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TITLE: Plutonium in the western North Pacific: Transport along the Kuroshio and implication for the impact of Fukushima Daiichi Nuclear Power Plant accident

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ABSTRACT:

This study examined Pu source terms in the western North Pacific (WNP) based on data collected in 2014 and 2015. The basin wide $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios ranged from 0.227 to 0.263 with an average value of 0.244 ± 0.011 , consistently higher than that of global fallout (~ 0.180). The spatial distribution of $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios showed higher values within the Kuroshio region, the main western ocean boundary current, as compared to the zone off of the Kuroshio. There was also an overall decreasing trend of $^{240}\text{Pu}/^{239}\text{Pu}$ along the Kuroshio path to its extensions. $^{239}+^{240}\text{Pu}$ activities in surface seawater exhibited a wide range from 1.15 to 4.30 mBq m⁻³ and their spatial distribution showed an increasing trend with latitude. Unlike the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios, which had heavier isotopic compositions in the Kuroshio mainstream compared to the zone off of the Kuroshio, the $^{239}+^{240}\text{Pu}$ activities were higher outside the Kuroshio than within the Kuroshio. These patterns in both Pu isotopic ratios and activities point towards a unique close-in fallout source, which levels down in its source term and has a high degree of scavenging during its transport along the Kuroshio, and can be traced back to a precursor, the North Equatorial Current, which originates near the Pacific Proving Grounds (PPG) with characteristically higher $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios. High $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios found in the zone outside the Kuroshio were transported from the Kuroshio via the formation and circulation of North Pacific Intermediate Water. We further revealed, using a simple two end-member mixing model, that the PPG source contributed $60 \pm 13\%$ of the Pu in the Kuroshio zone and $45 \pm 10\%$ in the zone off of the Kuroshio. Both the comparison of Pu isotopic composition in the WNP within a radius of 500 km or 1000 km off the Fukushima Daiichi Nuclear Power Plant (FDNPP) between prior to and post the accident and a simple first order mass balance calculation in terms of atmospheric deposition and release suggest that the Pu originating from the FDNPP accident, if any, was either negligible, or the input flux of $^{239}+^{240}\text{Pu}$ was too small to significantly alter the Pu isotopic composition in the ambient seawater. This article is part of a special issue entitled: 'Cycles of trace elements and isotopes in the ocean: GEOTRACES and beyond' - edited by Tim M. Conway, Tristan Horner, Yves Plancherel, and Aridane G. González.

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