Late Holocene Heavy Metals Record of Jakarta Bay Sediments

Rekam Jejak Logam Berat Holosen Akhir dalam Sedimen Teluk Jakarta

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ABSTRACT: This paper reports copper, zinc, lead, cadmium, and chromium records of Jakarta Bay sediment since 500 AD of a 150 cm long gravity core (TJ–17, 106.902488°E, 5.99381°S). The core was acquired from the eastern part of Jakarta Bay in 2010 onboard RV Geomarin I by the Marine Geological Institute–MEMR. We applied core scanning, grain size and elemental analyses to track environmental changes. Core scanning was conducted by multi sensor core logger, grain size analysis was carried out by sieving and pipette methods and heavy metal concentration was analyzed using atomic absorption spectrometry on bulk samples. The results yield baseline values of Cu at 12.88 ppm, Zn at 95.5 ppm, and at Cr 60.13 ppm. Vertical record of heavy metals show three stages of environmental changes in the region: (I) 476–933 AD, (II) 933 – 1799 AD, and (III) 1799 – 1991 AD. These changes are interpeted as related to land use changes caused by human activity in the West Java region since Tarumanagara to modern Indonesia.

***Keywords*:** heavy metals, Jakarta Bay, land use changes, anthropogenic activities

ABSTRAK: *Makalah ini melaporkan hasil penelitian kandungan tembaga, seng, timbal, kadmium dan krom dalam sedimen Teluk Jakarta sejak 500 M dari sampel sedimen yang diambil dengan gravity core sepanjang 150 cm di bagian timur Teluk Jakarta (TJ–17, 106.902488°BT, 5.99381°LS). Sampel diambil pada tahun 2010 dengan menggunakan KR Geomarin I oleh Pusat Penelitian dan Pengembangan Geologi Kelautan–KESDM. Analisis sampel terdiri atas core scanning, analisis besar butir dan unsur untuk menelusuri perubahan lingkungan di darat. Core scanning dilakukan dengan menggunakan multi sensor core logger, analisis besar butir dengan menggunakan metoda ayakan dan pipet, dan analisis kandungan logam berat dilakukan dengan metoda atomic absorption spectrometry pada bulk sample. Hasil analisis memberikan nilai rona awal (baseline values) untuk Cu adalah 12.88 ppm, Zn 95.5 ppm, dan Cr 60.13 ppm. Rekaman logam berat secara vertikal menunjukkan tiga tahap perubahan lingkungan: (I) 476 – 933 M, (II) 933 – 1799 M, and (III) 1799 – 1991 M. Perubahan ini diinterpretasikan sebagai perubahan tata guna lahan akibat aktivitas manusia di daerah Jawa Barat sejak Kerajaan Tarumanagara hingga Indonesia modern.*

Kata kunci: *logam berat, Teluk Jakarta, perubahan tata guna lahan, aktivitas manusia*

INTRODUCTION

The term heavy metal usually indicates elements with densities heavier than 5 g/cm^3 (Järup, 2003). Thus, some of trace elements, composing approximately 0.1% of the lithosphere, can be considered as heavy metals (Callender, 2003). Existing studies show that increase of heavy metal within sediments can be related to anthropogenic activities (Zhao *et al.*, 2017; Decena *et al.*, 2018). An increase of industrial activities in a region is usually followed by increase of waste disposal, either in solid, liquid or gaseous forms. Crounse *et al.* (1983) identified severe abdominal pain and anemia that is caused by copper poisoning.

Heavy metals disposed to the water bodies will ultimately be brought to the sea and deposited within the marine sediments that could be ingested by organisms and thus transferring the metals to the food chain (Calmano *et al.*, 1996). Even though geological condition of a region influence heavy metal concentration within the sedimen (Howarth and Thornton, 1983), enrichment of certain heavy metals such as lead, mercury, cadmium, zinc and copper in an

estuary is almost always related to anthropogenic activities (Forstner, 1983).

One of Indonesian water bodies that is most affected by human activities is Jakarta Bay that borders the northern part of Indonesian Capital, Jakarta. Despite providing the livelihood for local fishermen (Putri *et al.*, 2015), the bay also acts as waste disposal from surrounding areas (Cordova *et al.*, 2016) that is transported by thirteen natural rivers belonging to Ciliwung-Cisadane Watershed flowing into the bay (Rochyatun and Rozak, 2007). The bay also acts as means of transportation, as well as site of leisure. Those activities affect the environment of the bay. Such impact was observed by Farhan and Lim (2012) who found the inverse effect of urban pressures and pollutants from four main rivers in Jakarta Bay on the environment. During wet season (northwest monsoon), increase river discharge and runoff from Jakarta coastal plain that coincides with high tides would lead enchanced the possibility of flooding event.

