



Geochronology of Cadmium (Cd), Cuprum (Cu), and Arsenics (As) in Annual Band of Coral *Porites lutea* at Pantai Kondang Merak, Malang

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Abstract

Coral reef is a massive natural building block that mainly composed of hard coral. In ecological view coral reef is the center of biological activity for shelter, foraging and feeding place. *Porites lutea* is a common coral in reef flat area such as in Pantai Kondang Merak. This coral can form a massive with hillocky colony surface and has a slow linear extension rate, that made this coral has long longevity. The annual growth band of coral skeleton provide information of pollutants in the coastal extending back over several years ago due to ability of coral aragonite traped trace metal from environment. The aim of this research was to determine the concentration of trace metals of Cd, Cu and As along with the coral annual banding. The heavy metals detection was performed using ICP-OES (iCAP 7400 Series). The concentration of Cd, Cu and As in sample 1 (KM1) were 2.236 mg/kg, 9.726 mg/kg, and 2.474 mg/kg, while sample 2 (KM2) were 1.989 mg/kg, 19.157 mg/kg, and 2,064 mg/kg respectively. Two ways mechanism of trace metals to be trapped in the coral skeleton are by direct mechanism when trace metals in a form dissolved ion that were uptaken by coral then stored into coral skeleton and by indirect mechanism when particulate metals ingested by plankton then eaten by coral through coral tissue. The tracing of heavy metal in coral is provided important information of environment condition of the sea from 2009 - 2015 that may be used for authority decision regarding pollutant ambient in the sea environment.

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INTRODUCTION

Coral reef provides a basic need for coral reef fauna (shelter and foraging) so it becomes a primary producer in the ocean. Recently, coral reef ecosystem was threatened by global climate change, bleaching, disease and pollution (Beyer et al., 2018; Safaei et al., 2018). The massive *Porites lutea* has been well recognized as a biomonitor for time-series studies of geochronology or trace-back of marine condition from the past up to now. Many variations of sea properties (temperature), organic and inorganic materials even many trace minerals influence of coral growth. All valuable information lied and recorded on coral annual growth band in *P. lutea* that can be proxy indicators (Erler et al., 2016; Krishnakumar et al., 2015; McCutcheon et al., 2015). Heavy metals have got serious attention because of their hazards, endurance and high potential to accumulate on various marine biota.

Pantai Kondang Merak, like the other coasts in southern Java was surrounded by mountain and identified as flood area during west monsoon. During that time, heavy rainfall brought massive water and sediment load from the upper area resulted in the reduced salinity of seawater, an increase in suspended solid sediment and severe coral reef living inside. Scleractinian coral that live submerged in reef flat area may stress during a flood event. Sediment as storage and resource of heavy metals play important role of metal movement and transformation. High load sediment discharged from terrestrial allowed heavy metals also ran into the marine environment, (Martuti et al., 2019; Pourabadehei & Mulligan, 2016).

Several various natural ways of heavy metals to enter the marine environment are by rock erosion, leaching of soil, plant life and volcano eruption. Moreover, several anthropogenic activities that also have a role including water waste, industrial waste, fish pond and also agriculture (Burakov et al., 2018; Marrugo-Negrete et al., 2017; Zaborska et al., 2017). These metals are bound in coral aragonite by coral feeding. Plankton contained heavy metals are captured by a coral tentacle or trapped on coral mucus. After ingested by coral, the heavy metals still remaining in coral and bound into the organic matrix of the coral skeleton (Amir & Mohamed, 2017).

