

# Temporal and Spatial Distribution of Heavy Metal in Sediment of Urban Coastal Waters: A Case Study in Jakarta Bay, Indonesia

## *Sebaran Temporal dan Spasial Logam Berat di Sedimen Perairan Pesisir: Studi Kasus Teluk Jakarta, Indonesia*

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**ABSTRACT:** Heavy metals, hazardous chemical substances, increase in marine environment due to anthropogenic discharges. However, due to the hydrodynamic of the marine system these metals could vary both temporal and spatial distribution of metals in Jakarta Bay. This study was to reveal the temporal and spatial distribution of metals in sediment over the bay and to assess the environmental condition. Sediment samples were collected in 11 stations of March (transitional season) and June (dry season) 2013. The result showed that the concentration of heavy metals varied spatially, in which elevated concentration occurred adjacent terrestrial indicating the enrichment of metal-anthropogenic source, but insignificant temporarily.

**Keywords:** heavy metals, spatial distribution, temporal distribution, anthropogenic activities, Jakarta Bay.

**ABSTRAK:** Logam berat merupakan bahan berbahaya yang tersebar di lingkungan laut karena pengaruh aktifitas antropogenik. Akan tetapi, logam berat ini dapat terdistribusi secara temporal ataupun spasial di Teluk Jakarta akibat sistem hidrodinamika laut. Tujuan dari penelitian ini adalah untuk mengetahui distribusi spasial dan temporal logam berat dalam sedimen dan untuk menilai kondisi lingkungan teluk. Sedimen diambil dari 11 stasiun pada bulan Maret (musim transisi) dan Juni (musim kering) tahun 2013. Hasil penelitian menunjukkan bahwa distribusi spasial logam berat bervariasi, dimana konsentrasinya meningkat di lokasi dekat daratan yang mengindikasikan tingginya sumber logam-antropogenik namun secara temporal tidak signifikan.

**Kata kunci:** logam berat, distribusi spasial, distribusi temporal, kegiatan antropogenik, Teluk Jakarta.

### INTRODUCTION

Heavy metals, are very likely bio-accumulated into organisms causing adverse effects (Muse et al., 2006; Hyun et al., 2007; and Velusamy et al., 2014). These chemical pollutants, are introduced into marine environment through several mechanisms such as river runoff and direct discharge from urban or industrial activities as their major sources in estuarine and coastal area (Kurun et al., 2007; Sakellari et al., 2011; and Chakraborty et al., 2014;). Metals exhibit different function with regard to the organisms (Yamada et al., 2006 and Zhou et al., 2007) i.e. as essential for growth, zinc (Zn), copper (Cu), but as toxic, cadmium (Cd), nickel (Ni) and lead (Pb).

Weathering of watershed rock and contaminated soil become one of metal inputs into the aquatic environment, and seasonal alteration stimulates the distinctive rate of metals release (Hosono et al., 2011; Oliva et al., 2012). The metals usually are stored in waste dumping site, soil and deposited sediment.

Either anthropogenic or natural storages may be subject to erosion. The alteration of hydrological properties could mobilize those metals (Schiedek et al., 2007), including flooding, for example, the increase of Cd, Cu, Cr, Pb and Zn occurring in Odra River, western Poland in 2010 (Ibragimow et al., 2013).

Jakarta bay is considered as complex shore-ecosystem area, and its geographical position near the most active city in Indonesia gives particular pressure to its ecosystem like heavy metals input as a contaminant (Williams, et al., 2000; and van der Meij, et al., 2009). Yoo et al. (2014) and Nur et al. (2001) explained that Jakarta is considered as one of the most vulnerable cities in Southeast Asia due to climate change. The coastal area receives metal contaminant mainly derives from direct discharge of approximately 1,100 tons of solid waste. This massive pollutant discharge degrades aquatic sustainability causing an adverse effect on both marine life such as large scale mortality of coral reef in