Riverine runoff of an area is affected by land use changes, as was observed in Nyando river basin, Kenya, by Kundu and Olang (2011). Changes in land cover due to human activities are found to increase sedimentation in several deltas of norther Java (Lavigne and Gunnell, 2006). Increasing open spaces inland would increase sediment volume flowing into the river that in turn would increase deposition of sediment, as well as heavy metals, in the bay.

To date, heavy metal analysis for environmental purposes are usually limited to surface sediment (Permanawati *et al.*, 2013) or by concentrating the analysis on the silt and clay fractions of marine surface sediment (Zhuang and Gao, 2014). Previous study on historical heavy metal record from Jakara Bay was concentrated on tracing the impact of stricter environmental regulations on heavy metal contamination from the Bay (Hosono *et al.*, 2011). The objective of this study is to determine the baseline value of heavy metal concentration in the bay from long sediment core sample and to identify environmental changes of Jakarta Bay and surrounding area.

METHODS

This study uses one 150-cm core taken from the western part of Jakarta Bay (TJ-17) during Geomarin I cruise in September 2010 as part of thematic mapping project of Marine Geological Institute of Indonesia (MGI) - Ministry of Energy and Mineral Resources (MEMR). Core location is presented in Figure 1 and Table 1. The core was split longitudinally and divided into archive and working parts. Core description was carried out onboard Geomarin I and core sampling was conducted at the Core Laboratory of MGI in Cirebon. The sampling interval for grain size analysis is 10 cm and for heavy metal analysis is 5 cm (Figure 2).

Table 1. Information on core sample that is used in this study.

No.	Core Number	Longitude (E)	Latitude (S)	Water Depth (m)	Core length (cm)	Sediment Type
1	TJ-17	106.902488	-5.99381	20.78	150	Silt and Clayey Silt

Sediment Age Determination

Determination of the age of marine sediments can be conducted by various methods, among others are ^{210}Pb and ^{14}C. ^{210}Pb dating method was conducted by determining the ratio of ^{210}Pb to ^{206}Pb wihin the sedimen column (Lowe and Walker, 1997). The method relies upon the amount of atmospheric ^{210}Pb fallout in the sediment (Appleby, 1998) to calculate mass accumulation rates (MARs) of recent sediments (Boer *et al.*, 2006). Even though ^{210}Pb dating method yielded reliable data for areas with nonuniform accumulation (Appleby, 1998), the validity of this method needs to be assessed with independent method, such as ^{137}Cs or ^{241}Am (Appleby, 1998; Boer *et al.*, 2006).

Field observation shows that the seafloor sediments of Jakarta Bay is mostly disturbed by fishing and marine activities, while no tephra layer was observed in the core to be used as age control (Figure 3). Thus, the age model of Core TJ-17 was developed from radiocarbon data. The ^{14}C analysis was conducted on molluscs shell fragments that were found in two different layers (Table 2). The analysis was carried out at the Australian National University Radiocarbon Dating Centre using accelerated mass spectrometer (AMS) that use ^{14}C half life of 5568 years (Fallon *et al.*, 2010). The ^{14}C age was calibrated into calendar age (AD) by applying reservoir age of 391 from Sabui Bay, South Kalimantan (Southon *et al.*, 2002). The caledar age was calculated by CALIB 7.1 (Stuiver *et al.*, 2019; Stuiver and Reimer, 1993).

Grain Size Analysis

Previous studies (Balkis *et al.*, 2009) showed the affinity of heavy metal to fine grained sediments as opposed to coarse grain sediments. As we performed no separation between coarse and fine grain sediments, we conducted grain size analysis to determine the sediment