Copper (Cu) with atomic number of 29 is widely used for a conductor of electricity, herbicides, and antifouling in the paint. Copper is an essential trace mineral for almost all organism but the high level concentration of Cu in coral

will affect on the photosynthesis process of coral symbionts, disturb the fertilization and give negative impact on coral metabolism (Al-Harahsheh et al., 2015). Cadmium (Cd) is an element with atomic number 48 that mostly found in sulfide mineral. This metal is used for electroplating, paint pigments, and alloy. For biota, Cd has a role in metallothioneins activity and hence disrupts the process of internal chemical and physical body conditions. Cadmium in the seawater caused by upwelling events where the stored cadmium in the sea floor (sediment) brought to the surface and enriched of seawater (Auger et al., 2015). Arsenic (As) is abundant and mobile metalloid in the environment, where naturally occurred from earth crust. This metal cycle is started from dust that was resulted from weathering rocks and then dissolution in water by rain. The occurrence of this metal usually associated with mud, hydroxides and some organic materials. The anthropogenic sources of this metal were from rodenticides, pesticides, and herbicides that all used for agriculture purposes (Mirlean et al., 2016).

Considering the importance to monitor the marine pollutant in Pantai Kondang Merak with an accurate way through trace mineral accumulation in two colonies of *P. lutea* this research becomes first time report of coral reef health condition at south Java sea. The main objective of this study was to determine of Cu, Cd and As in coral skeleton as useful method for interpreting the past environmental condition. .

Coral reef under the threats from many factors one of them is heavy metal pollutant. Along increasing number of pollutant from natural and human in Pantai Kondang Merak, that had affected on deterioration of coral health. Incorporate heavy metals in coral aragonite is tools to analyse and monitor the heavy metals contamination in the sea water. Furthermore, more advance technic such as isotopic work on these trace minerals should also useful tracing the sources of them. In order coral skeleton can give extensive record of heavy metals history in this location, it will become valuable data for policy maker regarding setting a heavy metals threshold regulation.

METHODS

Study area and research location

In May 2015, two colonies of *P. lutea* were collected from two sites in Pantai Kondang Merak, South Malang (Figure 1). Trace metals analysis from coral annual banding of *P. lutea*, was carried out from August to September 2016 at the Marine, Industrial and Environmental

Laboratory, Isotope and Radiation Application Center (PAIR), National Nuclear Energy Agency (BATAN) Pasar Jumat, South Jakarta.

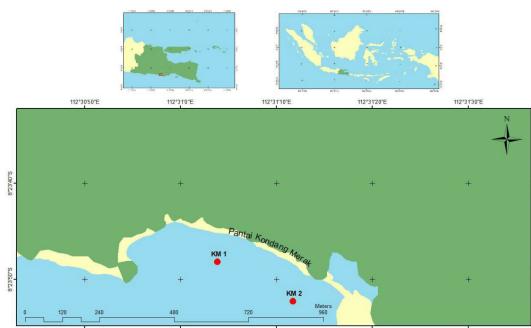


Figure 1. The research locations are marked with round tips in Pantai Kondang Merak, Malang

Data collection

Coral colonies (KM1 and KM2) were obtained from reef flat area of the Pantai Kondang Merak, with 1-3 m depth. Each coral colony with 16-18 cm in diameter were taken by a handspike then kept alive in a cool box with seawater. Prior to analysis, the colonies were soaked under five percent hypochlorite solution (2 l) in a plastic chamber. After 24 hours of coral colonies washing with tap water in the Marine Science University of Brawijaya's lab, if the coral tissue was not removed completely, samples would be soaked under bleach solution. After all, samples tissues were removed then rinsed in distilled water and dried at 60 °C for 72 hours (Krishnakumar et al., 2015). Each sample then was cut into 1 cm perpendicular to the perceived growth direction using a masonry saw with a 1-m diameter (Barnes & Lough, 1999).

Sample preparation

The coral slab imaged with X-ray photography resulted in a positive photo of the annual growth ring of coral. The positive image then was scanned and printed to guide of the skeleton. The coral slabs were washed using DDW and then soaked into 0.2 N HNO₃ or nitric acid and hydrogen peroxide (H₂O₂) on the ultrasonic bath to remove all tissue and organic materials. Sub-sampling was started from the top to downward of coral annual banding that representing of coral growth from recent to past. Using a dentist driller to collect about 20 mg weight of coral skeleton powder, all samples were placed into a 15 ml vial bottle and dissolved into 25% nitric acid (HNO₃) and 5% HCl solutions. All solvents were vigorously mixed with a Vortex® for 10 seconds, and the last samples were centrifuged at 2000 rpm for 30 minutes before analyzed at ICP-OES (iCAP 7400 Series).

