



Anthropogenic Pu distribution in Tropical East Pacific

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ABSTRACT

The geographical distribution of the anthropogenic radionuclides ²³⁸Pu and ²³⁹⁺²⁴⁰Pu in the Tropical East Pacific in 2003 was studied from the viewpoint of material migration. We measured the contents of Pu isotopes in seawater and in sediment from the sea bottom. The distributions of Pu isotopes, together with those of coexisting nitrate and phosphate species and dissolved oxygen, are discussed in relation to the potential temperature and potential density ($\sigma\text{-}\theta$). The Pu contents in sediment samples were compared with those in the seawater. Horizontal migration across the Equator from north to south was investigated at depths down to ~800 m in the eastern Pacific. The Pu distribution at 0–400 m correlated well with the distribution of potential temperature. Maximum Pu levels were observed in the subsurface layer at 600–800 m, corresponding to the depth where $\sigma\text{-}\theta \approx 27.0$. It is suggested that the Pu distribution depends on the structure of the water mass and the particular temperature and salinity. The water column/sediment column inventory ratio and the vertical distribution of Pu may reflect the efficiency of scavenging in the relevant water areas.

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1. Introduction

Radionuclides emitted during atmospheric tests of nuclear weapons have been widely used as proxies for tracing the migrations of materials (Doney, 1992; Tosaki et al., 2007). Most of these nuclides were deposited on the oceans, which occupy about 70% of the Earth's surface. It is believed that nuclides present as insoluble species become adsorbed by suspended materials in seawater. The suspended materials, together with the radionuclides that they contain, subsequently sink from the surface to the sea bottom, whereas soluble species, such as ¹³⁷Cs, remain in the surface waters for many years (Yamada and Wang, 2007) and migrate both vertically and horizontally under the influence of oceanic currents.

All isotopes of Pu are artificial radioactive nuclides. Most of the isotopes [²³⁸Pu ($T_{1/2} = 87.7$ yr), ²³⁹Pu ($T_{1/2} = 2.411 \times 10^4$ yr), ²⁴⁰Pu ($T_{1/2} = 6.563 \times 10^3$ yr), ²⁴¹Pu ($T_{1/2} = 14.254$ yr), ²⁴²Pu ($T_{1/2} = 3.833 \times 10^5$ yr), and ²⁴⁴Pu ($T_{1/2} = 8.08 \times 10^7$ yr)] that are found in the environment were discharged from above-ground tests of nuclear weapons during the 1950s and early 1960s (Perkins and Thomas, 1980; Aarkrog, 2003). In addition, some Pu nuclides originate from various accidental releases and from reprocessing of nuclear materials (Kershaw et al., 1999).

Several investigations have been carried out by using environmental Pu isotopes. The chemical forms of Pu in seawater were evaluated by Aston (1980), who focused on its valence state and stability constant with inorganic ligands. The Pu nuclides in size-fractionated particles were analyzed by Dai et al. (2001), who reported that the most Pu exists in a low-molecular-weight fraction, with a few percent of Pu in a colloidal form. Several researchers have measured the atom ratios of Pu isotopes in sea sediment and in the ferromanganese crust by means of accelerator mass spectrometry (Paul et al., 2001; Wallner et al., 2000). Pu isotopes of mass numbers up to 244, formed a result of multiple neutron capture by ²³⁹Pu, have been measured. In addition, several researchers have measured Pu concentrations and ²⁴⁰Pu/²³⁹Pu atom ratios in seawater, sediment, and land soil (Buesseler and Sholkovitz, 1987; Buesseler, 1997; Cooper et al., 2000; Kelley et al., 1999). Seawater and sediment from the North Pacific exhibited a wide range of ²⁴⁰Pu/²³⁹Pu ratios (0.19–0.34), and higher ratios tended to be found near the sites of nuclear tests. In terms of the vertical distribution in seawater, the concentration of ²³⁹⁺²⁴⁰Pu is generally low in surface waters, increases to a maximum at 500–1000 m, and then decreases in deeper waters. Vertical and horizontal distributions, including the GEOSECS data, have also been reported elsewhere (Bowen et al., 1980; Livingston et al., 2001). Lee et al. (2005) reported ²³⁹⁺²⁴⁰Pu activities in sea sediments. A computer simulation of the scavenging and fate of Pu isotopes in seawater was carried out by Periañez (1998).

