

Plutonium Content in the Western North Pacific Waters

by

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Abstract

The content of plutonium in sea water collected in the western North Pacific off Japan was determined. Plutonium was analyzed by the α -ray spectrometric method after isolating it from a large volume of sea water by hydroxide coprecipitation and an anion exchanger. The plutonium content in sea water ranged from 1.8 to 22.6×10^{-15} g/l in the surface waters. Even in the intermediate or deep waters, comparatively high concentration of plutonium was determined. The ratio of $^{238}\text{Pu}/^{239}\text{Pu}$ ranged from 7 to 28%.

Introduction

The presence of a trace amount of plutonium in nature was first reported by SEABORG and his co-worker (1948). It was confirmed later by PEPPARD *et al.* (1951) and SEABORG (1951). Naturally occurring plutonium which is the product of neutron capture of ^{238}U was discovered in various uranium minerals and other minerals (LEVINE and SEABORG, 1951; CHERDYNTSEV, 1965) with a concentration ranging from 10^{-14} to 10^{-10} g/g. ^{239}Pu to uranium ratio in minerals is of the order of 10^{-12} to 10^{-11} . On the other hand, artificially produced plutonium has been released into the atmosphere through the nuclear weapon testings. Fallout plutonium is derived by neutron capture of ^{238}U in bombs or escape of fissile materials from the nuclear devices. At present, most of the plutonium in the global environment has been originated from the nuclear bombs.

It is well known that among plutonium isotopes produced in bombs, ^{239}Pu is the most abundant nuclide (SENTYURIN, 1967). The presence of a few percent of ^{238}Pu is also reported.

The contents of fallout plutonium in the air, rain water, soil, human organs and daily diet have been reported by many researchers, but, little is known on plutonium in the marine environment. Recently, FOLSOM *et al.* (1964) reported the plutonium concentration in sea water and marine organisms at the Pacific coast of North America. According to their results, the concentration in sea water was 5.6 to 49×10^{-15} g/l and the enrichment factor for zooplankton was as high as 2,600.

In the present paper the result of studies on the content in sea water in the

western North Pacific off Japan will be described.

Samples and method of analysis

The locations of water sampling are shown in Fig. 1. Surface water samples were collected during the eleventh cruise of the Japanese Expedition to Deep Sea (JEDS-XI, May to June, 1967) along 150°E and in the area of the Shikoku Basin on board the M. S. Ryufu-Maru-II (1,599 tons) which belongs to the Japan Meteorological Agency. Deep water samples were collected during the cruise of KT-67-16 (Aug. 1967) on board the M. S. Tansei-maru (250 tons) which belongs to the Ocean Research Institute, University of Tokyo at the station 33°05'N, 143°15'E. Coastal water samples were collected in the Sagami Bay on the pier of the Fisheries Research Laboratory, Kanagawa Prefecture.

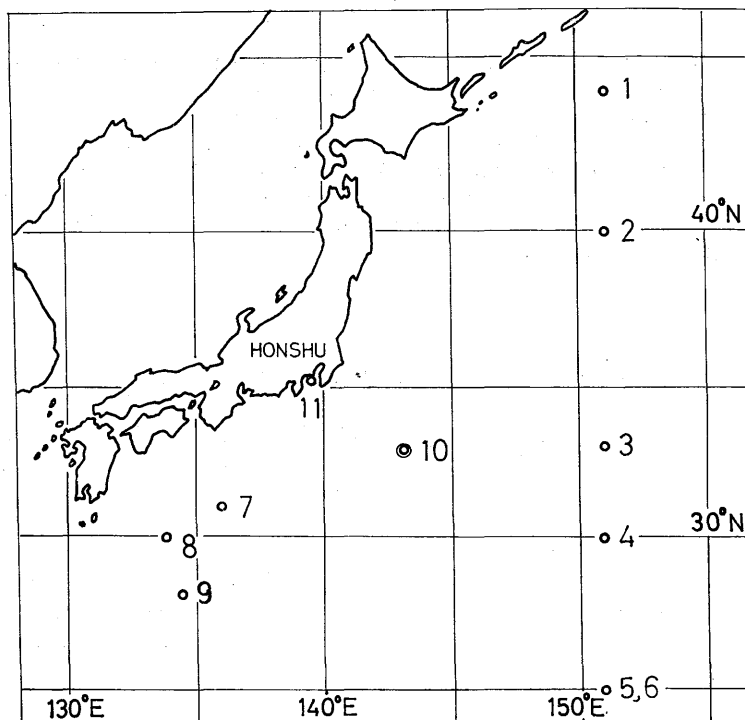


Fig. 1. Location of water sampling sites in the western North Pacific; double circle shows site of deep water sampling.