Trace metals determining

The operating condition of ICP-OES before analysis of trace metal Cd were purging for 2 hr, the cooling water was turned on and the trace elements (Cu, Cd, and As) were inserted. After all the indicator lights were turned green on the Qtegra software dashboard, ICP-OES could be used. To avoid error message from this machine, the warming up time should be not less for 15 minutes. Standard solutions and samples placed on an autosampler rack then measured using different intensity wavelength each heavy metals as follow: Cd 228.802, Cu 327.396, and As 193.759.

Trace metals concentration

The concentration of each heavy metal was calculated based on the sample weight tested to determine the actual concentration of metal elements based on the formula by Hutagalung (1997) as follows:

Note:

M = Heavy metal concentration in the sample (mg/ kg)

C = Concentration obtained from the calibration curve (ppm)

V = Volume of sample solution (ml)

B = sample weight (gram)

RESULTS AND DISCUSSION

Skeleton X-ray

KM1 coral sample (Figure 2A) was 6 years old, if calculated backward from 2015 to 2010 and has an average growth rate of 1.09 cm/yr (Table 1). While the KM2 coral sample (Figure 2B) was 1 year older than KM1 and this coral had lived for 7 years start from 2015 to 2009 and has an average growth rate of 1.74 cm/yr (Table 1). The growth rate of *P. lutea* is normal with average growth rate ranging from 1.09 - 1.74 cm/year.

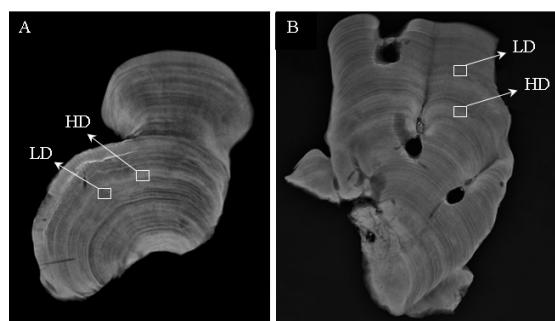


Figure 2. The result of X-ray on coral *P. lutea* skeleton, A) KM1; B) is KM2. HD: high density; LD: low density

Table 1. The growth rate of *P. lutea* each year in Pantai Kondang Merak

Year	Annual growth of <i>P. lutea</i> (cm/year)	
	KM 1	KM 2
2009	-	1.87
2010	1.47	1.39
2011	0.91	1.22
2012	1.31	2.17
2013	1.1	1.86
2014	0.92	1.6
2015	0.84	2.09
2016	-	-
Average	1.09	1.74

The concentration of trace minerals in *P. lutea*

The concentration of heavy metals from KM1 sample showed the heavy metal Cd was 1.442 - 2.912 mg/ kg and the highest concentrations were in 2011 and 2013. Then the concentration of Cu was 8.7 - 10.67 mg/ kg and the highest concentration was in 2013, while As concentration was 0.97 - 3.846 mg/ kg and the highest concentration was in 2012 (Figure 3A). Similar pattern was found in KM2 sample where Cd concentration (0.961 - 3.448 mg/ kg) was lower than Cu (12.5 - 30.288 mg/ kg) and As had the lowest concentration of 0.469 - 2.955 mg/ kg. Figure 3B also shows the peaks of each heavy metals within years which the highest concentration of Cd, Cu, and As was in 2012, 2015, and 2012 respectively.

The three heavy metals. (Cd, Cu, and As) showed a different range of concentration in *P. lutea* skeleton. The concentration of heavy metals absorbed in coral skeleton each year also have been showed different concentration and different with other biota such as plant, bivalve and human that stored and accumulated of heavy metal in their tissue. Because of this, the geochronical (history) of pollutants cannot be traced back Jafarabadi et al. (2018) explained that the absorption of heavy metals in coral skeleton is the lowest compared by their tissue and zooxanthellae. This is because coral skeleton had more intragranular porosity that made the amount of trace metals released through a porous structure (Jiang et al., 2017). Bioaccumulation process of heavy metals in coral skeleton depends on biomineralization process on endodermis layer that took a long time process (Akiva et al., 2016). Lastly, coral has a selective mechanism of heavy metals absorption in which not all types of metals can be transported from coral tissue into a coral skeleton

(Darvishnia et al., 2016).

