

Artificial Radionuclides in the Western North Pacific: A Review

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Abstract. Artificial radionuclides in the marine environment pose a significant concern along various political, health and environmental aspects since they were introduced as a by-product from the nuclear weapon testing, particularly in the northwestern Pacific Ocean. While radiological concern is confined only in special cases, the introduction of artificial radionuclides has been proven to be very useful tracers for the ocean processes. Various input of artificial radionuclides to the Pacific Ocean were compiled. Time series variation of the artificial radionuclides in the surface waters and in the water column was described. Finally, retrospective studies in the artificial radionuclide concentrations in the surface water was proposed using the dated corals taken from the northwest Pacific Proper away from the influence of coastal ocean.

Keywords: artificial radionuclides, input, temporal change, behavior, coral records

1. INTRODUCTION

The present sources of anthropogenic radionuclides in the marine environment, consisting of stratospheric and tropospheric fallout from atmospheric nuclear testing, accidental releases from civilian and military nuclear facilities including satellites, radioactive waste dumping in the Pacific Ocean are reviewed. The total activity of anthropogenic radionuclides in the world's ocean from fallout from nuclear weapons testing has been estimated to be more than 10^5 PBq (1 PBq = 10^{15} Bq), with tritium contributing more than 99% to this inventory. The single largest contribution to radioactivity in the marine environment from accidental releases from land based nuclear installations has come from the accident at the Chernobyl nuclear power station in April 1986.

We have chosen ^{90}Sr , ^{137}Cs and Pu isotopes as representative of anthropogenic radionuclides to study their distribution and behavior in the Pacific Ocean. On a global scale, ^{137}Cs and ^{210}Po , the most representative of anthropogenic (^{137}Cs) and natural (^{210}Po) marine radioactivity, the average annual individual doses from ingestion of marine food was estimated to be $0.03 \mu\text{Sv}$ from ^{137}Cs and $9 \mu\text{Sv}$ from ^{210}Po . The annual dose of ^{137}Cs for a hypothetical critical group consuming 100 kg of fish and 10 kg of shellfish per year would be $3 \mu\text{Sv}$, while the

contribution from ^{210}Po would be $160 \mu\text{Sv}$ for the year of 2000. These values are well below the accepted value for the public of 1 mSv (Livingston and Povinec, 2000).

Among the world oceans, the depositional situation in the Pacific is more complex than other oceans due to the presence of proving grounds in the tropical Western Pacific. The 68 near surface nuclear tests conducted at Bikini and Enewetak produced not only local fallout but regionally deposited tropospheric fallout, which led to widespread deposition to the Western North Pacific on a scale similar to that from global fallout. The Western Pacific ocean is, therefore, unique in the history of atmospheric nuclear weapons test fallout in that it is the one area of the globe where fallout was substantially enhanced over the patterns globally observed (Livingston and Povinec, 2002). Substantial excesses of measured inventories of ^{137}Cs , ^{90}Sr , and $^{239+240}\text{Pu}$ were found at many locations in the Western North Pacific compared to estimated delivery from global fallout. In this review, we have made an attempt to compile the various sources of artificial radionuclides into the Pacific Ocean and their current levels in the water column. Utility of artificial radionuclides as geochemical tracers is partly reviewed, and retrospective study on the level of artificial radionuclides in the northwestern Pacific Basin is proposed briefly.

2. INPUT

The three main sources of man-made radionuclides are nuclear weapons testing in the atmosphere, the Chernobyl accident, and waterborne discharges from nuclear reprocessing plants in the world ocean (Aarkrog, 2003). Other sources including sea-dumping of nuclear wastes, routine discharges from nuclear power plants, sunken nuclear submarines, satellite failures, lost nuclear weapons and the use of radioisotopes in medicine, industry and science, have had orders of magnitude lower contribution to the contamination of the World Ocean. Previously, Hamilton *et al.* (1996) assessed the input of artificial radionuclides and established a baseline dataset on the current level of key artificial radionuclides in the Pacific Ocean.

2.1 Atmospheric nuclear testing

Atmospheric nuclear testing resulted in fallout of nuclear material on the surface of the Earth and usually classified according to their detonation types (Buesseler and Sholkovitz, 1987). Global or stratospheric fallout refers to very small sized fallout particles ($\sim 1 \mu\text{m}$ or less in diameter) which received sufficient height to become entrained in the stratosphere. The fallout debris has a residence time in the atmosphere on the order of one year or more. This is the most abundant type of fallout. Both large and small yield nuclear tests conducted at high altitudes and large yield surface-based nuclear tests have produced considerable stratospheric fallout. The stratospheric fallout tends to concentrate over the mid-latitude in the northern hemisphere and western Pacific due to the climatology of stratospheric air intrusions into the troposphere (Waugh and Polvani, 2000), as

discussed by Hirose *et al.* (2001). Tropospheric fallout refers to fallout debris which is retained within the troposphere either as larger fallout particles, which are deposited rapidly within the vicinity of the nuclear testing site (so-called close-in fallout), or smaller fallout particles which do not reach stratospheric altitudes and are deposited within the hemisphere of injection in the days to weeks following a particular testing event (so-called intermediate fallout). Any nuclear test that is conducted at or just above the ground (i.e. surface based) will generate a substantial fraction of tropospheric fallout. Recently Aarkrog (2003) estimated input of anthropogenic radionuclides into the world ocean based on the UNSCEAR (2000). According to UNSCEAR (2000) the total fission yield of atmospheric nuclear test explosions is 189 Mt (TNT). Among total explosions, 29 Mt were deposited as local fallout close to the test sites. Hence, 160 Mt became global fallout. As nearly all local fallout (28 Mt fission) deposited at the US test sites at Marshall Islands in the Pacific Ocean (0–30°N latitude belt), the local fallout debris partly imbedded in minerals (corals) in the region. The production estimate of ^{90}Sr is 3.88 PBq Mt⁻¹. In the case of $^{239,240}\text{Pu}$ and ^{137}Cs , the production estimates were calculated assuming the activity ratios of $^{239,240}\text{Pu}/^{90}\text{Sr}$ and $^{137}\text{Cs}/^{90}\text{Sr}$ ratios to be 0.0175 and 1.53, respectively (Aarkrog, 2003) and the values are given in Table 1. However, a contribution of the plutonium deposited by the French atmospheric nuclear tests in the sediments of Mururoa and Fangataufa was observed to be confined to the adjacent waters only within the 22 km from the atolls in 1993–1996 after French tests were conducted from 1966 to 1974 (Chiappini *et al.*, 1999).

2.2 Satellite burn up in the air

Nuclear energy sources are used in some types of satellites for generation of

Table 1. Inputs (PBq) from weapons testing in the Pacific Ocean (year 2000).

Latitude	Stratospheric fallout (global fallout)			Tropospheric fallout (local or close in fallout)		
	^{90}Sr	^{137}Cs	$^{239+240}\text{Pu}^*$	$^{90}\text{Sr}^*$	$^{137}\text{Cs}^*$	$^{239+240}\text{Pu}^*$
90–60°N	1.3	2.1	0.023			
60–30°N	71.6	114.6	1.253			
30–0°N	65.8	105.3	1.152	113	171.8	3.8~6.3 ⁺ (1.978)
0–30°S	26.6	42.6	0.466			
30–60°S	25.8	41.3	0.452			
60–90°S	3	4.8	0.053			
Total	194.1	310.6	3.397	113	171.8	3.8~6.3 ⁺ (1.978)
Grand total	307.1	482.5	7.2 ⁺ ~9.7 ⁺ (5.374)			

Note: $^{239+240}\text{Pu}^*$ and $^{137}\text{Cs}^*$ inputs derived from weapons testing were calculated assuming $^{239+240}\text{Pu}/^{90}\text{Sr}$ and $^{137}\text{Cs}/^{90}\text{Sr}$ ratios to be 0.0175 and 1.53, respectively (Aarkrog, 2003). Values with ⁺ are based on the soil profiles (Nakano and Povinec, 2003) and values in parenthesis are based on the fission yield.

