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

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The Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident led to the formation of a radioactive pollution plume in soils of this region of Northeastern Japan. Although radiocesium (^{134}Cs and ^{137}Cs) is currently the most abundant radionuclide, trace levels of plutonium (Pu) have been detected in soil, sediment and vegetation samples collected in the vicinity of FDNPP. A previous study [1] quantified the ^{241}Pu/^{239}Pu activity ratio in sediment (n=5) transiting the coastal rivers of the region and demonstrated that Fukushima-originating Pu could been transported over relatively long distance (~45 km) from FDNPP. However, spatial and temporal variations of Pu signatures in sediment had not been determined yet.

Here, we used a Multi-Collection ICP-MS to detect Pu isotopes (i.e. ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu) and quantify its atom ratios and activity in flood sediment deposits (n=11) collected between November 2011 and November 2014 along the Niida River, draining a severely contaminated part of the inland radioactive plume.

The ^{241}Pu/^{239}Pu atom ratios measured in sediment decreased significantly from 2011 (e.g. ^{241}Pu/^{239}Pu = 0.07980±0.00041), to 2014 (0.01404±0.00020) , when they reached values close to those reported for the global fallout in the Northern Hemisphere (0.00167±0.0003 [2]). Accordingly, Pu found in this sediment was composed of a mix of Pu from FDNPP and from global fallout. Fukushima-originating Pu contribution decreased from 50% in 2011 to 10% in 2014. The results confirm the quick export of sediments from Fukushima coastal catchments to the Pacific Ocean. They also likely reflect the effectiveness of decontamination works that started in 2013 in this catchment. Future investigations could use the Pu atom ratios to further trace the impact of these remediation works on the evolution of radionuclide concentrations found in sediment transiting Fukushima coastal rivers.

[1] Evrard, O., et al., *Environ Sci Technol*, 2014. 48(16): p. 9334-40. [2] Zhang, Y., et al., *Science of The Total Environment*, 2010. 408(5): p. 1139-1144.