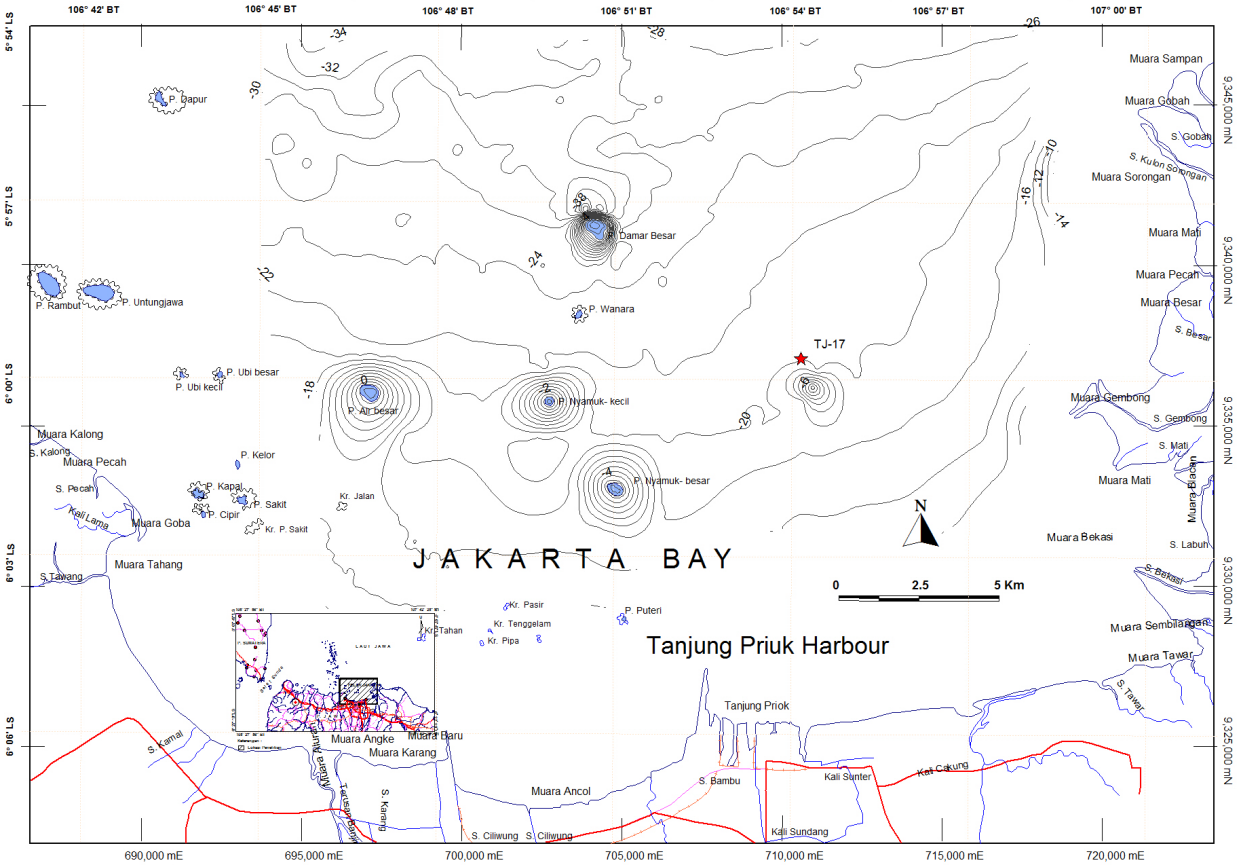

Figure 1. Sampling location Core TJ-17 (red star). Bathymetric data from Zuraida *et al.* (2010).

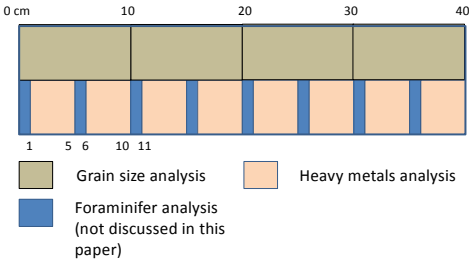

Figure 2. Sampling strategy for grain size and heavy metals analyses.

Table 2. AMS ^{14}C dates of molluscs shell fragments and their converted calender age (AD).

Depth (cm)	^{14}C age	Cal ^{14}C age (AD)
60	665 ± 30	1626 ± 170
145	1875 ± 30	515 ± 162

type to compare it to heavy metals data. Grain size analysis was carried out at MGI - MEMR, by the dry sieving method (Folk, 1980) for samples with particles >63 µm followed by pippette analysis for particles <63 µm. Determination of sediment type follows Folk (1980) classification.

Heavy Metal Analysis

Heavy metal analysis of Jakarta Bay sediment in this study was conducted on 12 bulk samples. Sample preparation include complete digestion following Kouadia and Trefry (1987). About 0.5 g of samples were treated with HCl and $HClO_4$ to remove silica. Samples were then digested by HNO_3 and HCl . Prior to analysis with *atomic absorption spectroscopy* (AAS), the aliquots were diluted by HNO_3 10%. The use of AAS for heavy metals study has been conducted in various places, such as Karnaphuli River in Bangladesh (Ali *et al.,* 2016). The analysis was conducted at Geological Survey of Indonesia-MEMR. Concentration of copper (Cu), lead (Pb), zinc (Zn) and chromium (Cr) was measured by flame AAS method with limit of detection of 0.01 ppm, while cadmium (Cd) concentration was analysed using graphite furnace AAS (GFAAS) with detection limit of 0.01 ppb.