heat or electricity. More than 30 nuclear powdered satellites in the Cosmos series of the Former Soviet Union were launched. At the end of the operation time, the normal procedure is to boost the satellite to a higher orbit, with a lifetime of at least 500 years, to allow for the decay of the fission products before the satellite with its nuclear reactor re-enters the earth's atmosphere and burns up (IAEA, 2001). US navigational satellite "Transit 5BN-3" with a SNAP-9A radioisotope generator containing 630 TBq of ^{238}Pu metal failed to achieve orbit and re-entered the atmosphere at 120 km altitude and burned up over the West Indian Ocean north of Madagascar in 1964. The nuclear fuel was vaporized during re-entry and was dispersed worldwide. The North Pacific appears to have received 0.0345 PBq of ^{238}Pu as of year 2000, calculated from Aarkrog (1988). In 1970, SNAP-27 radioisotope generator containing 1.63 PBq of ^{238}Pu from the Apollo 13 landed as intact as designed in the deep ocean south (water depth 6000 m) of Fiji Islands in the vicinity of the Tonga Trench. However, no release of nuclear fuel was evidenced (IAEA, 2001).

2.3 *Civilian nuclear facility accident*

The world's worst nuclear power accident occurred at Chernobyl (51°23' N, 30°06' E) in the Former USSR (now Ukraine). Some 8 of the 140 tons of fuel, which contained plutonium and other highly radioactive materials (fission products), were ejected from the reactor along with a portion of the graphite moderator (<http://www.chernobyl.co.uk>). Chernobyl accident released various fission and activated products, and uranium and plutonium into the air. Among the long-lived ones, ^{137}Cs , and ^{134}Cs (3.7×10^{16} and 1.9×10^{16} Bq, respectively) were released from Chernobyl accident and accounted for 5.1×10^{-3} of the total release of caesium isotopes released in the Northern Hemisphere stratosphere and among which 0.12–0.42 PBq was deposited over the East Sea (Sea of Japan, surface area 1.008×10^6 km²) (Miyao *et al.*, 1998).

2.4 *Nuclear reprocessing plants*

Nuclear reprocessing is carried out to recover U and Pu from spent fuel for reuse in reactors. The major reprocessing plants are in the United Kingdom (Sellafield), France (Cap de La Hague), and Japan (Tokai-mura since 1977 and Rokhasho-mura since 2005 (<http://www.nsnet.gr.jp>)). Since most discharges were made from the European plants, the discharge of radionuclides from the nuclear reprocessing is apparently negligible in the Pacific Ocean, which was evidenced by the low ^{129}I levels in the North Pacific Ocean and Arctic Sea (Cooper *et al.*, 2001).

2.5 *Ocean dumping of nuclear wastes*

The use of nuclear energy for the generation of electric power, as well as the applications of radioisotopes in research, industry and medicine, are unavoidably associated with the production of radioactive wastes. The treatment and ultimate disposal of the low level radioactive solid wastes present a special problem due

Table 2. Radioactive waste dumping by Japan, Republic of Korea, New Zealand, USA in the Pacific Ocean.

Country	Date	Coordinate		Depth range (m)	Container number	Radioactivity (GBq)
		Latitude	Longitude			
USA	1946–1962	21°28' N–52°25' N	119°35' W–174°52' W	896–5487	163	554247
New Zealand	1954–1976	42°13' S–42°15' S	175°00' W–176°10' E	2600–2834	39	1039
Republic of Korea	1968–1972	37°20' N	130°44' E	2192	115	—
Japan	1955–1969	34°34' N–34°55' N	139°25' E–139°56' E	2400–2600	3031	15441

to the volumes involved. The search for safe and economical disposal methods for this kind of waste has thus always been of considerable importance to countries with nuclear programmes. Disposal into selected areas of the deep oceans, under carefully controlled conditions, soon appeared as a suitable solution for some types of packaged low and medium-level radioactive waste. For the Pacific Ocean, sea dumping operations were carried out on national basis, from 1948 and 1976 by Japan, New Zealand, Republic of Korea, and USA (Table 2, IAEA, 1991). The sea dumping of radioactive wastes of Former Soviet Union and Russian Federation from 1962 to 1992 was reported after 1992 contradicting her earlier report to the London Convention 1972 in 1988 (Table 3, IAEA, 1999).

2.5.1 USA, Japan, New Zealand, and Republic of Korea

The radioactive wastes were usually contained in metal drums of 200 liters lined with concrete or bitumen. The radioactive wastes were of mainly solid or solidified low level radioactive wastes totaling 0.89 PBq in the West Pacific and 0.55 PBq in the Northeast Pacific (Table 2). Some monolithic blocks were also dumped and sometimes a polymer matrix was used. Beta-gamma emitters represent more than 98% of the total radioactivity of the waste and tritium alone represents one third of the total radioactive material disposed of at the North-East Atlantic sites. They comprise fission and activation products such as ^{90}Sr , ^{137}Cs , ^{55}Fe , ^{58}Co , ^{60}Co , ^{125}I and ^{14}C . The wastes also contain low quantities of alpha-emitting nuclides with plutonium and americium isotopes representing 96% of the alpha emitters present (IAEA, 1991).

2.5.2 Former Soviet Union and Russian Federation

The former Soviet Union disposed of radioactive wastes in the Far Eastern Seas totaling 456 and 418 TBq for liquid and solid wastes, respectively (Table 3). Unlike the Arctic, no reactors containing fuel were dumped there. The Joint Japanese-Korean-Russian expeditions carried out during 1994 and 1995 took samples of seawater, bottom sediments and biota from dump sites and from reference sites. The results showed that the concentrations of ^{90}Sr , ^{137}Cs , ^{238}Pu and $^{239+240}\text{Pu}$ in the Far Eastern Seas were low and were predominantly due to global fallout (Hirose *et al.*, 1999; Petterson *et al.*, 1999; Ikeuchi *et al.*, 1999).

2.5.3 International regulations on the sea dumping of radioactive wastes

Radioactive wastes dumping operations at sea raised political, scientific and technological questions. Templeton (1997) provided detailed review on the

Table 3. Radioactive wastes disposed of in the Pacific by Former Soviet Union and Russian Federation. Solid wastes were disposed only in containers except large objects (Danilyan and Vysotsky, 1995; IAEA, 1999).

Region	Coordinate	Depth (m)	Date	LNW (TBq)	SNW (TBq)
1 East Sea (Sea of Japan)	41–42°N 133°10' E–134°30' E	3250–3700		0.056	
2 East Sea (Sea of Japan)	39°30' –41°10' N 131°10' –134°30' E	2900–3300		0.033	
3 Off Kamchatka	51°20' –53°N 146°40' –148°10' E	ca. 2300		0.004	
4 Off Kamchatka	48°–50°N 161°35' –162°45' E	ca. 5500		0.007	
5 East Sea (Sea of Japan)	42°17' –42°26' N 131°37' –132°20' E	1100–1500	1966–1992	4.325	
6 East Sea (Sea of Japan)	41°45' –41°55' N 131°47' –132°13' E	1900–3300	1986–1987 (1993)	18.084 (0.0139)	119.399
7 Off Kamchatka	52°28' –52°40' N 159°02' –159°12' E	1400–1500	1966–1992	13.001	
8 Off Kamchatka	52°28' –52°34' N 159°02' –159°11' E	2000–2570			110.692
9 East Sea (Sea of Japan)	41°36' –41°46' N 133°22' –134°42' E	3300–3400	1974–1992	401.087	82.144
10 East Sea (Sea of Japan)	40°10' –41°10' N 131°15' –131°35' E	2900–3300	1974–1984	19.816	105.385
Total				456	418

development of this issue. The first of the international meeting was held in 1958 at the United Nations Conference on the Law of the Sea. Subsequently, the International Atomic Energy Agency (IAEA) recommended measures for ensuring that disposal of waste into the sea would not result in unacceptable hazards to man (references in Templeton, 1997). Since that time, knowledge of oceanographic processes has improved markedly, providing better understanding of the physical transport process and of the pathways by which radionuclides are transported from marine dumping and disposal sites back to man. Finally, radioecology has developed to the stage where predictions of radionuclides cycling pathways and rates are possible. The IAEA has published many documents in a series “Safety and Technical Documents” covering relevant areas such as oceanographic models, bioaccumulation factors, sediment distribution coefficients, and effects of ionizing radiation on organisms. The number of international and regional conventions has also increased over the same time period. One resolution of the United Nations (UN) Conference on the Human Environment, held in Stockholm in 1972, provided the stimulus for the formulation of the Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter (London