In addition to the ²³⁸Pu released from nuclear tests and by accident, additional ²³⁸Pu was released when the Transit-5BN-3

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navigation satellite, powered by a SNAP-9A nuclear generator fuelled with ^{238}Pu , failed to achieve orbit and burnt up on reentry at an altitude of about 50 km over the Indian Ocean in 1964. In the Northern Hemisphere, the estimated release of ^{238}Pu was 230 TBq from nuclear tests and 110 TBq from SNAP-9A; the corresponding figures for the Southern Hemisphere were 59 TBq and 380 TBq, respectively (Harley, 1980). The ratio of ^{238}Pu inventory released by nuclear testing to that released from SNAP-9A is estimated to be 2.0 in the Northern Hemisphere and 0.16 in the Southern Hemisphere.

In the East Pacific, there is an oceanic ridge, named the East Pacific Rise, an upwelling region and also high nutrient low chlorophyll region resulting from the poverty of iron. Variations in climatic conditions such as surface temperature occur in the Equatorial Pacific because of the El Niño Southern Oscillation (Tourigny and Jones, 2009). On the other hand, horizontal migration should be uncomplicated because there are few islands to block the oceanic currents. The North Equatorial Current, North Pacific Current, Equatorial Counter Current, South Equatorial Current, California Current, and Peru Current are of importance for surface migration in the Tropical East Pacific.

It was of interest, therefore, to determine how Pu is distributed in the East Pacific, given the conditions mentioned above. If levels of isotopes in a variety of water-mass structures over an extensive region could be compared with each other and with previously reported levels, and if they could be correlated with levels of dissolved oxygen, water temperatures, and water densities, we might be able to understand which parameters affect the migration and distribution of Pu. Here, we report the vertical and horizontal distributions of ^{238}Pu and $^{239} + ^{240}\text{Pu}$ activities as well as the Pu inventory present in samples of seawater and sediment collected in the Tropical East Pacific during 2003, and we discuss our results in relation to material migration in seawater.

2. Experimental

2.1. Sampling

Samples of seawater and sediment from the Tropical East Pacific were collected during the KH-03-1 “Hydra” expedition of the *R/V Hakuho-Maru* in 2003. The locations of the sampling stations, the water depths, and the sampling dates are listed in Table 1, and the locations are shown in Fig. 1. Large-volume water samples (250 L) were collected at various depths between the surface and the bottom by using acoustically triggered quadruple PVC sampling bottles. Additionally, samples of sea sediment with diameter of 7 cm from the surface of the sea bottom to a depth of about 30 cm below the sea bottom were taken by using a multiple corer at locations HY-1, HY-2, HY-3, HY-6, and HY-9. The sediment cores were sliced into 1-cm-thick pieces. Additional samples of seawater (12 L) were collected by using a CTD Carousel multi-sampling (CTD-CMS) system fitted with a dissolved-oxygen (DO) sensor. The water pressure, water temperature, salinity, and DO were measured by devices incorporated into the

Table 1

Location of the stations, the water depths, and the sampling dates.

Station	Location	Water depth	Sampling date
HY-1	20° 00' N 140° 00' W	5309 m	June 28, 2003
HY-2	16° 31' N 123° 00' W	4208 m	July 2, 2003
HY-3	8° 02' N 95° 27' W	3635 m	July 7, 2003
HY-6	0° 01' N 95° 27' W	3219 m	July 10, 2003
HY-9	7° 59' S 95° 01' W	3882 m	July 14, 2003
HY-11	15° 08' S 85° 50' W	4680 m	July 26, 2003
HY-12	20° 00' S 101° 00' W	4114 m	July 30, 2003
HY-15A	25° 00' S 116° 00' W	2867 m	Aug. 2, 2003
HY-17	28° 30' S 127° 47' W	4037 m	Aug. 5, 2003
HY-18	26° 00' S 140° 00' W	4411 m	Aug. 8, 2003