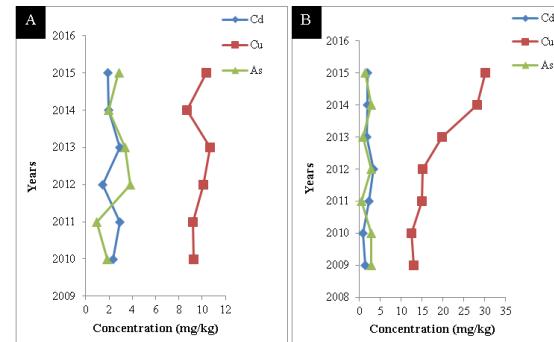


Figure 3. Graph of trace metals Cd, Cu and As in KM1 (A) and KM2 (B) samples taken from growth annual banding of *P. lutea*.

The higher concentration of Cd and As in 2012-2013 seems to be related to upwelling Kelvin wave (uKW) in June 2012 at the eastern part of Indian Ocean (Baumgart et al., 2010; Dey et al., 2018). Cadmium is similar with others in organic material that sinking and accumulating to the sea bed through a biological process. Cadmium is similar to phosphorous that depleted into the water column due to biological activity through upwelling process (Matthews et al., 2008). Rich of plankton during upwelling is suggested to have a strong correlation with the skeletal extension of stony coral beside another factor such as coral metabolism, salinity, temperature, and light. Naturally, the number of arsenic in the ocean was 20-40 nM, a very small amount. Besides, from the mining process (gold mining) As can be resulted from a hydrothermal vent that was commonly found in the Indo-Pacific area (Price et al., 2013). The stable arsenate is As (V) and other is arsenic As (III) which often be found in nature. During upwelling event, mass cooler deep water raised toward sea surface brought nutrient and contained Cd-As sedimentrich .

The mechanism of trace metal cadmium bound in coral skeleton could be in several ways, such as through a biological process, the trace metal has possibly been contained in the plankton (food) and trapped in the skeletal cavity due to undigested food. ; the substitution of trace metal during skeletal forming in endodermis cell and kept in the crystal lattice; and absorbed by coral tissue (Al-Ouran, 2005; Al-Rousan et al., 2007; Jayaraju et al., 2009). While As can be the uptake in two ways: cell diffusion and trophic transfer (Price et al., 2013).

Phosphate in organic form (Pi) is an essential mineral to a living organism, it has a key role for the building block of protein and DNA, me-

tabolism of a cell, and build the skeletal mineralization (Spina et al., 2013). Arsenate chemically has a similar compound to the phosphate, some time coral taking the wrong substances such as As(V) or As(III) for specific transporter in the cell wall (Price et al., 2013). Two organoarsenic biotases detected in natural water are dimethylarsinate (DMA) or methylarsonate (MA). Marine algae including plankton sometimes contain DMA and MA. The source of As accumulated in coral skeleton suggested from diet trophic level (Ohki et al., 1996).

Copper (Cu) had the highest concentration of heavy metals in *P. lutea* skeletal, that was 5 times fold than Cd and As. High concentration of Cu in coral skeleton because Cu as essential trace element for coral that was required for metabolism process as enzyme catalyst, inhibition factor of oxidation in photosystem II, and useful for

induction of superoxide dismutase (SOD) activity algal symbionts (zooxanthellae) (Pätsikkä et al., 2002; Que Jr & Tolman, 2008).