Convention 1972) which entered into force in 1975. The Convention binds contracting parties to take all practical steps to prevent the pollution of the sea by the dumping of waste and other matter that is liable to create hazards to human health, to harm living resources and marine life, to damage amenities or to interfere with other legitimate uses of the sea. The London Convention 1972 entrusts the IAEA with specific responsibilities for dumping of radioactive wastes at sea, and for making recommendations for radiological dose assessment and surveillance. In 1974, the IAEA established a provisional definition of high level wastes or other radioactive matter unsuitable for dumping at sea, and for dumping other material at sea. The IAEA recommendations were adopted by the London Convention 1972 in 1976, revised in 1978 and again revised in 1985. However, dumping operations was suspended in 1983 as a consequence of a voluntary moratorium decided by Contracting Parties to the London Convention. Since 1983 a moratorium on the dumping of low-level radioactive wastes has been in place pending the completion of scientific and technical studies on the wider political, legal, economic and social aspects of radioactive waste dumping (e.g. CRES, 1996). Following a completion of these studies, the parties agreed in 1993 to amend the Annexes I and II to the London Convention 1972 to ban the dumping of all radioactive wastes for a 25-year period. The resolution also requires to complete a scientific study relating all radioactive wastes and other radioactive matter other than high level wastes or matter at each 25-year interval. This legally binding prohibition entered into force on February 1994. However, Russia, the principal recent “dumper” of radioactive waste into the oceans, has not agreed to accept the nuclear waste dumping ban until now.

Ongoing military and civilian uses of nuclear materials will continue to result in the production of nuclear wastes to be disposed of safely in the environment. Therefore it is extremely important to make periodic detailed comparative analyses of land- and ocean-based operations, including models for release rates and process of radionuclide transport, concentration and dispersion. This effort will require the international cooperation between scientists, policy analysts, legal and environmental organizations (Vartanov and Hollister, 1997).

However, natural radionuclides are also present in all materials, including natural and inert materials, which can also contain artificial radionuclides such as fallout due to past atmospheric nuclear testing. Therefore, the Contracting Parties to the London Convention 1972 recognized the need to develop definitions and guidelines so that candidate materials (those wastes or other matter not otherwise prohibited from disposal at sea in accordance with Annex I to the Convention) containing less than *de minimis* levels of specific activity, can be regarded as “non-radioactive” and may be disposed of at sea subject to the other provisions of the Convention. In 1997, Contracting Parties to the London Convention 1972 agreed to request the IAEA to develop further the concept of *de minimis* levels and, in particular, to “provide guidance for making judgment on whether materials planned to be dumped could be exempted from radiological control or whether a specific assessment was needed”. The IAEA provided it advice in this regard (IAEA, 2003).

Table 4. Accidents at sea resulting in actual or potential release to the Pacific Ocean (IAEA, 2001).

Country	Date	Region	Depth	Content
Russian Federation	1997	Sea of Okhotsk		RTG power supply, ⁹⁰ Sr-sealed source, 1.3 PBq
Soviet Union	1987	Sea of Okhotsk		RTG power supply, ⁹⁰ Sr-sealed source, 25.3 PBq
USA	1970	South Pacific Tonga	6000	SNAP-27 generator, 1.63 TBq
USA	1965	South of Kyushu	4800	Skyhawk Jet A-4E1 nuclear weapon
USA	1962	Johnston Island		Nuclear test device
Indonesia		Indonesian waters	300–4058	²⁴¹ Am, ¹³⁷ Cs, 4077.9 GBq, abandoned by cementing
Malaysia	1990–95	South China Sea	1800–3132	¹³⁷ Cs, ²⁴¹ Am, 333 Gbq, abandoned by cementing in drill well
USA	1982–88	Pacific Coast, Offshore Australian		²²⁶ Ra, ²⁴¹ Am, ¹³⁷ Cs, 703 GBq

2.6 Accidents and losses at sea involving radioactive material

Data on the accidents and losses at sea involving radioactive material was compiled by IAEA. In 1989, pursuant to the recommendations of the London Convention 1972, the IAEA started to gather information on accidents and losses at sea. Seven possible sources of radioactive material entering the marine environment as a result of accidents and losses have been identified. These are nuclear powered military surface or underwater vessels, nuclear weapons and military vessels capable of carrying such weapons, nuclear powered civilian ships, nuclear energy sources used in spacecraft, and satellites and in the deep sea as acoustic signal transmitters, radioisotope thermonuclear generators (RTG) used, for instance, to supply power to lighthouse, cargoes of nuclear material in transit, and sealed radiation sources. The inventory of accidents and losses involving radioactive material in the Pacific Ocean (Table 4) was taken from IAEA (2001). IAEA (2001) further listed the losses of sealed sources resulting in actual or potential release to the marine environment. A radioactive source designed in such a form that the probability of dispersion of its radioactive contents is extremely low. Sealed sources may be used, for example in teletherapy and brachytherapy and in scientific devices, as well as in a number of industrial and other medical applications. In the context of this report most of the sealed sources have been used in offshore drilling and logging.

3. CURRENT LEVEL OF ARTIFICIAL RADIONUCLIDES IN THE NORTH PACIFIC OCEAN

IAEA and Japan have recently established an extensive database on the marine radioactivity, GLOMARD and HAM, respectively, and IAEA has analyzed the database through the Coordinated Research Programme (CRP) on Worldwide Marine Radioactivity Studies (IAEA, 2001). Current level of artificial radionuclides in the North Pacific Ocean is largely derived from IAEA (2001) and Hirose and Aoyama (2003).

Table 5. The effective half-life and estimated concentration of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in the North Pacific Ocean on 1 January 2000.

Radionuclide	Effective half-life, $T_{1/2} \pm 1\sigma$	Estimated concentration
^{90}Sr	12.4 ± 1.1	$0.9\text{--}1.3 \text{ mBq l}^{-1}$
^{137}Cs	12.7 ± 1.4	$1.7\text{--}2.8 \text{ mBq l}^{-1}$
$^{239,240}\text{Pu}$	6.7 ± 0.9	$0.9\text{--}2.8 \text{ } \mu\text{Bq l}^{-1}$

3.1 Temporal changes in the artificial radionuclide concentrations in surface waters

Time series variations of anthropogenic radionuclide (^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$) concentrations during the period from 1971 to 1998 were examined to estimate their surface concentrations in the year 2000. In general, surface concentrations of these radionuclides have significantly decreased over recent decades exponentially with time (Table 5). Therefore, time variation of these radionuclide concentrations may be described as a first order reaction and reaction constant is some combination of radioactive decay constant and rate of removal from the surface ocean at a given site. The effective half-lives of these radionuclides in the surface ocean is also given in Table 5.

