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Pu-239 and Pu-240 inventories and Pu-240/ Pu-239 atom ratios in the water column off Sanriku, Japan.

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A magnitude 9.0 earthquake and subsequent tsunami occurred in the Pacific Ocean off northern Honshu, Japan, on 11 March 2011 which caused severe damage to the Fukushima Dai-ichi Nuclear Power Plant. This accident has resulted in a substantial release of radioactive materials to the atmosphere and ocean, and has caused extensive contamination of the environment. However, no information is available on the amounts of radionuclides such as Pu isotopes released into the ocean at this time. Investigating the background baseline concentration and atom ratio of Pu isotopes in seawater is important for assessment of the possible contamination in the marine environment. Pu-239 (half-life: 24,100 years), Pu-240 (half-life: 6,560 years) and Pu-241 (half-life: 14.325 years) mainly have been released into the environment as the result of atmospheric nuclear weapons testing. The atom ratio of Pu-240/Pu-239 is a powerful fingerprint to identify the sources of Pu in the ocean. The Pu-239 and Pu-240 inventories and Pu-240/Pu-239 atom ratios in seawater samples collected in the western North Pacific off Sanriku before the accident at Fukushima Dai-ichi Nuclear Power Plant will provide useful background baseline data for understanding the process controlling Pu transport and for distinguishing additional Pu sources. Seawater samples were collected with acoustically triggered quadruple PVC sampling bottles during the KH-98-3 cruise of the R/V Hakuho-Maru. The Pu-240/Pu-239 atom ratios were measured with a double-focusing SF-ICP-MS, which was equipped with a guard electrode to eliminate secondary discharge in the plasma and to enhance overall sensitivity. The Pu-239 and Pu-240 concentrations were 2.07 and 1.67 mBq/m3 in the surface water, respectively, and increased with depth; a subsurface maximum was identified at 750 m depth, and the concentrations decreased with depth, then increased at the bottom layer. The total Pu-239+240 inventory in the entire water column (depth interval 0-bottom) was 69.8 Bq/m2. This was significantly higher than the expected cumulative deposition density of atmospheric global fallout. The Pu-240/Pu-239 atom ratios were 0.22 in the surface water and increased gradually with depth reaching 0.26 at the 5000 m depth. The obtained Pu-240/Pu-239 atom ratios were higher than the mean global fallout ratio of 0.18. These high atom ratios proved the existence of close-in tropospheric fallout Pu from the Pacific Proving Grounds in the Marshall Islands.