Determination of natural baseline level of each heavy metal could be conducted either by empirical or statistical method, including using core samples that

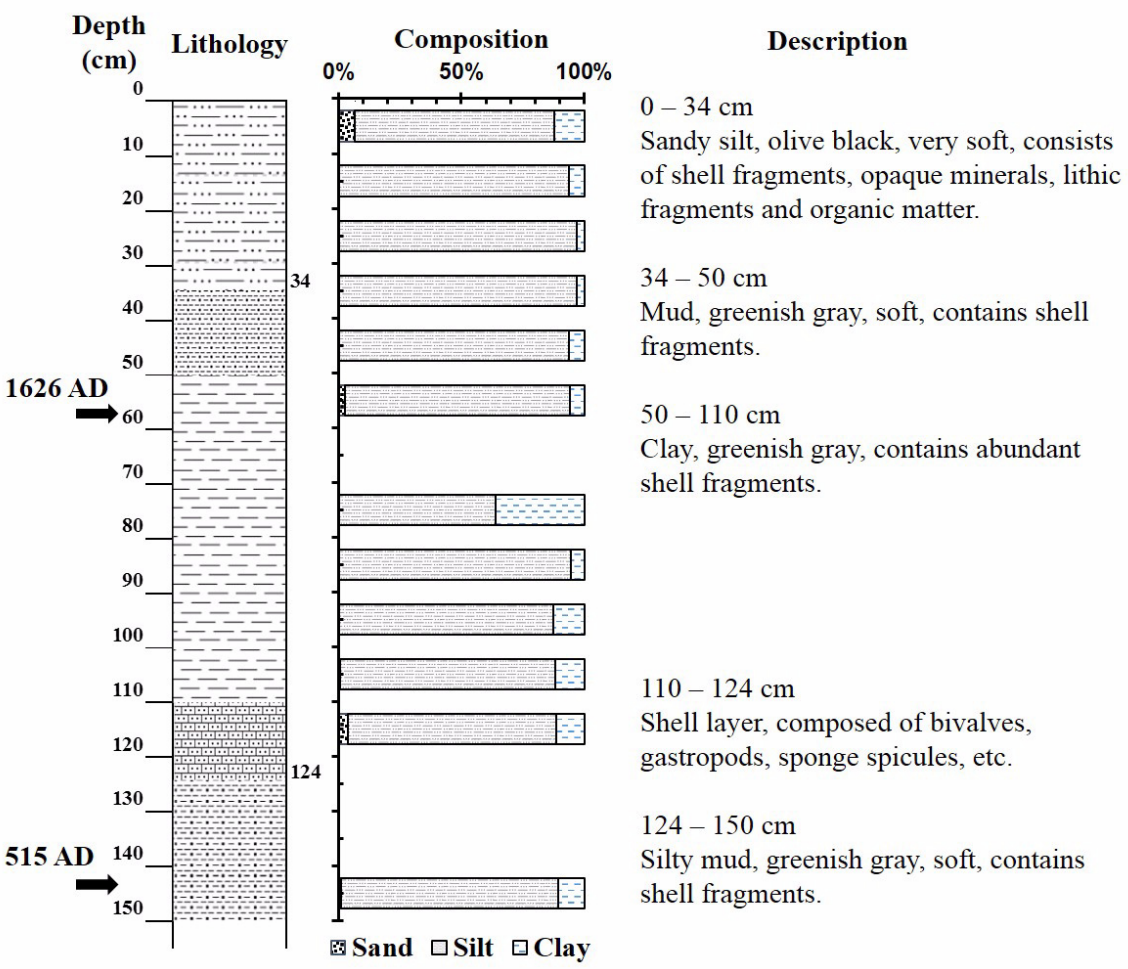

Figure 3. Lithologic column of TJ-17. Arrows indicate the layers where samples for ^{14}C dating were obtained. Numbers above arrows indicate calendar age. Composition of size fraction is in cummulative percentage.

penetrate pre-anthropogenic section (Birch, 2017). Abrahim and Parker (2008) determined pristine pre-industrial sediments in Tamaki Estuary by averaging five low concentration samples in the relatively "flat" baseline that was observed in the core. This paper follows the method of Abrahim and Parker (2008).

Core Scanning

To recover fast and reliable historical data from the cores, we conduct non-destructive analysis using multi sensor core logger (MSCL-S) Geotek at MGI, MEMR. The core logger recorded magnetic susceptibility, colour spectrophotometry, and elemental composition of the sediment. Elemental composition of the sediments was used to identify terrigenous influx into site location and environmental condition.

Magnetic susceptibility (MS) of sediments is determined by the abundance of magnetic minerals contained in the sediments and is measured using Bartington MS2E point sensor on split core. Magnetic minerals in the sediments are products of weathering of existing rock formation on land which include volcanic deposits, sedimentary and igneous rocks. The result is reported as SI unit time 10^{-5}.

Color spectrometry was conducted by Minolta CM2600d spectrophotometer that quantify the color of wet sediments using color space that was defined by the International Commission on Illumination (CIE) in 1976. The color space, known as CIE L*a*b* or CIE Lab, designates color into three categories: L* (lightness) from black (0) to white (100); a* from green (-) to red (+); and b* from blue (-) to yellow (+). Previous study revealed linear correlation between L* and carbonate content in marine sediments where lighter sediments contains higher carbonates and vice versa (Rothwell and Rack, 2006). A study from Cendrawasih Bay, Papua, shows inverse correlation between L* and organic content (Sari *et al.*, 2014). This paper applied L* as indicator of organic content in the TJ-17 sediments.