3.2 Vertical distribution of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in the water column

The evolution of vertical profiles of ^{90}Sr and ^{137}Cs concentrations in the water column of the North Pacific was observed by comparing data from GEOSECS cruise in 1973 and IAEA cruise in 1997 (Povinec *et al.*, 2003) during those two time periods. GEOSECS stations 223, 224, 225, 226 and IAEA97 stations 1 and 3 may be compared due to their geographical positions are near and share similar oceanographic characters as done by Povinec *et al.* (2003). A little more detailed examination is attempted here. These stations were located in the downstream of the Kuroshio Extensions and its primary and export production are $\sim 100 \text{ g C m}^{-2}$ and 10 g C m^{-2} , respectively (Falkowski *et al.*, 2003). Those primary production and export production values are read from figures and subjected to some uncertainties. GEOSECS stations 229 and IAEA97 station 6 may be compared due to their proximity and both stations were located in the oligotrophic gyre and the primary and export production are $< 20 \text{ g C m}^{-2}$ and $< 2 \text{ g C m}^{-2}$, respectively (Falkowski *et al.*, 2003). For the sake of discussion, the former area is termed as eutrophic and the latter as oligotrophic area. In the eutrophic area, the vertical distribution pattern of ^{90}Sr and ^{137}Cs did not change during the 24 years time interval, with the subsurface maximum and gradual decrease to the 1000 m depth where the permanent thermocline resides. The concentrations of both ^{90}Sr and ^{137}Cs are very low ($< 0.1 \text{ Bq l}^{-1}$) with very little variation with depth below the thermocline to the near bottom. However, the surface concentration in 1997 decreased $\sim 1/3$ of the values observed in 1973 for ^{90}Sr and $\sim 1/4$ for ^{137}Cs . In the oligotrophic region, the vertical distribution pattern

is very similar to each other in GEOSECS and IAEA97 cruises and ^{90}Sr and ^{137}Cs concentrations peaked both at the subsurface and decreased toward the thermocline, located at 500 m, with very little variation with depth. The current surface concentration of ^{90}Sr and ^{137}Cs concentration has decreased more to less than half of those observed 24 years ago. Since the half-lives of ^{90}Sr and ^{137}Cs are 28.78 and 30.07 years, respectively, the decrease in concentration could be explained by their radioactive decay and their involvement of particle formation processes *in situ*, if the residence time of surface waters within this oligotrophic gyre is much longer than the time elapsed between the two measurements, 24 years.

For $^{239+240}\text{Pu}$, in the eutrophic region, several changes during the 24 years time period can be noted as observed for ^{90}Sr and ^{137}Cs . General vertical distribution pattern is maintained during this period with surface minimum, subsurface maximum and decrease with depth to ca. 2000 m and slight increase below 5000 m toward the bottom as reported by Bowen *et al.* (1980). However, the location of the subsurface maximum in the eutrophic water moved down from ca. 420 m to 800 m from 1973 to 1997 and the subsurface peak concentration decreased to as much as ca. 1/4 of the concentration observed 24 years ago. However, in the oligotrophic region, the subsurface concentration peak in 1997 decreased only ca. 1/7 of the concentration observed in 1973 and the position of the subsurface concentration peak moved down from ca. 600 m to 700 m. Since ^{239}Pu and ^{240}Pu decay very slowly, the decrease in their concentrations factors and processes other than radioactive decay must cause the decrease in their concentrations. The primary mechanism causing the decrease in the surface $^{239+240}\text{Pu}$ concentration and its downward movement is the export flux of Pu-laden particles from the surface to the deep. In the open ocean, Moore and Dymond (1988) observed a strong correlation between ^{210}Pb removal rates from the water column and the flux of particulate organic carbon. Deuser *et al.* (1983) further observed that the removal of particulate Al to deep water is intimately linked to the rapid downward transport of organic matter, although the concentration of particulate aluminum was variable. However, the particles in the deep ocean are largely biogenic, made of organic matter. Bacon *et al.* (1985) also reported that sinking $^{239+240}\text{Pu}$ flux was well correlated with the mass flux in the oligotrophic gyre in the Atlantic Ocean. It appears that more than 5 fold differences in the export flux between the eutrophic and oligotrophic region explains the relative decrease in $^{239+240}\text{Pu}$ concentration in the northwest Pacific. However, we need to confirm this hypothesis by obtaining the sinking $^{239+240}\text{Pu}$ fluxes in both regions.

Livingston and Povinec (2002) argued that the decrease of plutonium concentration could be caused due to the advection of water masses bearing lower level of plutonium into the region. Subsequently, a term for the water mass movement was included in the plutonium scavenging model coupled with an Ocean General Circulation Model (Nakano and Povinec, 2003). This modeling confirmed the importance of advection of water masses in the distribution of plutonium in the ocean. They also confirmed that local fallout originated about 10 years earlier than global fallout and higher $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios were found

in deeper and bottom waters near Bikini and Enewetak atolls (159.5°E 15.5°N) as well as central North Pacific (170°E 30°N).

3.3 *Biological control versus abiological control of sinking flux of artificial radionuclides*

Since most fallout artificial radionuclides were delivered to the surface of the sea and a substantial fraction of total input has arrived at the deep sea floor, the understanding of the mechanisms of downward movement of these radionuclides is crucial to assess their consequences and environmental impacts in the ocean. Most artificial radionuclides introduced into the marine environment is particle reactive but not necessarily biologically active. Although marine particles have a wide range of sources and characteristics, the majority in the open ocean is phytoplankton cells. Moreover, the phytoplankton cells were eaten by zooplankton. Zooplankton subsequently defecated their feces. This fecal formation acts as a packaging mechanism for the phytoplankton cells and thereby acts as ballast for the subsequent sinking to the interior of the sea and eventually to the sea floor. This mechanism is believed to be primary vehicle for the artificial radionuclides and many other chemical compounds delivered to the deep sea floor.

In general, marine phytoplankton concentrates Pu, Am from seawater and result in volume/volume concentration factors generally $0.3\text{--}6.3 \times 10^5$ for Pu and $0.3\text{--}6.9 \times 10^5$ for Am (Fisher *et al.*, 1983). An apparent distribution coefficient, K_d , which is defined as the concentration in the solid phase (Bq kg^{-1})/concentration in the liquid phase (Bq l^{-1}), is used to characterize the particle reactivity of radionuclides. Strict use of K_d implies that reversible adsorption/desorption equilibrium is established; however, the mechanisms acting in sediments may be non-reversible and may not be in equilibrium. To a first approximation, an adsorption/desorption process described by an apparent K_d of $1.7\text{--}11.8 \times 10^5$ for Pu and $3.5\text{--}8.9 \times 10^5$ for Am in the bottom sediment (Malcom *et al.*, 1990). Their detailed depth profiles of apparent K_d for Pu and Am in the porewaters indicated that the sediment/interstitial water system is not in equilibrium. Any post depositional mobility could not be observed for Pu and Am isotopes in anoxic marine sediments (Carpenter and Beasley, 1981). ^{137}Cs is also observed to be virtually non-exchangeable and strongly adsorbed by clay particles in estuarine sediments (Cundy and Croudace, 1995).

Higgo *et al.* (1977) measured the Pu concentrations in whole euphausiids, fecal pellets, exoskeletons and derived the flux rates of Pu through living euphausiids. They further calculated removal time of the Pu from the surface mixed layer using $1 \times 10^{-4} \text{ kg m}^{-3}$ of dry zooplankton biomass in the world ocean and obtained 3.6 yr as the removal time. Since Pu is not actively taken up by zooplankton, and any Pu taken-up is largely expelled from the zooplankton body as fecal pellets (Hong *et al.*, 2002). The velocity of sinking of pellets varied from 126 to 862 m d^{-1} . In general, settling rates of fecal pellets and other particulate matter is believed to be ranging from <1 to >1000 meters a day at sea (Buesseler *et al.*, 2000).

Table 6. Particulate fluxes of Pu isotopes in the Pacific Ocean.

