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Pacific Proving Grounds radioisotope imprint in the Philippine Sea sediments

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ABSTRACT

Radionuclide concentrations were studied in sediment cores taken at the continental slope of the Philippine Sea off Mindanao Island in the equatorial Western Pacific. High resolution deposition records of anthropogenic radionuclides were collected at this site. Excess ^{210}Pb together with excess ^{228}Th and anthropogenic radionuclides provided information about accumulation rates. Concentrations of Am and Pu isotopes were detected by gamma spectrometry, alpha spectrometry and ICP-MS. The Pu ratios indicate a high portion (minimum of 60%) of Pu from the Pacific Proving Grounds (PPG). This implies that the transport of PPG derived plutonium with the Mindanao Current southward is similarly effective as the previously known transport towards the north with the Kuroshio Current. The record is compared to other studies from northwest Pacific marginal seas and Lombok basin in the Indonesian Archipelago. The sediment core top was found to contain a 6 cm thick layer dominated by terrestrial organic matter, which was interpreted as a result of the 2012 Typhoon Pablo-related fast deposition.

1. Introduction

From 1946 to 1958 the United States conducted an extensive nuclear test programme at Bikini and Enewetak atolls, Marshall Islands (Fig. 1), which were part of the Pacific Proving Grounds (PPG). At the Marshall Islands, 65 atmospheric and underwater tests were performed, many of them with high yield. Approximately 70% of the total yield of all U.S. atmospheric tests were performed at these two atolls, causing a high degree of local and regional fallout. In fact, because the nuclear explosions were performed at or close to ground level or underwater, Bikini and Enewetak account for 96% of local and regional nuclear tests fission yield globally (UNSCEAR, 2000). An estimated 90 TBq of $^{239+240}Pu$ have been deposited in the atoll lagoons' sediments and they represent an important secondary regional source of anthropogenic radionuclides. Plutonium is continuously being released into the open ocean with a rate of approximately 0.2 $TBq \cdot yr^{-1}$ (Lindahl et al., 2010, and references therein).

Another significant plutonium source in the marine environment is the integrated global fallout from nuclear explosions in the atmosphere. The tests injected radionuclide particles into the stratosphere, in which they were dispersed and deposited worldwide (Hamilton, 2005). The first significant insertion of radionuclides into the stratosphere was connected with the 1952 Ivy Mike thermonuclear test. Nuclear testing in the atmosphere continued until a three year moratorium on all nuclear testing between 1958 and 1961, and was resumed until October 1963, when the partial nuclear test ban treaty came into effect. The most intensive years regarding atmospheric testing were between 1961 and 1963, mainly due to a large contribution from the U.S.S.R. high yield tests at Novaya Zemlya. Later fallout from Chinese tests at Lop Nor 1964–1980 and French tests in French Polynesia 1966–1974 contributed comparatively little to the fallout (UNSCEAR, 2000). The estimated $^{239+240}Pu$ inventory in the North Pacific from the global fallout is presumed to be 6 PBq (Lindahl et al., 2010).

Apart from the integrated global fallout and the PPG, an additional source of ^{238}Pu in the sediments in the tropical Pacific is fallout from the navigation satellite SNAP-9A with a ^{238}Pu

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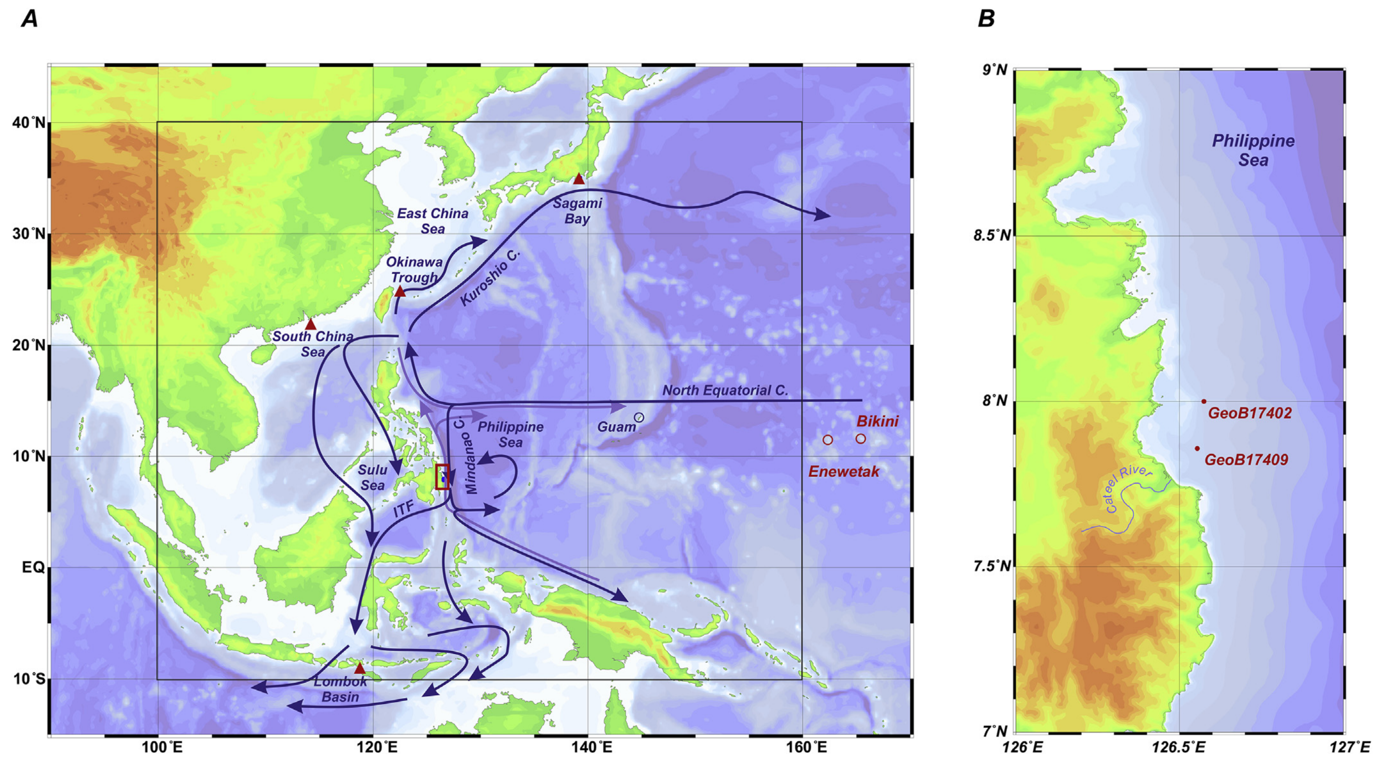