The water sample was collected on board into a 500 liter tank of polyvinyl chloride. Two grams of ferric ion as chloride was added to water immediately after collection to coprecipitate plutonium with ferric hydroxide. The precipitate was dissolved in hydrochloric acid and the solution was stored in a polyethylene bottle.

In the land laboratory, the solution was dried up on a hot plate, followed by digestion with concentrated perchloric acid to decompose organic matter. After drying up the solution, the residue was dissolved in 8 M HCl, and iron was removed

from the solution through a solvent extraction with iso-propyl ether leaving only a trace amount of iron in the solution.

After expelling the trace of ether, plutonium isotopes were coprecipitated again with a trace amount of iron in the solution by adding saturated sodium hydroxide solution. The precipitate was separated through centrifugation, washed with distilled water and finally dissolved in 7.5 M HNO_3 .

Separation of plutonium isotopes was carried out by an anion exchange resin (Dowex 1 \times 2 of NO_3 form, from 50 to 100 mesh in a column of 6 mm dia. and 10 cm long) at a flow rate of 1 ml per minute. The resin was washed successively with 8 M HNO_3 and 10 M HCl . Finally plutonium isotopes which were absorbed on the resin was eluted with 0.5 M HCl .

The effluent containing plutonium was dissolved in a mixed solution of 1 ml of 2 M HCl and 20 ml of ethyl alcohol. The solution was placed in an electroplating unit and plutonium isotopes were electroplated on a cathode of stainless steel disk (30 mm dia. and 0.2 mm thick) for two hours at a voltage of 10 volts and a current of 250 mA. Then the disk was removed, and heated with a flame.

Measurement of α -ray intensities of ^{239}Pu (5.15 Mev) and ^{238}Pu (5.49 Mev) was done with an α -ray spectrometer which consists of a Frisch screen grid ion chamber or a solid state detector coupled with a multichannel pulse height analyzer. Duplicate analyses by using ^{238}Pu tracer show that the chemical yield is $60\pm 10\%$.

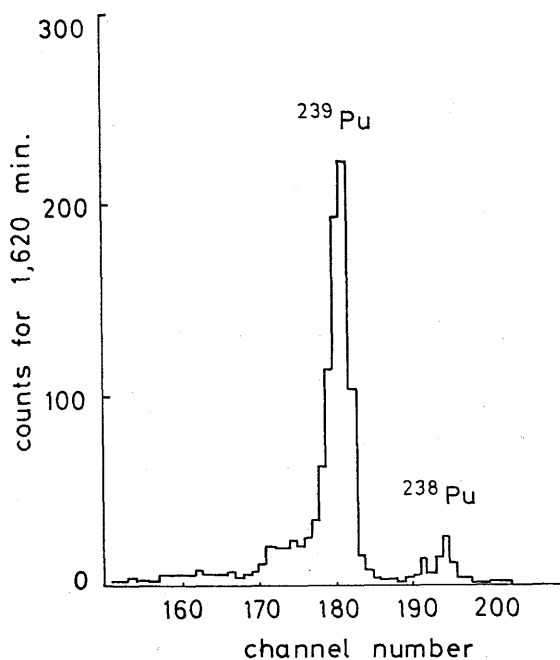


Fig. 2. α -spectrum of plutonium isotopes in sea water.
Location: 30°00' N, 150°56' E
Depth: 0 m

A typical α -spectrum of plutonium isotopes in sea water is given in Fig. 2.

Results and discussions

Table 1 gives the results of determination of plutonium concentration and the activity ratio of $^{238}\text{Pu}/^{239}\text{Pu}$ in sea waters of the western North Pacific off Japan and also in coastal waters at the Pacific coast of Japan.

Table 1. Plutonium content in the western North Pacific waters.