Region	Coordinates	Water depth (m)	Trap depth (m)	²³⁹⁺²⁴⁰ Pu concentration (mBq g ⁻¹)	Particulate ²³⁹⁺²⁴⁰ Pu flux (mBq m ⁻² d ⁻¹)	Org C flux (mg C m ⁻² d ⁻¹) / Total mass flux (mg m ⁻² d ⁻¹)	Remark
Equatorial North Pacific	15°21' N	5792	5582	35.8	0.35	/9.7 ⁶	
	151°W						
Panama Basin	5°6' N	3880	3791	5.3	0.96	/183 ⁶	1.19 mBq ¹³⁷ Cs m ⁻² d ⁻¹
	81°32' W						Anoxic basin
Santa Monica and San Pedro Basins	33°45' N	910	850	6.73	0.4 ³	34/767 ⁴	
	118°53' W						
East Sea (Sea of Japan) ⁷	39°40' N	3100	2800	3.7–5.2	0.2–4.4 ¹	0.7–35.4/50–1232 ²	Deep basin
	132°24' E						
	36°55' N	2160	1000	2.4–2.8	0.7–1.0	17.35/321	Deep basin
East China Sea	131°00' E						
	28°40.86' N	132–604	72–592	2.0–2.7	0.5–0.8	11.4/270.4	
	127°03' E			2.85–4.18	34.6–82.7	/1000–1500 ⁵	Continental slope, resuspension

1. Lee *et al.* (2003), 2. Hong *et al.* (1997), 3. Wong *et al.* (1992), 4. Crisp *et al.* (1979), 5. Yamada and Aono (2002), 6. Livingston and Anderson (1983), 7. this study.

Direct intercept of sinking particles in the sea is usually made by sediment trap. Buesseler *et al.* (2000) recently reviewed collection efficiency of sediment traps at sea. While traps are widely used to serve as accurate “rain gauges” in the oceans, the absolute accuracy of traps has not been well documented in the field. By and large, the data from traps have been internally consistent and this consistency and the “reasonableness” of the results have been one of the strongest arguments that the data cannot be too far from the true particle flux. However, differences between radionuclide fluxes calculated from measured water column ^{234}Th inventories and measured trap ^{234}Th fluxes have suggested that traps can both over- and under-collect in the field by up to a factor of 10 in the upper ocean (Buesseler *et al.*, 1991). Direct particle flux measurements and measurements of artificial radionuclides in the trap material from the Pacific Ocean is rare and

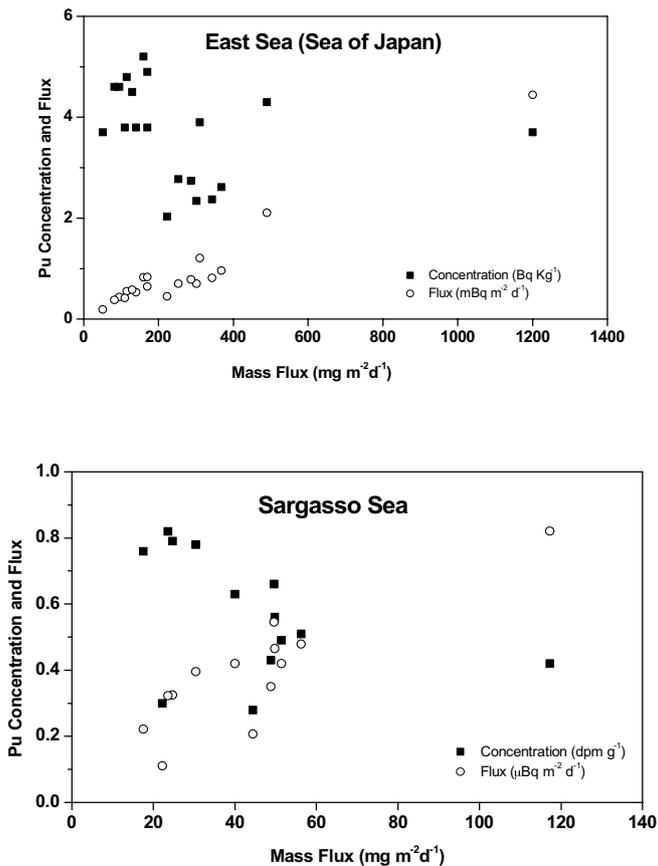


Fig. 1. Variation of concentration and flux of Pu with mass flux observed in the East Sea (Sea of Japan) in the Pacific and Sargasso Sea in the Atlantic from Bacon *et al.* (1985).

there is no measurement in the northwest Pacific Ocean proper where coastal influence is minimal (Table 6). Therefore it is urgent to obtain the artificial radionuclide sinking fluxes in the northwest Pacific Basin in order to understand the basin wide scavenging feature of these nuclides.

Deuser *et al.* (1983) conducted concurrent measurements of particle concentrations in the near-surface water and of particle fluxes in the deep water of the Sargasso Sea and showed a close coupling between the two for biogenic components. The concentrations of suspended matter appeared to follow an annual cycle similar to that of primary production and deepwater particle flux. Although the concentration of particulate aluminum in the surface water appeared to vary randomly with respect to that cycle, the removal of aluminum in deep water seemed to be intimately linked to the rapid downward transport of organic matter. The rate of scavenging of particle-reactive nuclides depends highly on the nature and composition of suspended particulate matter. For example, Chase *et al.* (2002) found that the affinity of particles for Pa and Be increases with their increasing opal content and decreasing carbonate content, while the affinity of

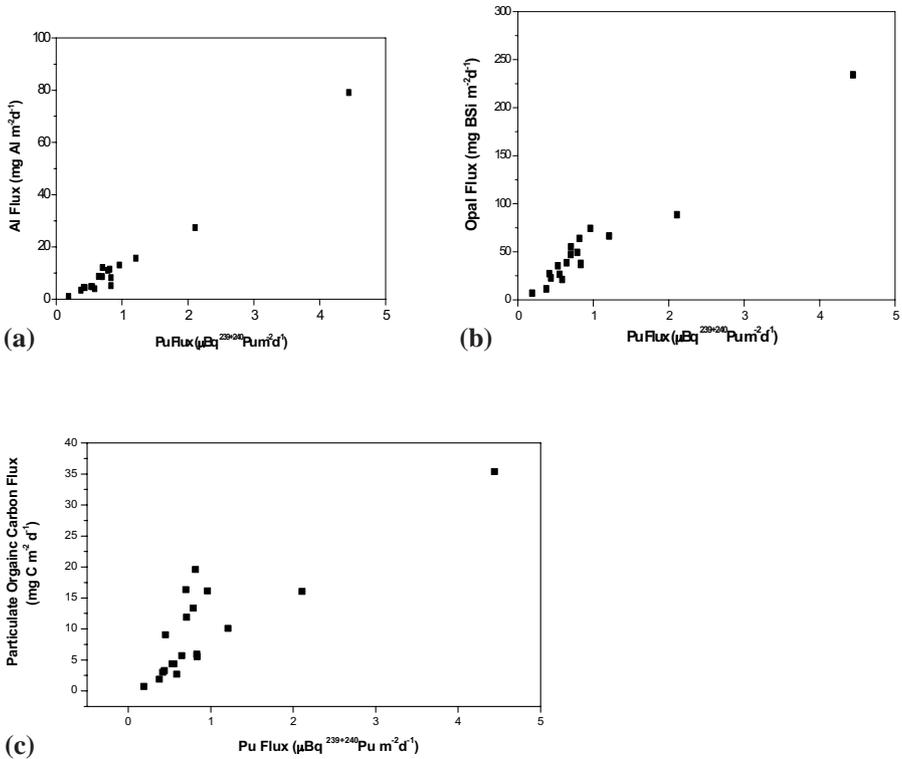


Fig. 2. $^{239+240}\text{Pu}$ settling flux ($\mu\text{Bq } ^{239+240}\text{Pu m}^{-2}\text{d}^{-1}$) versus fluxes of Al ($\text{mg Al m}^{-2}\text{d}^{-1}$), biogenic Si ($\text{mg BSi m}^{-2}\text{d}^{-1}$) and organic C ($\text{mg org C m}^{-2}\text{d}^{-1}$) in the deep basin of the East Sea (Sea of Japan).

particles for Th increases with increasing carbonate content and decreases with increasing opal content. Plutonium exists in four oxidation states (reduced: III and IV; oxidized: V and VI), having each state different solubility, complexation and adsorption properties (Sholkovitz, 1983). Since the chemical form of plutonium could vary with depth with water depth (Nelson and Lovett, 1978) and this would imply that the plutonium partition coefficient will not be constant through the water column either (Livingston *et al.*, 2001). It will argue against a simple complexation-scavenging equilibrium model (linear relationship between concentrations of particulate Pu and organic ligands in the surface waters) proposed by Hirose *et al.* (2003).