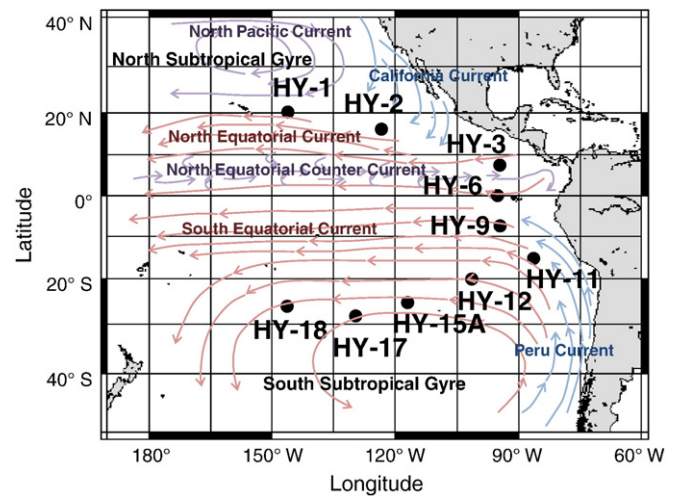


Fig. 1. The locations of sampling stations HY-1 to HY 18 where Pu isotopes were measured.

CTD-CMS. In addition, small portions of seawater samples collected with the CTD-CMS were subjected to analysis for salinity, DO, nitrate, silicate, phosphate, etc.

2.2. Analysis of Seawater

Unfiltered seawater samples (~250 L) were acidified with HCl to a pH of less than 1.5 and spiked with ^{242}Pu tracer and Fe^{3+} carrier. The solution was left to stand for more than 24 h to ensure chemical equilibration, then neutralized with aqueous ammonia to coprecipitate Pu with $\text{Fe}(\text{OH})_3$. The precipitate was separated from the solution and brought back to Japan for subsequent chemical analysis.

The Pu isotopes were analyzed by a procedure similar to that described by Kinoshita et al. (2007). Most of iron atoms were removed from the solution by solvent extraction with diisopropyl ether, and the remaining iron and Pu atoms were precipitated with aqueous ammonia. The precipitate was dissolved in 8 M HNO_3 , and reduced with NaNO_2 to ensure that all the Pu was in the Pu(IV) oxidation state. The solution was then passed through an anion-exchange column of Dowex 1X8, (100–200 mesh), and Th ions were eluted with 8 M HCl. Pu ions were subsequently eluted with 8 M HCl–0.1 M HI solution. This column separation was repeated to purify the resulting Pu solution. The Pu solution was then evaporated to dryness and the residue was dissolved in aqueous ammonium sulfate. The resulting solution was used to prepare an electrodeposited sample on a stainless-steel disk for assay by alpha spectrometry. The typical chemical efficiency of the entire procedure was 50%.

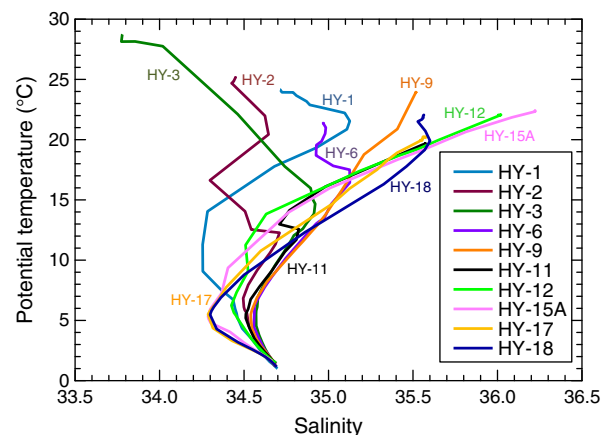


Fig. 2. Potential temperature vs. salinity (θ -S) diagrams for each HY station.

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