Fig. 1. (A) Overview map of the NW Pacific with the principal currents, plotted after Hu et al. (2015). The study site at the Mindanao continental slope is highlighted by a red rectangle. The previous studies referred to in Section 4.5 are marked by triangles. (B) Detail of the Mindanao east coast with locations of the sampling sites off Cateel, Davao Oriental province. This map was created in Ocean Data View software (Schlitzer, 2016).

radioisotope thermoelectric generator on-board, which burned up in the atmosphere at the altitude of 50 km above the Indian Ocean in 1964 (Krey, 1967; Hardy et al., 1973; Harley, 1980). This introduction of additional ^{238}Pu has changed the ^{238}Pu/$^{239+240}$Pu activity ratios globally. A higher portion, about 78% of the estimated 0.63 PBq of ^{238}Pu, was deposited in the southern hemisphere (Lindahl et al., 2010), where it can be used as a time marker in sediment cores (Hancock et al., 2011). However, in the latitudinal band of 0–10°N, a negligible contribution from this source is expected (Hardy et al., 1973).

Artificial radionuclides remain in the Pacific Ocean water column many years after their initial arrival, and their distribution is characterized by a subsurface maximum at depth between 500 and 1000 m (Povinec et al., 2003). Availability of particles near the continental margin due to higher productivity and suspended particle load, supports particle-reactive radionuclides (^{241}Am and plutonium isotopes) scavenging from the water column and their incorporation in the sediment (León Vintró et al., 2005). Conversely, depth distribution of artificial radionuclides in fast depositing anthropogenic sediment layers can provide information about changes of radioisotope concentration in the overlying water column in time, and activity and atom ratios of anthropogenic radionuclides contain information about their origin. Inventories ($Bq \cdot m^{-2}$) of these transient signals relative to steady state tracers like ^{210}Pb thus provide information on the magnitude of the sediments as sinks for a particular source. Elsewhere, plutonium isotopes have successfully been used as chronometers in Anthropocene sediment cores (Corcho-Alvarado et al., 2014).

Bikini and Enewetak atolls are located along the pathway of the North Equatorial Current (NEC), a westward moving major current, forming the southern boundary of the clockwise northern subtropical gyre. The NEC reaches the continental margin in the Philippine Sea and bifurcates into the northbound Kuroshio Current (KC) and the southbound Mindanao Current (MC). This current splitting takes place about 14°N at the surface and 20°N at a depth of 1000 m (Hu et al., 2015). Qu and Lukas (2003) estimated the average partition of MC and KC as 31.4 Sv[1] and 25.4 Sv, respectively, while large inter-seasonal variations were observed. The MC therefore carries North Pacific water masses southward, partially diverging them towards the Indonesian Throughflow. Below the thermocline, the Mindanao Undercurrent transports water of South Pacific origin northward. This poleward transport however happens in a greater distance from the Mindanao coast; within 50 km from the shore the transport in southward direction occurs up to a depth of 1000 m (Schönau et al., 2015).

Plutonium isotopes are reportedly being distributed from the PPG by the NEC and the KC towards the Pacific Ocean marginal seas, particularly in the East China Sea (Wang et al., 2017; Tims et al., 2010; Wang and Yamada, 2005; Lee et al., 2004) and along the Japanese west coast (e.g., Zheng and Yamada, 2004). Clear PPG signals are also reported in sediments from the Sulu and South China Seas (Dong et al., 2010; Wu et al., 2014). Recently, the PPG derived transuranics were found in the sediments of the Lombok basin off the Indonesian island of Sumba (Steinke et al., 2014; Pittauer et al., 2017). The transport likely follows the pathway southward with the Mindanao Current, and further through the Maritime Continent.

Up to this date, artificial radionuclide deposition history and the PPG effect has not been explored in the Philippine Sea, in the area south of the NEC bifurcation. Within this study we aim to examine the sources of artificial radionuclides in the continental slope sediment cores off the Philippine southern island of Mindanao

[1] A non-SI unit of volume transport Sverdrup (Sv; 1 Sv = 10^6 m^3s^{-1}) used in oceanography; not to be confused with an SI unit of radiation dose, Sievert (also Sv).