However, Pu scavenging onto the settling particulate matter may be more dependent upon the bulk mass flux rather than their chemical composition of the particulate matter as observed for other radionuclides and metals in the sea. Observations in both the Sargasso Sea in the Atlantic Ocean and the East Sea (Sea of Japan) exhibited that the Pu concentration in the settling particles decreases with the increase in bulk mass flux and the Pu settling flux increases with the increase in bulk mass flux despite their very different oceanographic conditions (Fig. 1).

In the East Sea, settling flux of $^{239+240}\text{Pu}$ appear to be dependent more strongly upon the Al and biogenic Si fluxes than organic matter flux both in the Japan Basin and Ulleung Basin for shallow (1000 m) and deep (2000 m and 2800 m) waters (Fig. 2). However, there is a relationship between the settling flux of $^{239,240}\text{Pu}$ and organic carbon for only deep Japan Basin traps (2800 m).

In the Uleung Basin, the dissolution flux of organic carbon, chlorophyll a were calculated by measuring the organic carbon and flux and chl. a in the trap material collected at 1000 m and 2000 m. The differences in org C flux and $^{239+240}\text{Pu}$ flux between 1000 m and 200 m traps were not positively linear, although we have only three observations. The dissolution rate between 1000 m and 2000 m depth intervals was calculated by subtracting the flux value measured at 2000 m from that at 1000 m depth (Fig. 3).

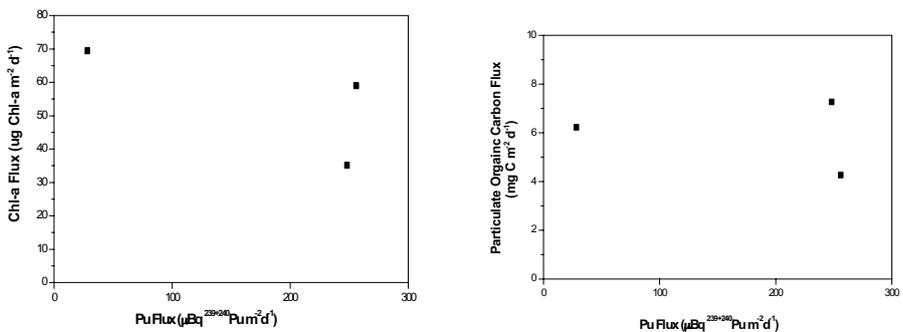


Fig. 3. Comparison of dissolution of $^{239+240}\text{Pu}$ ($\mu\text{Bq } ^{239+240}\text{Pu m}^{-2}\text{d}^{-1}$) and organic carbon ($\text{mg org C m}^{-2}\text{d}^{-1}$) and chlorophyll *a* ($\mu\text{g chlorophyll a m}^{-2}\text{d}^{-1}$) between 1000 and 2000 m depth interval.

Table 7. Activity and atom ratios of artificial radionuclides originated from the global fallout.

Radionuclide	Half-life (yr)		Global fallout (activity ratio) in 2000	Ref.
^3H	12.33	$^3\text{H}/^{90}\text{Sr}$	299	1
^{14}C	5730	$^{14}\text{C}/^{90}\text{Sr}$	0.3424	1
^{90}Sr	28.78			1
^{137}Cs	30.07	$^{137}\text{Cs}/^{90}\text{Sr}$	1.52	1
^{134}Cs	2.062			
^{238}Pu	87.74	$^{238}\text{Pu}/^{239+240}\text{Pu}$	0.030	2, 3, 5
^{239}Pu	24110	$^{239}\text{Pu}/^{90}\text{Sr}$	0.0105	1
^{240}Pu	6563	$^{240}\text{Pu}/^{90}\text{Sr}$	0.007	1
^{241}Pu	14.35	$^{241}\text{Pu}/^{90}\text{Sr}$	0.2283	1
		$^{241}\text{Pu}/^{239+240}\text{Pu}$	13–14	4
		$^{240}\text{Pu}/^{239}\text{Pu}$	0.6672 (activity ratio)	
		$^{239+240}\text{Pu}/^{137}\text{Cs}$	0.182 (atom ratio)	
			0.012 (activity ratio)	
^{241}Am	432.7			

1. Aarkrog (2003), 2. decay corrected to the year of January 1, 2000 using Aarkrog (1988) for the northern hemisphere, 3. Baskaran *et al.* (1995), 4. Hirose *et al.* (2001), 5. Baskaran *et al.* (2000).

4. UTILITY OF ARTIFICIAL RADIONUCLIDES AS GEOCHEMICAL AND OCEANOGRAPHIC TRACERS

As radionuclides enter the ocean, they become involved in various ocean processes depending on their chemistry including solubility and affinity to the particles and involvement in the biological and sedimentation processes occurring in the ocean. Since the concentrations, activity and atomic ratios of various nuclides are useful as time markers, the activity and atom ratios of artificial radionuclides in the marine environmental compartments has been studied extensively. Therefore, a large number of studies of fallout nuclides in the ocean have contributed either directly or indirectly to studies on ocean currents and mixing, particle association, transport and fate in the water column and bottom sediments, and biological processes and their interaction with both fallout radionuclides and their elemental analogues including radiation effects (Livingston and Povinec, 2002). Some of the isotope signatures that have been utilized for such purposes are compiled here for future reference (Tables 7 and 8). The global fallout itself provides a number of very useful isotopic signatures in marine research (Table 7). The isotopic signatures originated from the global fallout have been significantly modified by the introduction of various sources as mentioned above (Table 8) depending upon the region. However, the isotopic signatures of close-in fallout have been modified by the global fallout in the Western North Pacific, since the close-in fallout preceded major global fallout period. Based on the published nuclear test information, the Pacific Proving Grounds tests account for >95% of all surface-based nuclear weapons tests (based upon the yields calculated from Department of Energy (1982) and reported in Buesseler (1997)).

Table 8. Activity and atom ratios of artificial radionuclides originated from the various sources other than global fallout.

Source	$^{240}\text{Pu}/^{239}\text{Pu}$ (atom ratio)	$^{238}\text{Pu}/^{239+240}\text{Pu}$ (activity ratio, 2000)	$^{241}\text{Am}/^{239+240}\text{Pu}$ (activity ratio, 2000)	$^{137}\text{Cs}/^{134}\text{Cs}$ (activity ratio, 2000)	$^{137}\text{Cs}/^{90}\text{Sr}$ (activity ratio, 2000)	$^{239+240}\text{Pu}/^{137}\text{Cs}$ (activity ratio)	$^{238}\text{U}/^{235}\text{U}$ (atom ratio)
Chernobyl	0.40 ⁶	0.5 (4 Hirose)		85 ⁵	12.1 ⁵		
Effluent from Selafield dumped reactors in Kara Sea	0.242 ¹¹	0.27 ± 0.02 ³ 0.25–0.46 ⁵					
Coral rock and top soil (Mururoa Atoll) (1997)		0.54 ± 0.16 ⁴	1.69 ± 0.51				
Loose coral rocks		30.7 ± 8.6 ⁴	5.0 ± 1.2				
Natural uranium							137.88 ⁶ 15–32 before burn up ⁶
Reactor							
MAGNOX reactor (GCR)	0.23 ^{8,6}						
Pressurized heavy water reactor (PHWR)	0.41 ^{8,6}						
Advanced gas-cooled reactor (AGR)	0.57 ^{8,6}						
Pressure tube boiling water reactor (RBMK)	0.67 ^{8,6}						
Boiling water reactor (BWR)	0.40 ^{8,6}						
Pressurized water reactor (PWR)	0.43 ^{8,6}						
Weapon grade uranium							
Depleted uranium							<0.1 ⁶ 250–500 ⁶
Weapon production	0.01–0.07 ⁶						
Ivy/Mike shot Eniwetok Island	0.36 ⁷						
Rongelap atoll	0.276 ⁸						0.28
Bikini atoll	0.306 ⁸						0.12
Enewetak atoll (Runit Island)	0.065 ⁸						34.7
Enewetak atoll (Aej Island)	0.254 ⁸						1.06
Marshall Island	0.303 ⁸						0.20
Semipalatinsk, Chenaya Guba, Nevada test sites soils Nagasaki, Japan	0.037 ⁹						
Mururoa and Fangataua, sediments	0.035–0.05 ¹⁰						

* After fuel burn-up.

