Co in the digestive glands of common octopus *Octopus vulgaris* in the East China Sea. Marine Pollution Bulletin 60, 1193–1199.

Muramatsu, Y., Ruhm, W., Yoshida, S., Tagami, K., Uchida, S., Wirth, E., 2000. Concentrations of ^{239}Pu and ^{240}Pu and their isotopic ratios determined by ICP-MS in soils collected from the Chernobyl 30-km zone. Environmental Science and Technology 34, 2913–2917.

Muramatsu, Y., Hamilton, T., Uchida, S., Tagami, K., Yoshida, S., Robison, W., 2001. Measurement of ^{239}Pu/^{240}Pu isotopic ratios in soils from the Marshall Islands using ICP-MS. Science of the Total Environment 278, 151–159.

Nagaya, Y., Suzuki, Y., Nakamura, K., 1990. 239, ^{240}Pu and ^{137}Cs concentrations in some marine organisms, mostly from the Ibaraki and Aomori coasts, Japan, 1987–1989. Nippon Suisan Gakkaishi 56, 1599–1604.

Noshkin, V.E., Gatrousis, C., 1974. Fallout Pu-240 and Pu-239 in Atlantic marine samples. Earth and Planetary Science Letters 22, 111–117.

Noshkin, V.E., Wong, K.M., Eagle, R., Gatrousis, C., 1975. Transuranics and other radionuclides in Bikin Lagoon: concentration data retrieved from aged coral sections. Limnology and Oceanography 20, 729–742.

Noshkin, V.E., Robison, W.L., Wong, K.M., Brunk, J.L., Eagle, R.J., Jones, H.E., 1997. Past and present levels of some radionuclides in fish from Bikini and Enewetak atolls. Health Physics 73, 49–65.

Nozaki, Y., 2001. Elemental distribution. In: Steele, J.H., Thorpe, S.A., Turekian, K.K. (Eds.), Encyclopedia of Ocean Sciences, vol. 2. Academic Press, pp. 840–845.

NRIFS., 2009. Annual Reports of Environmental Radioactivity Research for 2007. The Ministry of Agriculture, Forestry and Fisheries of Japan, pp.27–48 (in Japanese).

Oikawa, S., Yamamoto, M., 2007. 2007. Plutonium concentration and ^{240}Pu/^{239}Pu atomic ratio in liver of squid collected in the coastal sea area of Japan. Journal of Environmental Radioactivity 93, 170–180.

Olivier, S., Bajo, S., Fifield, L.K., Gäggeler, H.W., Papina, T., Santschi, P.H., Schotterer, U., Schwikowski, M., Wacker, L., 2004. Plutonium from global fallout recorded in an ice core from the Belukha glacier, Siberian Altai. Environmental Science and Technology 38, 6507–6512.

Osterberg, C., Pearcy, W.G., Curl, Jr.H., 1964. Radioactivity and its relationship to oceanic food chains. Journal of Marine Research 22, 2–12.

Pettersson, H.B.L., Amano, H., Berezhnov, V.I., Chaykovskaya, E., Chumichev, V.B., Chung, C.S., Gastaud, J., Hirose, K., Hong, G.H., Kim, C.K., Kim, S.H., Lee, S.H., Morimoto, T., Nikitin, A., Oda, K., Povinec, P.P., Suzuki, E., Tkalin, A., Togawa, O., Veletova, N.K., Volkov, Y., Yoshida, K., 1999. Anthropogenic radionuclides in sediments in the NW Pacific Ocean and its marginal seas: results of the 1994–1995 Japanese–Korean–Russian expeditions. Science of Total Environment 237 (238), 213–224.

Povinec, P.P., Livingston, H.D., Shima, S., Aoyama, M., Gastaud, J., Goroncy, I., Hirose, K., Kwong, L.H.N., Lee, S.H., Moriya, H., Mulsow, S., Oregioni, B., Pettersson, H., Togawa, O., 2003. IAEA'97 expedition to the NW Pacific Ocean-results of oceanographic and radionuclide investigations of the water column. Deep-sea Research part II 50, 2607–2637.

Science and Technology Agency of Japan (STA), 1983. Analytical method of radioactive strontium. Radioactivity Measurement Series No.2, (in Japanese).

Suzuki, Y., Nakamura, R., Ueda, T., 1973. Cesium-137 contamination of marine fishes from the coasts of Japan. Journal of Radiation Research 14, 382–391.

Suzuki, Y., Nakahara, R., Nakamura, R., Ueda, T., 1979. Roles of food and seawater in the accumulation of radionuclides by marine fish. Bulletin of the Japanese Society of Scientific Fisheries 45, 1409–1416.

Umezu, T., 1994. Oceanic distribution of ^{108m}Ag estimated using squid liver- a result of squid watch -. Water report 4, 9–11.

UNSCEAR., 2000. Source and Effect of Ionizing Radiation, United Nations Scientific Committee on the Effect of Atomic Radiation, United Nations, New York.

Watanabe, Y.M., Watanabe, S., Tsunogai, S., 1991. Tritium in the Japan Sea and the renewal time of the Japan Sea deep water. Marine Chemistry 34, 97–108.

Yablokov, A.V., 2001. Radioactive waste disposal in seas adjacent to the territory of the Russian Federation. Marne Pollution Bulletin 43, 8–18.

Yamada, M., Zheng, J., 2008. Determination of ^{240}Pu/^{239}Pu atom ratio in coastal surface seawaters from the western North Pacific Ocean and Japan Sea. Applied Radiation and Isotopes 66, 103–107.

Yang, H.S., Hwang, D.W., Lee, H.P., Kim, G., 2002. Distribution of ^{90}Sr in coastal seawater, sediments and organisms off two atomic power stations in Korea. Journal of Environmental Radioactivity 59, 105.

Yoshida, K., 1999. Biological transport of Cs-137 to deep sea bottom – using for case the rattail fish *Coryphaenoides yaquinae*. Aquabiology, 21, 210-218, 1999 (In Japanese with English abstract).

Yosho, I., Hayashi, I., 1994. The bathymetric distribution of *Chionoecetes opilio* and *C. Japonicus* (Majidae; Brachyura) in the western and northern areas of the Sea of Japan. Bulletin of the Japan Sea National Fisheries Research Institute 44, 59–71.

Young, D.R., Folsom, T.R., 1979. Cesium accumulation in muscle tissue of marine fishes. Health Physics 37, 703–706.

Zheng, J., Yamada, M., 2004. Sediment core record of global fallout and Bikini close-in fallout Pu in Sagami Bay, Western Northwest Pacific margin. Environmental Science and Technology 38, 3498–3504.

Zheng, J., Yamada, M., 2005. Vertical distributions of $^{239+240}$Pu activities and ^{240}Pu/^{239}Pu atom ratios in sediment cores: implications for the sources of Pu in the Japan Sea. Science of the Total Environment 340, 199–211.