# Isotopic evidence for the sources of Pu in the sediments collected from Sagami Bay, Japan

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In order to understand the fate of Pu in the ocean, Pu in Pacific sediments should be studied because Pacific sediments are believed to be the ultimate sink for Pu present there. Several studies on the Pu in Pacific sediments have been reported, especially shelf and slope studies on both eastern and western Pacific margins. On the eastern side, a comprehensive review about the work on the California basin sediments was presented by Sholkovitz (1). Beasley et al. (2) reported on transuranic measurements in shelf sediments off Washington and Oregon. On the western side, our laboratory has made a series of marginal studies on Pu in sediments in the East China and Yellow Seas and in the Seto Inland Sea. Also, Yamada and Nagaya (3-4) investigated 239,240Pu and 137Cs in sediments from Sagami Bay and Tokyo Bay. All these studies reached the same major conclusion that the Pacific marginal sediments underlying waters with high biological productivity and larger suspended sediment loads are major sinks for Pu removal and sequestration. In addition, general excess inventories of Pu over those anticipated from global fallout were observed. The above mentioned studies drew conclusions mainly based on the total activity of 239,240Pu and the activity of 137Cs; the source of the additional Pu input in these marginal sediments, however, remained unknown because of the lack of a distinctive 240Pu/239Pu signature.

Information on Pu isotopic compositions in sediments is very useful in understanding the source of Pu present there since the 240Pu/239Pu ratio has proved to be a powerful fingerprint to identify the Pu contamination. The atomic ratio of 240Pu/230Pu in fallout may vary, depending upon the specific weapon design and test yield. The global fallout average 240Pu/239Pu atom ratio is 0.18, based upon atmospheric aerosol sampling, soil samples and ice core data. It is also recognized that different test series can be characterized by either lower or higher ratios. For example, fallout from the Nevada test site has a lower 240Pu/239Pu ratio, averaging 0.035 (5), while elevated 240Pu/239Pu ratios (0.21-0.36) in fallout from the 1950s from the Pacific Proving Grounds were reported (6). Recently, we have developed a highly sensitive method for the determination of 240Pu and 239Pu in marine sediment samples by means of ICP-MS (inductively coupled plasma mass spectrometry) with a shield torch system. In the present study, we have applied this method in order to determine 240Pu and 239Pu and their ratios in sediment core samples collected in Sagami Bay. Based on the 240Pu/239Pu ratio signature, we identified the additional Pu input in Sagami Bay was from the transported close-in fallout from Bikini and Enewetak nuclear weapon tests. We demonstrate that Pu activity and isotopic ratio profiles of marine sediment cores contain records of global fallout and close-in fallout Pu, which provide chronological information on the recent sedimentation. We propose that the transport route is through the oceanic currents, e.g. the North Equatorial Current and Kuroshio Current. Using a two fallout end-member model, we resolved the relative contribution of Pu between global fallout and close-in fallout in Sagami Bay sediments.

References

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