Table 1. The geographical coordinates of sampling sites of sediment in Jakarta Bay

| Sampling Point | Longitude   | Latitude  |
|----------------|-------------|-----------|
| 1              | 106° 45'21" | 6° 05'31" |
| 2              | 106° 45'58" | 6° 05'24" |
| 3              | 106° 46'48" | 6° 05'32" |
| 4              | 106° 52'50" | 6° 04'04" |
| 5              | 106° 56'19" | 6° 05'01" |
| 6              | 106° 57'22" | 6° 04'50" |
| 7              | 106° 57'47" | 6° 02'55" |
| 8              | 106° 56'21" | 6° 02'48" |
| 9              | 106° 48'50" | 6° 03'50" |
| 10             | 106° 46'36" | 6° 03'36" |
| 11             | 106° 54'00" | 6° 02'10" |

Jakarta Bay (van der Meij et al., 2010) and human life (Firman et al., 2011).

Spatial distribution of heavy metals in the sediment of Jakarta bay is likely to change over time because of the hydrodynamic of the bay. The aim of this study was to reveal the spatial distribution of metals (Cd, Cu, Fe, Ni, Pb, and Zn) in the sediment of Jakarta Bay and to assess their environmental condition.

## METHODS

Sediment samples were collected from Jakarta Bay in March (representing transitional season) and June 2013 (dry season). 11 stations were selected to represent sediment of Jakarta Bay in the area of 106°45.35'E – 106°57.79'E and 6°2.17'S – 6° 5.83S (Table 1), based on the distance between estuary and land (Figure 1). There were seven points (1-7) representing stations located close to river mouths and harbor and the other 4 sites (8, 9, 10, 11) located away from coastline.

In each station, sediment samples were collected using stainless steel ponar grab the surface sediment (250 g) was then subsampled using PE spoon and put into an acid pre-cleaned polyethylene sample-bottle. To minimize the chemical alteration, all of those samples

were stored in the 4°C cool box during transportation to the laboratory (Loring and Rantala, 1992).

Metal analysis was performed based on USEPA 3050b methods and the procedure was divided into three consecutive steps. The first step was the addition of nitric acid and the last step was the addition of concentrated HCl. All chemicals used in this analysis were analytical grade purchased from Merck. To ensure clean analysis, all glassware used in this procedure were soaked in HNO<sub>3</sub> (1+1) for 24 hours then were rinsed using distilled water before used. After those acid

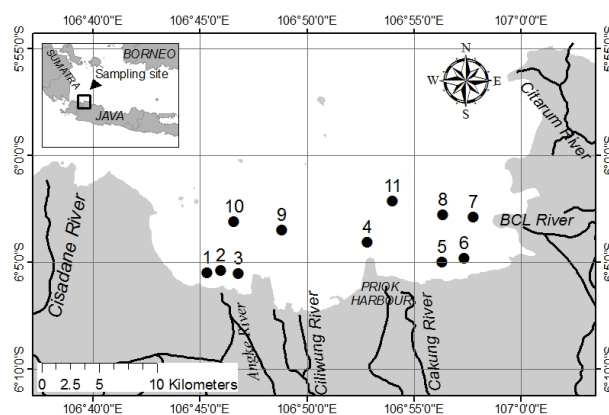


Figure 1. The sampling sites of sediment collection

addition steps completed, the sample was filtered and the filtrate was injected to FAAS (Flame Atomic Absorption Spectrophotometer) Varian SpectraAA 20-plus<sup>®</sup> for the measurement of Cd, Cu, Fe Ni, Pb, and Zn.

Quality control laboratory was carried out to perform for both precision and accuracy test using Certified Reference Material CRM PACS-2<sup>®</sup> analysis. Table 2 presented the analysis result of CRM. Cu, Pb, Fe, Cd, Zn and Ni.