Sediment composition measurement is observed by using x-ray fluorescence (XRF), Olympus Innov-X,

scanner that is attached to the MSCL-S. The XRF scanner can measure wide range of major, minor and trace elements and comparable to traditional laboratory instrument (Kenna *et al.*, 2010) and report the result in ppm. Paleoenvironmental condition was determined from magnetic susceptibility and elemental ratios of Ti/Ca, Fe/Ca and Fe/Ti (Blanchett, 2010; Croudace *et al.*, 2006).

The intensity of heavy metal in a given sediment can be expressed by various methods, such as Geoaccumulation Index (I_{geo}). I_{geo} involves simple calculation that includes multiplication factor to reduce lithogenic effect (Kowalska *et al.*, 2018). I_{geo} was applied to quantify the level of heavy metal pollution in marine sediments of Laizhou Bay and Zhangzi Island of China (Zhuang and Gao, 2014). The calculation of I_{geo} is:

$$I_{geo} = log_2\left[\frac{C_m\ sample}{(1.5\ x\ C_m\ background)}\right]$$

where C_m is concentration of heavy metal m in both sample and background. The background values that are used vary from global value of heavy metal in shale (Zahra *et al.*, 2014) to the baseline value of the element (Abrahim and Parker, 2008). Barbieri (2016) defined background values as "*Natural contents of substance in the soil completely dependent on the compositional and mineralogical characteristic of the parent/source geological material*" and baseline values as "*Actual mostly diffuse range of concentration of an given element in a specific area dependent both on the nature of the parent geological/source material and on the historic diffuse release into the environment of contaminants from anthropogenic sources*".

RESULTS

Age Model

The age model for core TJ-17 that is located to the west of Muara Gembong was developed in three parts based on the assumption that the rate of sedimentation in the area is linear. The rate of sedimentation between 60 and 145 cm was calculated by assuming linear sedimentation rate (LSR) between two ^{14}C age controls that gave a rate of sedimentation during this time as 0.07 cm/year. Sedimentation rate between 0 and 60 cm was determined by assuming that the core top is modern deposit (equal or younger than 1950. See Fallon *et al.*, 2010), while the bottom part was calculated by assuming LSR (Figures 4a and 4b). The age yielded by ^{14}C dating allows us to reconstruct changes in heavy metal concentration in the Jakarta Bay for the last two millenia.

Sediment Grain Size

Grain size analysis on core samples yielded the predominance of silt and clayey silt on all of the samples (Figure 3). Macroscopic observation indicates that samples containing sand and gravel are mostly composed of biogenic carbonates and thus are not considered in the classification of sediment type.

Heavy Metal Concentrations

Copper concentration rose from 12.30 to 29.70 ppm, with average value of 17.66 ppm, and highest concentration was reached at 1895 AD. The pattern shows stepwise increase of Cu that consists of three stages: (1) 476–933 AD where concentration ranges between 12.3 and 15.5 ppm with an average of 13.6 ppm; (2) 933–1799 AD where concentration ranges 15.3–19.1 ppm and an average of 16.9 pp; and (3) 1799–1991 AD where concentration ranges 19.4–29.7 ppm with an average of 25 ppm.

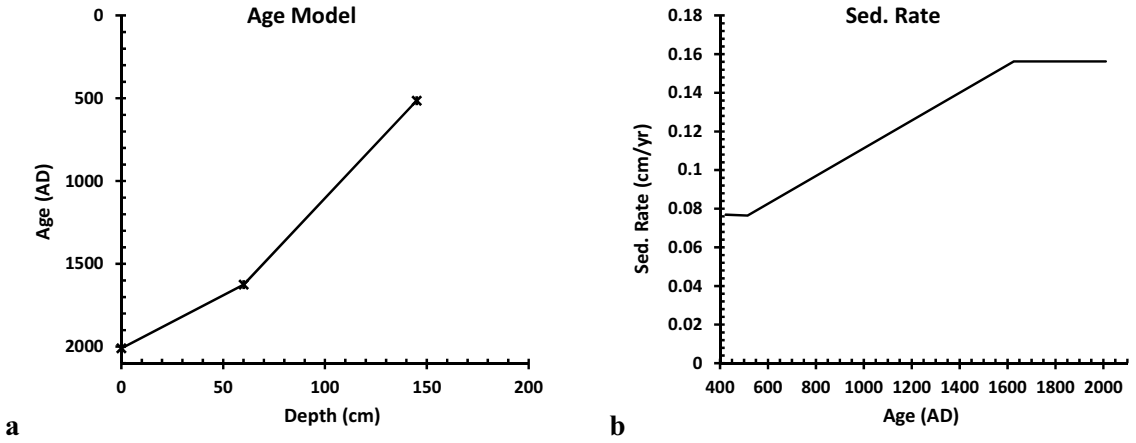

Figure 4. Age model for TJ-17 based on two ^{14}C ages (a) and sedimentation rate of core TJ-17 (b).

