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Sources of plutonium to the tropical Northwest Pacific Ocean since the mid-20th century: a natural coral archive

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Patric Lindahl[1], 浅海 竜司 [2*], 井龍 康文 [3], Paul Worsfold[1], Miranda Keith-Roach[1], Min-Seok Choi[4]
Patric Lindahl[1], Ryuji Asami[2*], Yasufumi Iryu[3], Paul Worsfold[1], Miranda Keith-Roach[1], Min-Seok Choi[4]

[1]University of Plymouth, UK, [2] 琉球大学・超域, [3] 名古屋大学・院環境, [4]KBSI, Korea
[1]University of Plymouth, UK, [2]University of the Ryukyus, Japan, [3]Nagoya University, Japan, [4]KBSI, Korea

The main source of plutonium (Pu) to the Pacific Ocean is fallout from atmospheric nuclear weapons testing between 1945 and 1980. Other sources (e.g. discharges from the nuclear industry, radioactive waste dumping and burn-up of nuclear powered satellites) have only made a relatively minor impact. Between 1945 and 1980, 543 atmospheric weapons tests were carried out worldwide with an estimated total released fission energy yield of 189 Mt (UNSCEAR, 2000). Due to the well-defined spatial and temporal inputs of Pu and its physical and chemical properties, Pu can be used a tracer for various oceanic processes such as water mass transport, particle fluxes and scavenging (Lindahl et al., 2010 Mar. Environ. Res.). Massive Porites corals, living in tropical/subtropical shallow waters, have annually-banded aragonite skeletons with high- and low-density increments and grow rapidly, which can provide chronological control and allow high-resolution sampling. Because of such benefits, geochemical composition in coral skeletons has been most widely used as paleoclimate proxies for temperature and chemical composition of seawater in many studies. Plutonium is incorporated in the coral skeleton during growth with a constant relationship between Pu in the coral and Pu in the surrounding seawater. Therefore, the historical Pu signal in the surrounding seawater can be reconstructed by analyzing well-dated coral cores

Here we determine the total Pu activity concentrations and Pu atom ratios in the annual growth bands of an accurately dated modern coral core from Guam Island using multi collector inductively coupled plasma mass spectrometry (MC-ICP-MS) to reconstruct the 1943-1999 Pu records in the tropical Northwest Pacific (Lindahl et al., 2011 Geochim. Cosmochim. Acta). The coral chronology was established in previous works (Asami et al., 2004 Palaeogeogr. Palaeoclimatol. Palaeoecol.; 2005 J. Geophys. Res.), in which coral d18O was determined with high-temporal resolution (biweekly to monthly) over the period 1787-2000. Dating of the coral was based on visual observations of soft X-radiograph images, which showed well-developed annual high- and low-density skeletal growth bandings. Selections relating to annual skeletal growth increments (approximately from January to December) were then assigned. The Pu atom ratios in the coral bands were used to distinguish the source(s) of Pu contamination and characterize the isotopic input from notable nuclear tests. Close-in fallout from the former US Pacific Proving Grounds (PPG) in the Marshall Islands and global fallout were identified as the two main sources. The Guam site was dominated by PPG close-in fallout in the 1950s. In addition, a higher Pu atom ratio was observed that could be attributed to fallout from the Ivy Mike thermonuclear detonation in 1952. The atom ratio decreased in the 1960s and 1970s due to increase in the global fallout with a low Pu atom ratio. Recent coral bands (1981-1999) are dominated by the transport of remobilized Pu, with high Pu atom ratios, from the Marshall Islands to Guam Island along the North Equatorial Current. This remobilized Pu was estimated to comprise 69% of the total Pu in the recent coral bands, although its contribution was variable over time.
Keywords: coral skeleton, annually-dated bands, plutonium, tropical Pacific Ocean, nuclear weapons test, North Equatorial Current