The data of heavy metals for spatial distribution were presenting in map generating using ArcGIS 10.2 software. All data were analyzed normality test prior the statistical analysis. The Principal Component Analysis (PCA) was performed for multivariate data. Assessments of the sediment quality were based on

Table 2. The accuracy and precision of each metal

| No | Element | No. of sample (N) | LOQ  | Accuracy       |        |              | Precision |
|----|---------|-------------------|------|----------------|--------|--------------|-----------|
|    |         |                   |      | Expected Value | Result | Recovery (%) | RSD (%)   |
| 1  | Cu      | 15                | 0.10 | 310            | 315    | 102          | 0.6-2.3   |
| 2  | Pb      | 15                | 1.00 | 183            | 186    | 102          | 0.1-1.8   |
| 3  | Cd      | 15                | 0.04 | 2.11           | 2.25   | 107          | 0.2-2.3   |
| 4  | Zn      | 15                | 0.10 | 364            | 388    | 107          | 0.2-2.4   |
| 5  | Ni      | 15                | 0.40 | 39.5           | 37.1   | 94           | 2.4-4.1   |
| 6  | Fe      | 15                | 0.30 | 40900          | 39493  | 97           | 0.7-4.1   |

LOQ = Limit of Quantification  
RSD = Relative Standard Deviation

Enrichment Factor (EF) and Index of Geo-accumulation ( $I_{geo}$ ).

Enrichment Factor (EF). In this study, Fe normalization is run due to Fe distribution is not related to other metals, and usually, Fe has high natural concentration (5.63%, Taylor 1964). The EF can be formulated as:

$$EF = \frac{\left(\frac{C_i}{C_r}\right)_{sample}}{\left(\frac{C_i}{C_r}\right)_{background}} \quad (1)$$

$C_i$  sample describes the concentration of metals in the sample and  $C_r$  sample represents the concentration of reference metals in the sample.  $C_i$  background and  $C_r$  background explain the concentration of metals in the background and the concentration of reference metals in the background, respectively. EF value is interpreted following description of Varol (2011);  $EF < 1$  (no enrichment);  $1 < EF < 3$  (minor enrichment);  $3 < EF < 5$  (moderately enrichment);  $5 < EF < 10$  (moderately severe enrichment);  $10 < EF < 25$  (severe enrichment);  $25 < EF < 50$  (very severe enrichment) and  $> 50$  (extremely severe enrichment).

Index of Geo-accumulation ( $I_{geo}$ )

In this study, crustal average has been used as reference value.  $I_{geo}$  can be expressed as the following:

$$I_{geo} = \frac{\log_2 C_n}{1.5 B_n} \quad (2)$$

$C_n$  and  $B_n$  area concentration of metals in the sample and in the background, respectively. Factor 1.5 is the correction factor due to the lithogenic effects. Normalization of metals concentration was done by a comparison with iron (Fe) concentration. Caeiro et al. (2005) clustered  $I_{geo}$  scores into 6 classes which  $I_{geo} < 1$

(unpolluted state);  $1 < I_{geo} < 2$  (very lightly polluted);  $2 < I_{geo} < 3$  (lightly polluted);  $3 < I_{geo} < 4$  (moderately polluted);  $4 < I_{geo} < 5$  (highly polluted) and  $I_{geo} > 5$  (very highly polluted).

## RESULTS

The concentration of all metals showed a slight difference of median value between March and June. The deviations of concentration in March were lower than those observed in June (Table 3). However, statistically, the concentrations of the metals indicated insignificant differences between March and June suggesting the metals were distributed similarly.

The dendrograms were drawn by complete linkage methods using Euclidean distance measurement. The concentration of metal in sediment between March and June observation indicated differences in the spatial distribution (70% of similarity), in which there were seven clusters (Figure 2a) and six clusters (Figure 2b), respectively. The composition of the station in each cluster depicted the diverse of metals distribution in sediment. The stations located close to the shoreline tend to be clustered indicating the stations influenced by the anthropogenic sources.

The distribution pattern of metals in March was relatively different, compared with that observed in June showing several high values of metals (Figure 3). These later values appeared in the eastern part of the shoreline of the bay except for Fe and Zn.