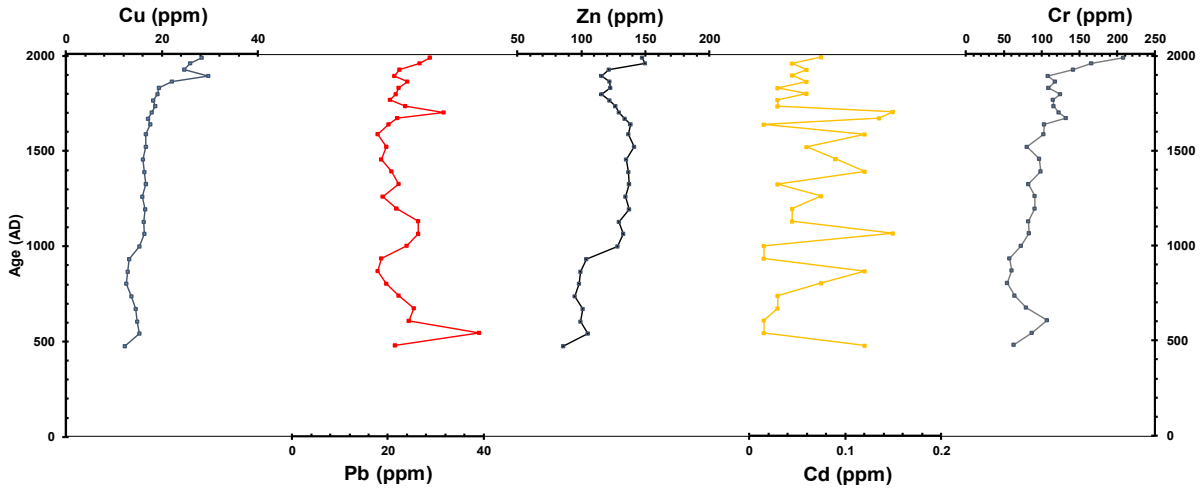

Figure 5. Concentration of Cu, Pb, Zn, Cd and Cr of discrete samples analysed by AAS.

Lead fluctuates between 17.9 – 39.1 ppm, with an average of 23 ppm. Two strong peaks are detected at 541 AD (39.1ppm) and 1703 AD (31.6 ppm), and one weak peak at 1129 AD (26.4 ppm). Those peaks show weak cyclicity with a period of approximately 600 years.

Zinc concentration ranges between 85.8 – 149.9 ppm, an average of 122.8 ppm and maximum concentration was detected at 1959 AD. This metal also shows stepwise within similar intervals of Cu with concentration range of: (1) 85.8 – 105.2 ppm with an average of 98.5 ppm; (2) 115.9 – 141.4 ppm with an average of 132.2 ppm; and (3) 115.7 – 149.9 ppm with an average of 129.9 ppm.

Cadmium concentration ranges between 0.015 – 0.15 ppm with an average of 0.064 ppm and maximum level was reached at 1703 AD. The pattern show strong cyclicity from the base of the core to 1735 AD with concentrations vary between 0.015 – 0.15 ppm and an average of 0.068 ppm. From 1735 to 1991, Cd concntrations show narrow fluctuation between 0.03–0.075 ppm and an average of 0.05 ppm.

Concentration of Cr ranges between 54.54–207.75 ppm with an average of 100.74 ppm and highest concentration was detected at the core top (1991 AD). The metal shows similar stepwise increase like Cu and Zn within the same periods: (1) the concentration ranges 54.54–107.79 ppm with an average of 71.95 ppm; (2) 73.18–132.1 ppm with an average of 99.73 ppm; and (3) 108.43–207.75 ppm with and average of 141.84 ppm.

Baseline Levels

Baseline level of certain heavy metal can be calculated providing the vertical distribution of that metal shows relatively stable concentration in the lower part of the core before increasing to the surface. Calculation of baseline level followed Abrahim and Parker (2008) by averaging five low concentration of each heavy metal.

Heavy metal records from core TJ-17 exhibit relatively stable concentrations of Cu, Zn and Cr that increase upwards (Figure 5), while Pb and Cd higher fluctuation on the lower part of the core. Baseline levels of Cu, Zn, and Cr are 12.88 ppm, 95.5 ppm, and 60.13 ppm, respectively (Table 3). The baseline levels for Pb and Cd can not be calculated as there is no baseline that can be observed (Figure 5).