4. Irlweck and Hmecek (1999), 5. Aoyama *et al.* (1991), 6. Warneke *et al.* (2002), 7. Koide *et al.* (1985), 8. Muramatsu *et al.* (2001), 9. Yoshida and Muramatsu (2003), 10. Chiappini *et al.* (1999), 11. Lee *et al.* (2001).

By comparing marine water column data with the soil $^{239+240}\text{Pu}$ inventories, it is estimated that up to 60% of the Pu inventory in the North Pacific originated from local sources (Bowen *et al.*, 1980). The two primordial isotopes of uranium are ^{235}U and ^{238}U with $^{238}\text{U}/^{235}\text{U}_{\text{present}} = 137.88$ and this atomic abundance ratio has no significant variation in nature except in fossil natural nuclear reactors. Deviations from this ratio in environmental materials can therefore only be explained by the addition of technologically modified uranium (Table 8).

Among artificial radionuclides, plutonium, unlike uranium, is virtually anthropogenic in origin and its main isotopes found in the environment, ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu and ^{242}Pu , are derived from civil and military sources, and they deserve a special attention as a oceanographic tracer because plutonium exists as various physical/chemical forms and various oxidation states and their activity ratios or atomic ratios provide a great deal of information regarding the sources of Pu and their behavior in the sea. Buessler and Sholkovitz (1987) reviewed the atom ratio of $^{240}\text{Pu}/^{239}\text{Pu}$ of the fallout. Differences in weapons design appear to account for much of the variability seen in the historical $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios. Within any given design or series of tests, a larger nuclear yield produces greater neutron fluxes resulting in the enhanced production of the higher masses of Pu. The U.S. nuclear testing program dominated the early atmospheric fallout record in terms of total fallout yields. In the early 1950s, stratospheric fallout was characterized by a $^{240}\text{Pu}/^{239}\text{Pu}$ ratio greater than 0.30, dropping off to the 0.21–0.26 in the mid-1950s. A low fallout with atomic ratio of 0.035 from the Nevada Test site produced during the above ground testing in the early 1950s. Later atmospheric nuclear tests in the early 1960s were dominated by the large yields of former Soviet Union tests. These former USSR tests accounted for ca. 75% of the total yield. The average $^{240}\text{Pu}/^{239}\text{Pu}$ ratio of this material was found to be 0.18. Buessler and Sholkovitz (1987) further suggested that the solubility and reactivity of Pu from different fallout sources might not be necessarily identical, as was often previously assumed.

5. RETROSPECTIVE STUDIES ON THE ARTIFICIAL RADIONUCLIDES IN THE SURFACE OCEAN

5.1 Works up to date

Fallout strontium and plutonium for the above ground nuclear testing of the 1950's and 1960's was not systematically monitored through the ocean studies. However, the history of some of the artificial radionuclides contamination in surface seawater has been revealed by studying annually banded corals as time discriminant recorders of seawater composition (Table 9). Plutonium has been measured in the annual bands of the corals *Favites virens* from Bikini Lagoon (Noshkin *et al.*, 1975) and *Porites* of French Frigate Shoals Island (24°N, 166°W, Buessler, 1997) in the Pacific and *Montastrea annularis* from St. Croix (Benninger and Dodge, 1986) and the Florida Keys (Purdy *et al.*, 1989) in the Atlantic. Meece and Benninger (1993) conducted CaCO_3 coprecipitation experiments with low levels of oxidized and reduced Pu and a complementary suite of other radionuclides

to evaluate the utility of the Pu coral record as a tracer of seawater Pu history. According to them, Reduced Pu (III and IV), Am, Th and Pb were inferred to be incorporated via adsorption, and U was incorporated via solid solution formation. However, oxidized Pu (VI) was reduced to Pu (V) and then sorbed onto the solids. There is no discrimination of $^{90}\text{Sr}/\text{Sr}$ in corals relative to its composition in seawater and coral took up Pu isotopes in proportion to its concentration in the water (Table 10). All the compiled $^{240}\text{Pu}/^{239}\text{Pu}$ data was plotted with time (Fig. 4).

In the terrestrial environments, ice cores and plant archives were used for the reconstruction of deposition history of the artificial radionuclides (Table 9). Koide *et al.* (1985) analyzed artificial radionuclide concentrations in ice cores and in air filters, and demonstrated that there was a characteristic pattern from stratospheric fallout of the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio with time. It started with the highest ratio (0.34–0.28 depending on geographic location), decreased to values of about 0.22 just prior to the moratorium, hovers around 0.18 during the 1960's decreases to low values during 1969–1970 and then rises slightly. No explanation was given for the slight rise in the atomic ratio of $^{240}\text{Pu}/^{239}\text{Pu}$ after 1970 by both Koide *et al.* (1985) and Warneke *et al.* (2002). Recent increase in $^{240}\text{Pu}/^{239}\text{Pu}$ after 1986 may be attributable to the Chernobyl fallout. A value of 0.44 was also observed in lichen samples collected in Sweden in 1986–1988 (Lindahl *et al.*, 2004).

5.2 Time variation of Pu isotopes in the northwest Pacific in the last two decades: A plan

$^{239+240}\text{Pu}$ concentrations and $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios in coastal seas adjacent to the Korean Peninsula were determined during the period from 1999 to 2002 to assess the current distribution and to identify sources of Pu isotopes in both water and bottom sediments. $^{239+240}\text{Pu}$ concentrations around the Korean Peninsula is greater than those in the western North Pacific. The $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios in dissolved seawater ranged from 0.18 to 0.33 with an average value of 0.25 ± 0.03 (Kim *et al.*, 2004), which are significantly higher than that of global fallout, and they are comparable to those observed in the deep waters near the Marshall Islands and the areas under the influence of the US Pacific Proving Grounds of Marshall Islands (Buesseler, 1997). $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio in the subsurface bottom sediments (>3 cm) was higher (0.23) than that (0.18) of the surface sediments (0–3 cm) in the 2200 m deep Ulleung Basin, southwestern basin of the East Sea (Sea of Japan) (Kim *et al.*, 2003). Therefore, it appears that Pu isotopes are continuously transported both dissolved and particulate forms from the Pacific to the East Sea (Sea of Japan) and these Pu are eventually scavenged in the East Sea. Hence, the $^{240}\text{Pu}/^{239}\text{Pu}$ signature appears to have changed from the tropospheric fallout to the global fallout until 1980s and then tropospheric fallout again became the dominant source from the Marshall Islands to the western North Pacific Ocean. In order to understand the temporal variations of the changes in Pu sources, it is necessary to reconstruct the past changes in the atom ratio of $^{240}\text{Pu}/^{239}\text{Pu}$ in the northwest Pacific Ocean, especially for the last two decades where the

Table 9. Time series studies on the artificial radionuclide concentration evolution using various geochemical reservoirs.