No.	Location		Depth	t	S	Sample	^{239}Pu	$A^{238}\text{Pu}/A^{239}\text{Pu}$
	N	E	(m)	($^{\circ}\text{C}$)	(%)	size (l)	($\times 10^{-15}$ g/l)	(%)
1	44 $^{\circ}$ 00'	150 $^{\circ}$ 58'	0	6.4	33.06	500	7.2	15
2	39 $^{\circ}$ 57'	151 $^{\circ}$ 02'	0	15.3	34.48	500	8.5	23
3	32 $^{\circ}$ 58'	151 $^{\circ}$ 06'	0	22.1	34.79	500	6.0	6.7
4	30 $^{\circ}$ 00'	150 $^{\circ}$ 56'	0	21.9	34.86	500	18.7	11.7
5	24 $^{\circ}$ 58'	151 $^{\circ}$ 00'	0	25.3	34.98	500	22.6	7.4
6	24 $^{\circ}$ 58'	151 $^{\circ}$ 00'	0	25.3	34.98	500	20.5	10.1
7	31 $^{\circ}$ 01'	136 $^{\circ}$ 19'	0	23.7	35.04	500	1.8	28
8	30 $^{\circ}$ 00'	133 $^{\circ}$ 40'	0	22.7	34.83	500	3.1	26
9	28 $^{\circ}$ 10'	134 $^{\circ}$ 33'	0	23.7	34.96	500	4.1	8.8
10	33 $^{\circ}$ 05'	143 $^{\circ}$ 15'	0	19.1	34.83	500	4.8	22
			1,000	3.3	34.43	500	4.9	10
			3,000	1.7	34.69	500	3.9	6.0
11	Jogashima pier		0	25.8	33.44	500	20	8.9
			0	25.8	33.44	500	10	7.8
			0	25.8	33.34	500	10	10

As to the plutonium concentration in surface waters, the result shows a rather wide range of variation from 1.8 to 22.6×10^{-15} g/l. Even in the water at 3,000 m depth in the western North Pacific, 3.9×10^{-15} g/l of plutonium was detected. Three samples of coastal water show contents from 10 to 20×10^{-15} g/l. The above values are of the same order of magnitude with those obtained by FOLSOM (1964) in the coastal waters of the eastern North Pacific.

With respect to the ratio of $^{238}\text{Pu}/^{239}\text{Pu}$, the observed ratio in sea water of 7 to 28% is in accordance with the ratio of fallout plutonium at New York in 1967.

Two samples of surface waters were taken simultaneously at the stations in the Shikoku Basin of the western North Pacific. In these samples ^{239}Pu , ^{137}Cs and ^{90}Sr were analyzed in the laboratory. The content of ^{137}Cs and ^{90}Sr are respectively 0.17 pCi/l and 0.18 pCi/l, and ^{239}Pu activities ranged from 0.1% to 1% of the ^{90}Sr activity.

The average ratio of ^{239}Pu to ^{90}Sr in fallout through the end of 1966 was 1.6% in Tokyo (MIYAKE, KATSURAGI, SUGIMURA, 1968). The reason for the lower ratio of ^{239}Pu to ^{90}Sr in sea water than that in land fallout calls for further study.

According to the result of study on ^{239}Pu fallout at Tokyo since 1958 through the end of 1966 (MIYAKE, KATURAGI, SUGIMURA, 1968), the integrated amount of ^{239}Pu fallout was 0.92 mCi/km^2 . Assuming that the same amount of ^{239}Pu has fallen on the ocean surface and has been mixed in the 100 m layer homogeneously, a concentration of about $1 \times 10^{-2} \text{ pCi/l}$ ($1.6 \times 10^{-13} \text{ g/l}$) of ^{239}Pu will be expected, which is much higher than the observed values. However as shown in Table 1, ^{239}Pu has been detected even in deep water to 3,000 m depth, and an average concentration of 2 to $10 \times 10^{-4} \text{ pCi/l}$ (1 to $6 \times 10^{-15} \text{ g/l}$) of ^{239}Pu will be anticipated taking the deep penetration of the nuclide into consideration. The calculated concentration of ^{239}Pu is of the same order of magnitude with the observed results in the western North Pacific.

With regard to the $^{239}\text{Pu}/\text{U}$ ratio in sea water, the ratio ranged from 2×10^{-10} to 9×10^{-9} , which is less than that in continental rocks and that in ground water observed in USSR (2×10^{-9} to 7×10^{-7} ; CHERDYNTSEV, 1965).

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北太平洋西部海水中のプルトニウム含量について

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1967年春から夏にかけて採取した北太平洋西部海水中のプルトニウム含量について報告する。プルトニウムは、500 l の海水から、水酸化鉄との共沈によって濃縮し、陰イオン交換法を用いて単離精製したのち、ステンレス板上に電着し、 α 線波高分分析法によって定量する。

表面水のプルトニウム含量は、 $1.8 \sim 22.6 \times 10^{-15} \text{ g/l}$ をしめし、中層水および深層水でも、かなり高い含量がみとめられた。

プルトニウムの同位体のうち主要なものは ^{239}Pu であるが、少量の ^{238}Pu がみとめられ、 $^{238}\text{Pu}/^{239}\text{Pu}$ の放射能比は $7 \sim 28\%$ をしめた。

海水中の $^{239}\text{Pu}/\text{U}$ 比は $2 \times 10^{-10} \sim 9 \times 10^{-9}$ であり、火山岩やソ連の陸水中の値 ($2 \times 10^{-9} \sim 7 \times 10^{-7}$) よりも低い。