Calculated baseline levels followed the order: Zn>Cr>Cu. Baseline level of Zn is nearly equal to Zn in shale (Table 3), while baseline levels of Cr and Cu are below geochemical background. We compare our data with stream sediment sampling from Citarum river that was conducted in 2015 by Center for Geological Survey (Table 3) to assess the influence of local geology. The comparison shows that baseline levels of Zn, Cr and Cu are much lower than stream sediments which might be related to sediment deposition within dams and floodgates along the river.

Geoaccumulation Index (I_{geo})

Calculation of I_{geo} applied background value of each heavy metal instead of baseline value considering no baseline value can be calculated for Pb and Cd. We employed geochemical background value of the five heavy metals in the shale (Turekian and Wedepohl, 1961) for the calculation following Zahra *et al.* (2014). I_{geo} is then categorized into six classes that reflect sediment quality (Zahra *et al.*, 2014). Mean I_{geo} of TJ-17 shows that I_{geo} of five metals that were analysed are below 0 that correspond to class 0 (Table 3). The sediment quality of this class is considered unpolluted.

Table 3. Baseline level and mean Igeo of TJ-17 sediments. Geochemical background is from shale of Turekian and Wedepohl (1961). Stream sediment sampling data from Center for Geological Survey (PSG, 2015). Igeo classification follows Zahra *et al.* (2014).

Heavy Metal	Geochemical Background (ppm)	Stream Sediment Sampling (ppm)	Baseline level (ppm)	I_{geo}	I_{geo} Class	Sediment Quality
Cu	45	14 – 40	12.88	-1.97	0	Unpolluted
Pb	20	23 - 89	-	-0.40	0	Unpolluted
Zn	95	121 – 236	95.5	-0.23	0	Unpolluted
Cd	0.3	-	-	-3.18	0	Unpolluted
Cr	90	104 – 256	60.13	-0.49	0	Unpolluted

Core Scanning Result

Magnetic susceptibility of marine sediment is affected by mineral composition, particle size and diagenetic process. Ferro- and ferrimagnetic particles, that are mainly terrigenous particles, yield highest susceptibility and very small particles of those minerals would have very high MS (Blanchett, 2010). Application of MS as indicator of terrigenous input needs to be verified by elemental ratio such as Ti/Ca and Fe/Ti (Figure 6).

MS and Ti/Ca show similar pattern of relatively constant value from 450 AD to 1700 AD and doubles after 1700 AD (Figure 6). Those patterns suggest that MS is carried mainly by Terrigenous Iron Oxide or TIO (Blanchett, 2010). Fe/Ti shows narrow fluctuations between 10 and 20 and increase slightly to the surface. This pattern differ from MS pattern, thus showing that the sediments are unaffected by diagenetic process. Comparison between MS to Ti/Ca and Fe/Ti indicate that MS record of this core reflects terrigenous input into the site. Figure 6 shows an increase of terrigenous input since 1700 AD that peaked at 1831 AD.