Howard & Brown (1984) hypothesized the theory of how trace metals incorporated in coral skeleton. Metals were partitioned in coral skeleton in 3 sites: skeletal aragonite, crystal lattice, and organic component. During aragonite formation, metal ion was taken up by crystal lattice and developed with equation of $\text{CaCO}_3 + \text{Me}^{2+} \xrightarrow{\text{1/4}} \text{MeCO}_3 + \text{Ca}^{2+}$. Cu has a divalent radius of 0.72 Å and electronegativity of 1.9, these factors suggested the influence of element incorporated with coral skeleton, so that the higher amount of Cu was absorbed in coral skeleton of *P. lutea*. The process binding of heavy metal in coral skeleton have variety mechanism: through substitution of calcium ion (Ca) on coral lattice, trapping small organism (plankton) in skeleton cavities, uptake

Table 2. Comparison of heavy metals (Cd, Cu, and As) between Pantai Kondang Merak and the other areas

Type samples	Location	Concentration (ppm)			References
		Cd	Cu	As	
<i>Coral Reef</i>	Gulf of Mannar, India	0.7	2.4	10.64	Kumar et al. (2010)
<i>Siderastrea siderea</i>	Costa Rica	7.5	2	-	Guzmán & Jiménez (1992)
	Panama	7.6	3.8	-	
<i>Porites lutea</i>	Red Sea	0.052	5.7	-	Hanna & Muir (1990)
<i>Porites andrewsi</i>	Tuticorin	7.21	10.65	-	Jayaraju et al. (2009)
<i>Acropora formosa</i>	Coast, South India	4.53	2.46	-	
		3.25	2.56	-	
<i>Montipora digitata</i>	Pulau Tidung	1.42	-	-	Mellawati & Bachtiar (2011)
<i>P.stephensoni</i>	Pulau Tikus	1.27	-	-	
	Pulau Bokor	1.28	-	-	
<i>P.astreoides</i>	North-West Coast of Venezuela	-	16.33	-	Bastidas & García (1999)
<i>Goniastrea sp</i>	Samalona	-	3.8	-	Pratikto (2014)
	Barranglombo	-	3.78	-	
	Bone Batang	-	0.6	-	
<i>P. lutea</i>	Pantai Kondang Merak (KM1)	2.236 ± 0.59	9.726 ± 0.76	2.474 ± 1.07	This research
	Pantai Kondang Merak (KM2)	1.989 ± 0.77	19.157 ± 7.31	2.064 ± 1.07	

organic material from coral tissue and coral feeding process (Dar et al., 2018).

Trace metals comparison to others area

In previous research showed vary of concentration trace metals (Cd, Cu, and As) in coral (Table 2). Corals have the ability to absorb heavy metals into their skeletons from the water environment in which they grow. Binding of trace metals into lattice of coral skeleton reflect their concentrations in water column where it grew. The availability of trace metals spread out in water column due to the upwelling, anchoring, dredging, volcano activity, and terrestrial input. Hence, various environment of coral reef grew in around the world with different degree of pollution, colony size, and species resulted in different concentration of heavy metals bound in the skeleton (Luthfi et al., 2017).

The observation on heavy metals in coral showed important information regarded history of heavy metals pollution at South of Java Sea where the location is far and not directly connecting to industrial waste. The existence of heavy metals in the ocean (Pantai Kondang Merak where connected into the Indian Ocean) may be influenced by a natural condition such as volcanic or plate shift, sediment stirring and deposition from organic matter. This also gave information about the ability of coral to absorb the trace mineral in their skeleton that can be used to described the environment condition in the past. In the future, Pantai Kondang Merak water may be more polluted with other heavy metal because the road infrastructure is being developed in surrounding area. That condition may attract tourism and industrial investment resulted in more pollutant accumulated in the sea. These will be the threats for the coral reef ecosystem in the future unless the government or non-government organization adopt clear and good management strategies to protect coral reef form pollution.

CONCLUSION

Annual growth band of *P. lutea* in Pantai Kondang Merak showed different absorption of heavy metals. The average concentration of Cd, Cu and As of KM1 were 2.236 mg/kg, 9.726 mg/kg, and 2.474 mg/kg respectively while in the for KM2 they were 1.989 mg/kg, 19.157 mg/kg, and 2.064 mg/kg. These differences may be caused by the number of trace metal that consumed by coral, number of cadmium from environment and speed of skeletal formation.

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