Reference	Material	Site	Collection year	Period covered	Radionuclide
Koide <i>et al.</i> (1985)	Ice core	Ross Ice Shelf, Antarctica (19, 82°22' S 168°40' W)	1976	1954–1968	²⁴⁰ Pu, ²³⁹ Pu, ²⁴¹ Pu, ²³⁸ Pu, ¹³⁷ Cs
		Dome C, Antarctica (74°39' S 123°10' E)	1977	1963–1973	
		South Dome, Greenland (65°11' N 43°50' W)	1975	1956–1965	
		Dye-3, Greenland (65°11' N 43°50' W)	1980	1954–1965	
Dibb <i>et al.</i> (1990)	Ice core	Antarctic Pole		1950 (?)–1989(?)	Total beta, ¹³⁷ Cs
Warneke <i>et al.</i> (2002)	Ice core	Dome du Gouter (4300 m), Mont Blanc (ca. 46°N 7°E)		1945–1990	²⁴⁰ Pu, ²³⁹ Pu, ²³⁸ U, ²³⁵ U, ¹³⁷ Cs
Warneke <i>et al.</i> (2002)	Grass	IACR Rothamsted archive (Harpenden, 25 miles South of London)	1945–1990	1945–1990	²⁴⁰ Pu, ²³⁹ Pu, ²³⁸ U, ²³⁵ U
Benninger and Dodge (1986)	Coral, <i>Montastrea annularis</i>	St. Croix, US Virgin Island, A (18°20' N 64°50' W)	1980	1951–1980	²³⁹⁻²⁴⁰ Pu, ²³⁸ U, ²²⁸ Th, ²¹⁰ Po, ²²⁶ Ra, ²²⁶ Ra, Ca, Mg, Sr, Na
Purdy <i>et al.</i> (1989)	Coral, <i>Montastrea annularis</i>	The Rocks, Florida Keys, A (24°57' N 80°33' W)	1983	1940–1982	⁹⁰ Sr, ²³⁹⁻²⁴⁰ Pu (seasonal ⁹⁰ Sr activity for 1973 and 1974)
		New Ground reef, A (24°40' N 82°25' W)	1983		⁹⁰ Sr
	<i>Diploria strigosa</i>	North Rock reef, Bermuda, A (32°20' N 64°45' W)	1983	1951–1982	⁹⁰ Sr
Noshkin <i>et al.</i> (1975)	Coral, <i>Favites virens</i>	Bikini Lagoon, NP (ca. 11°36' N 165°22' E)	1972	1954–1972	²³⁹ Pu, ²⁴⁰ Pu, ²³⁸ Pu, ²⁴¹ Pu, ²⁴¹ Am, ¹⁵⁵ Eu, ²⁰⁷ Bi, ⁶⁰ Co, ¹³⁷ Cs, ^{102m} Rh, ¹²⁵ Sb, ⁹⁰ Sr, ²¹⁰ Pb

Reference	Material	Site	Collection year	Period covered	Radionuclide
Toggweiler and Trumbore (1985)	Coral	Tarawa, NP (1°25' N 173° E)		1959–1978	⁹⁰ Sr
	Coral	Oahu, NP (21° 18' N 158° 07' E)		1958–1979	⁹⁰ Sr
	Coral	Fanning, NP (3° 51' N 159° 21' E)		1951–1978	⁹⁰ Sr
	Coral	Fiji, SP (18° S 175° W)		1949–1978	⁹⁰ Sr
	Coral	Cocos, I (12° 30' S 96° 50' E)		1954–1978	⁹⁰ Sr
	Coral	Mauritius, I (21° 17' S 57° 33' E)		1968–1978	⁹⁰ Sr
	Coral	Galapagos, SP (1° S 90° W)		1974–1976	⁹⁰ Sr
	Coral	Tonga, SP (20° S 175° W)		1962–1978	⁹⁰ Sr
	Coral	Great Barrier Reef, Australia, SP (23° S 151° E)		1969–1979	⁹⁰ Sr
Buesseler (1997)	Coral, <i>Porite</i>	French Frigate Shoal Island, NP (24° N 166° W)	1978	1955–1964	²⁴⁰ Pu, ²³⁹ Pu
Hong <i>et al.</i> (in preparation)	Coral, <i>Poites australiens</i>	Ishigaki Island, P (24° 30' N 124° 20' E)	1998	1945–1998	²³⁹ Pu, ²⁴⁰ Pu, ²³⁸ Pu, ²⁴¹ Am, ¹³⁷ Cs, ⁹⁰ Sr, ²¹⁰ Pb, ²³⁸ U, ²²⁸ Th, ²²⁸ Ra, ²²⁶ Ra

Note: NP, SP, I, and A stand for the North Pacific, South Pacific, Indian, and Atlantic Ocean, respectively.

Table 10. Concentration factor (CF) and distribution coefficient (K_d) for coral skeleton.

Radionuclide	Factor and coefficient		Reference
^{90}Sr	CF	1.1×10^3	Noshkin <i>et al.</i> (1975)
	K_d (for stable Sr)	1.040 ± 0.008	Benninger and Dodge (1986)
$^{239,240}\text{Pu}$	CF	2.7×10^3	Noshkin <i>et al.</i> (1975)
	K_d	1.8 ± 1.2	Benninger and Dodge (1986)
^{137}Cs	CF	2.3×10^3	Estimated from Noshkin <i>et al.</i> (1975)

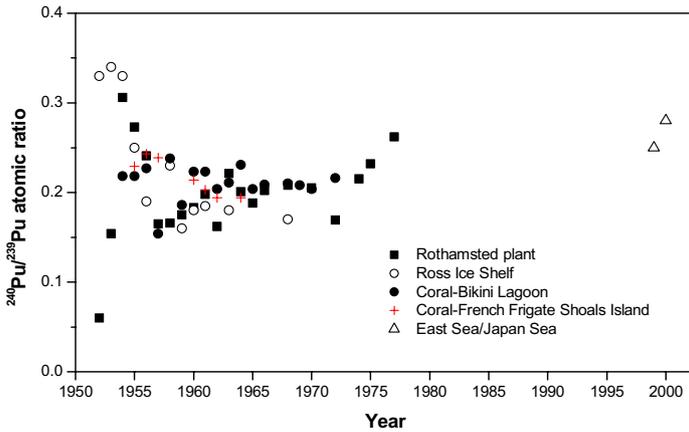


Fig. 4. Time variation of $^{240}\text{Pu}/^{239}\text{Pu}$ in various geochemical reservoirs over the surface of the Earth.

data do not exist. As a part of the collaborative work with Dr. H. Kawahata (Geological Survey of Japan), we have started to analyze dated coral samples taken from Isigaki Island which is located in the Pacific Basin proper well away from the influence of coastal waters. These coral samples will provide a unique opportunity to trace the time evolution of artificial radionuclides in the Kuroshio region. The Kuroshio water is the main ocean water feeds the seas adjacent to the Korean Peninsula.

6. CONCLUSIONS AND SUGGESTIONS

- Both global and close-in tropospheric fallouts are the major sources of artificial radionuclides currently present in the northwestern Pacific. Radioactive

waste dumping and other sources do not appear to be contributing significantly to the anthropogenic radionuclide levels in this region. However, other potential inputs should be carefully monitored and the current established databases of IAEA and Japan serve as the baseline for future work.

2. There is no clear understanding on the behavior of $^{239+240}\text{Pu}$ in the northwest Pacific Ocean proper. It appears that more than 5 folds differences in the export flux of Pu between the eutrophic and oligotrophic region explains the relative decrease in $^{239+240}\text{Pu}$ concentration in the northwest Pacific. However, there are no direct measurements of sinking Pu isotopes in the northwestern Pacific proper. We need to confirm this hypothesis by obtaining the sinking $^{239+240}\text{Pu}$ fluxes in both regions.

3. There is no systematic monitoring of the artificial radionuclides in seawater in this region, particularly beyond the 1980s. Therefore we need to analyze various geochemical reservoirs that accumulated high resolution temporal variation of the surface water concentrations.

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