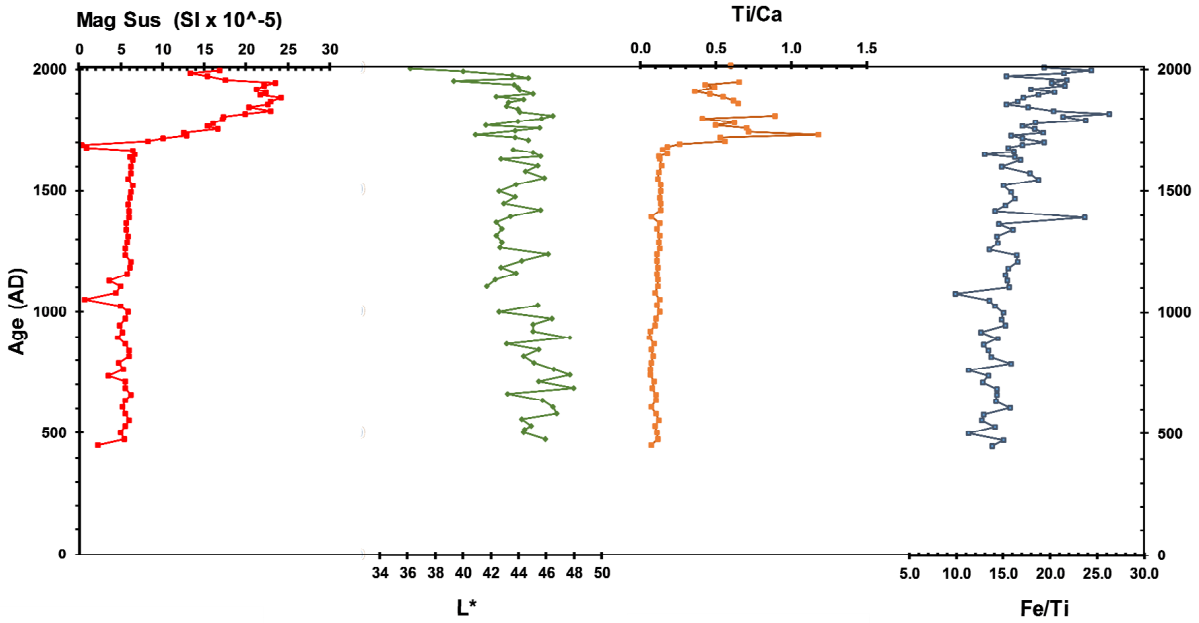

Figure 6. Comparison of MS to L*, Ti/Ca, Fe/Ti and Fe/Ca.

L* is declining upward and shows slight fluctuation around 44. The decline reflects increasing organic content, following Sari *et al.* (2014).

DISCUSSIONS

Factors Controlling Heavy Metal Concentration in the Sediment

There are numerous factors that control concentrations and vertical distributions of heavy metals in sediment, such as: grain size, composition of sediments, organic matter content, individual and combined effects of Eh and pH, etc. (Forstner and Wittmann 1979). Those factors might influence the

bonding and enrichment of heavy metal in marine sediment if the depositional environment, characterized by the chemical composition, salinity, pH, redox potential and hydrodynamic conditions, are favourable (Kljakovic-Gaspic *et al.,* 2008).

It is understood that sediment grain size and composition can influence heavy metal concentration. Jakarta Bay sediment is demonstrated to be composed mainly of silt and clayey silt (Figure 3) that might bind heavy metal to the sediments. Sediment composition is affected by local geology that influences natural background level. Heavy metal concentration (Figure 5) and XRF scanning of TJ-17 (Figure 6) show that drastic increase of terrigenous influx is not complemented by strong concentration increase of all heavy metal, thus suggesting partial contribution of terrigenous influx in the variation of heavy metal concentration in study area.

Positive correlation betwen organic content and Cu, Zn and Cr was observed in Arabian Sea (Shetye *et al.*, 2009), while correlation between organic content and Pb was reported from India (Sudhanandh *et al.*, 2011). Slight increase of organic content in TJ-17 that corresponds with increasing Cu, Zn, and Cr (Figure 5 and Figure 6) indicates that Cu, Zn and Cr accumulation in Jakarta Bay sediment is partially controlled by organic content.

Heavy Metal As a Reflection of Anthropogenic Activities Since 500 AD

Heavy metal records of Cu, Zn and Cr can be divided into three periods that are characterised by stepwise increase: (I) 476 – 933 AD, (II) 933 – 1799 AD, and (III) 1799 – 1991 AD. Period I is characterised by increase of Cu, Zn and Cr followed by slight decline before the end of the period (Figure 5). This period might be related to the development of agricultural irrigation and drainage ditches between 450 – 500 AD ordered by Purnawarman, the third king of Tarumanegara (Walker and Santoso, 1977). The slight decline might indicate limited change of landcover in the region that was indicated by low terrigenous input to site TJ-17.

The second period, Period II, is typically identified by 20% increase in Zn concentration before declining upward (Figure 5). During this period, Cu, Pb, and Cr show slight increase, while Cd exhibit cyclical fluctuation. The increase of heavy metals might be related to agricultural activities during those period. The slight decrease of Zn between 1600 and 1800 AD might be related to the destruction in the region caused by wars between Europeans and local rulers obstructing development of agriculture in the region as was decribed by Ricklefs (2005).

The third period, Period III, is characterised by increase of Cu, Pb, Zn, and Cr while Cd content is lower than Period II (Figure 5). The early Period III might be related to the start of *cultuurestelsel* or compulsory cultivation (Lavigne and Gunnell, 2006) that increase metal content. Sharp increase of heavy metal in late Period III is interpreted as the result of the development of modern Indonesia that resulted in landcover change such as for agriculture, housing, recreation, fuel consumption, industrial activities, etc.

CONCLUSION

Vertical distribution of heavy metals from site TJ-17 yields local baseline levels of: Cu, Zn, and Cr are 12.88 ppm, 95.5 ppm, and 60.13 ppm. No baseline levels can be calculated for Pb and Cd due to higher concentrations of both metals in the lower part of the core. I_{geo} calculation indicates that the sediments of site TJ-17 is relatively unpolluted. Our data suggested that heavy metal contents in study area are influenced by terrigenous input and organic content.

Based on stepwise increase of Cu, Zn and Cr, we can differentiate three periods of human activities: (I) 476 – 933 AD, (II) 933 – 1799 AD, and (III) 1799 – 1991 AD. Sharp increase of heavy metals in late Period III was related increasing human activities in modern Indonesia.

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