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Tracing the dispersion of contaminated sediment with plutonium isotope measurements in coastal catchments of Fukushima Prefecture

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The Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident led to important releases of radionuclides into the environment, and trace levels of plutonium (Pu) were detected in northeastern Japan. However, measurement of Pu isotopic atom and activity ratios is required to differentiate between the contributions of global nuclear test fallout and FDNPP emissions. In this study, we measured Pu isotopic ratios in recently deposited sediments along rivers draining the most contaminated part of the inland radioactive plume. To this end, we carried out a thorough chemical purification and concentration of Pu from sediment samples (5 g dry material) and precise isotopic measurements using a double-focusing sector field ICP-MS. Results showed that the entire range of measured Pu isotopes (i.e. ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu) were detected in all samples, although in extremely low concentrations. The $^{241}\text{Pu}/^{239}\text{Pu}$ atom ratios measured in sediment deposits (0.0017–0.0884) were significantly higher than the corresponding values attributed to the global fallout (0.00113 ± 0.00008 on average in the Northern Hemisphere between 31° – 71°N). The results indicated the presence of Pu from FDNPP, in slight excess compared to the Pu background from global fallout, representing up to ca. 60% of Pu in the analyzed samples. These results demonstrate that this radionuclide has been transported relatively long distances ($[^{239}\text{Pu}/^{241}\text{Pu}]$ 45 km) from FDNPP and deposited in rivers representing a potential source of Pu to the ocean.