

BPT002-P01

Room:Convention Hall

Time:May 25 14:00-16:30

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

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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  $\delta^{18}O$  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

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## Variation of expression of $\text{Ca}^{2+}$ -ATPase and skeletal growth of cultured corals (*Porites australiensis*)

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Trace elements, such as Sr/Ca and Mg/Ca ratios, of coral skeleton is a commonly used for reconstructing paleoenvironment, especially it has been used to estimate past sea surface temperatures. However the processes controlling trace elements incorporation in coral aragonite are poorly understood and recently it has been reported that skeletal growth might be altered variation of trace element rather than temperature. In zooxanthellate corals,  $\text{Ca}^{2+}$  is transported transcellularly to the calcification site by both calcium channels and by the carrier protein  $\text{Ca}^{2+}$ -ATPase. The pump enzyme  $\text{Ca}^{2+}$ -ATPase has a higher affinity for  $\text{Ca}^{2+}$  than for  $\text{Sr}^{2+}$ , and the  $\text{Ca}^{2+}$ -ATPase pump is activated by exposure of the polyp to light, suggesting that the skeletal Sr/Ca of rapid calcifiers might be lower than that of slow calcifiers because of differences in the Sr/Ca ratio in the bulk calcifying solution. However, there has been no study investigating the relationship between  $\text{Ca}^{2+}$ -ATPase and Sr/Ca and Mg/Ca in the skeleton of *Porites* coral which is dominantly used for the study on paleoclimate. In this study, *Porites australiensis* were cultured under controlled temperature and pH settings and expression level of  $\text{Ca}^{2+}$ -ATPase in each cultured corals was estimated. Also Sr/Ca and Mg/Ca ratios in the same coral colony were analyzed. We will discuss the relationship between skeletal growth rate and expression of  $\text{Ca}^{2+}$ -ATPase.

Keywords: coral skeleton,  $\text{Ca}^{2+}$ -ATPase, growth rate, culture

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## Biological controls on Sr/Ca, Ba/Ca and Carbon isotope in freshwater pearl mussel *Hyriopsis* sp.

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Biogenic skeletons of marine and freshwater organisms are important archives of paleoenvironmental information. Bivalves occur in various environments from polar region to low latitude region, in freshwater and marine, and have carbonate shells which are formed by addition growth. So they have the potential to contain the paleoenvironmental information within their isotope and elemental compositions. Freshwater environment sensitively reacts to the climate changes and is well known as a role of the material transportation from continental regions to seawater following chemical and physical weathering. So freshwater bivalves gather much attention as time-series paleoenvironmental archives. By this time, there are many reports on elemental and isotope fractionation of marine bivalves, but little is known about freshwater bivalves.

In this study, we analyzed the trace element (Sr/Ca and Ba/Ca) and stable isotope (oxygen and carbon) ratios of a commercially cultured freshwater pearl mussel *Hyriopsis* sp. shells (*Bivalvia Unionidae*) and ambient water in Lake Kasumigaura. Our aim was to assess what is the factors influencing Sr/Ca, Ba/Ca, and carbon isotope ( $d^{13}C_{SHELL}$ ) ratios freshwater bivalve shells.

Element ratios (Sr/Ca and Ba/Ca) in the three shell specimens were measured with a quadrupole ICP-MS and stable isotope ratios ( $d^{18}O_{SHELL}$  and  $d^{13}C_{SHELL}$ ) with an isotope ratio mass spectrometer. Shell structures were observed by the microscope and by inspection of the external shell. Trace element ratios (Sr/Ca and Ba/Ca) of water samples were calculated from the Ca, Sr and Ba concentrations, which were measured with an ICP-AES.  $d^{13}C_{DIC}$  was measured by isotope ratio mass spectrometer. We compare the element/isotopic profiles of shells among various parameters such as water temperature, annual growth rate, and elemental/carbon isotope ratios of ambient water.

Sr/Ca ratios of water remained relatively constant throughout years. Clear annual Sr/Ca cycles of shell suggest that Sr was incorporated into the shells at a high rate during warm seasons. If the Sr incorporation is governed by thermodynamic control, Sr/Ca profiles show a negative correlation with ambient temperature. However, between Sr/Ca and  $d^{18}O$  profiles negative correlation is found. That indicates primary controlling factor of a Sr incorporation into *Hyriopsis* shell is not thermodynamic partition. All three examined shells showed a clear decrease in annual average Sr/Ca with age. Inter- and intra-annual trends suggest that Sr incorporation was higher during periods of faster growth.

The Ba concentration and the Ba/Ca ratio of ambient water showed neither an annual periodicity nor a seasonal trend. In contrast to the findings of an inorganic aragonite precipitation experiment, Ba/Ca was positively correlated with ambient temperature. In addition, Ba/Ca and Sr/Ca ratios were positively correlated with each other. These results suggest that not thermodynamic equilibrium partitioning but the shell growth rate controls Ba incorporation into *Hyriopsis* shells.

The  $d^{13}C_{DIC}$  showed a wide range fluctuation from -14.63 to -8.62 per mill. The enrichment of  $d^{13}C$  was observed in summer samples. On the other hand, the  $d^{13}C_{SHELL}$  varied mainly from -12 to -9 per mill and its pattern was not symmetric and sinusoidal like the water temperature profile. A carbon isotopic mixing equation presented by McConnaughey et al. [1997] describes the combination of respired and environmental carbon incorporated into the precipitating carbonate. The calculated values of  $d^{13}C_{SHELL}$  under equilibrate with  $d^{13}C_{DIC}$  are ranged from -11.93 to -9.41 per mill and are consistent with observed shell data. Thus, the primary factor controlling the value of  $d^{13}C_{SHELL}$  is not the respired effects but the carbon isotope fluctuations of ambient DIC.

\*McConnaughey, T. A., J. Burdett, J. F. Whelan, and K. P. Charles (1997), *Cosmochim. Acta*, 61, 611-622.

Keywords: freshwater bivalve, Sr/Ca, Ba/Ca, Carbon isotope