### The Source of Metals

Figure 4a (March) and Figure 4b (June) depicted different trend of metal origin. In March, the source of Fe was distinguished from the source of cadmium, copper, nickel, lead and zinc (Figure 4a). Fe is an

Table 3. The heavy metals concentration in sediment of the Jakarta Bay

| Element          | Month | Minimum       |          |          |        |        | p*    |
|------------------|-------|---------------|----------|----------|--------|--------|-------|
|                  |       | Maximum value | value    | Mean     | Median | SD     |       |
| mg/kg dry weight |       |               |          |          |        |        |       |
| Cd               | March | 1.18          | 0.08     | 0.59     | 0.55   | 0.30   | 0.369 |
|                  | June  | 1.74          | 0.29     | 0.70     | 0.54   | 0.45   |       |
| Cu               | March | 94.80         | 16.40    | 46.90    | 33.6   | 29.20  | 0.992 |
|                  | June  | 157.00        | 16.60    | 49.80    | 35.9   | 40.70  |       |
| Fe               | March | 62313.00      | 40678.00 | 48063.00 | 46292  | 6889   | 0.057 |
|                  | June  | 60300.00      | 39573.00 | 51800.00 | 51114  | 6186   |       |
| Ni               | March | 38.40         | 13.70    | 24.70    | 23.60  | 6.10   | 0.438 |
|                  | June  | 75.80         | 15.90    | 29.40    | 26.70  | 16.00  |       |
| Pb               | March | 41.70         | 8.10     | 23.70    | 21.90  | 9.80   | 0.401 |
|                  | June  | 89.40         | 7.80     | 31.40    | 28.80  | 21.70  |       |
| Zn               | March | 503.00        | 72.10    | 206.00   | 144.00 | 137.00 | 0.196 |
|                  | June  | 1270.00       | 85.30    | 408.00   | 241.00 | 398.00 |       |

\* 95% of confidence level

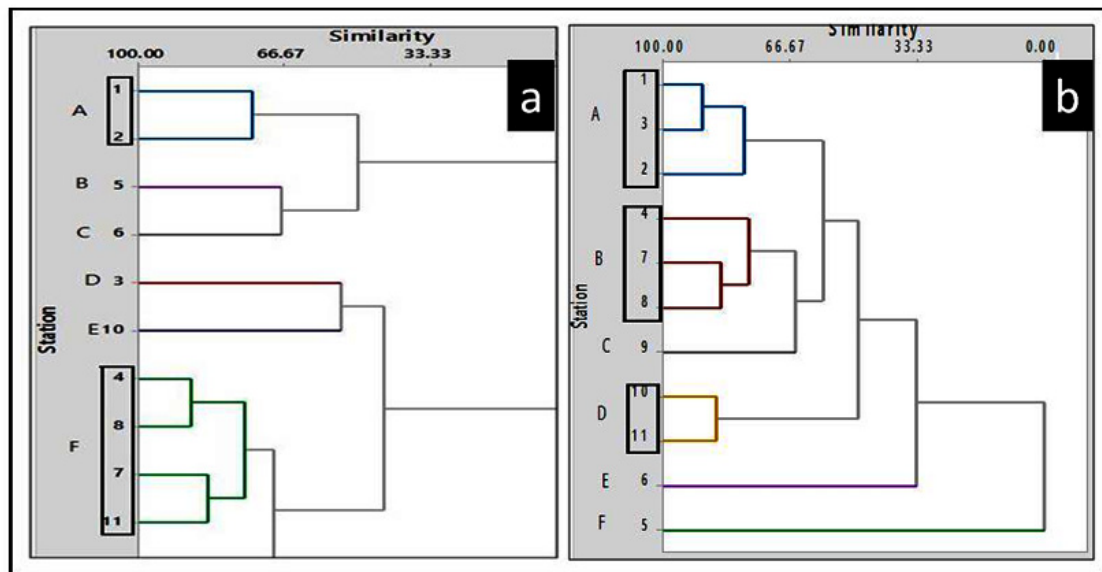


Figure 2. The dendrogram for clustering the stations: a. March; b. June

indicator for natural metal and Figure 4a described that Cd, Cu, Ni, Pb and Zn were positively correlated meaning the increase of one of the metals most probably indicated the increase of other metals. Since these metals (Cd, Cu, Ni, Pb and Zn) are influenced by anthropogenic source, the level of these metal represented the level of anthropogenic activities. Although, the exact source of the metal could not be clearly detected. Moreover, in June, Fe was positively correlated with Cd, Cu, Ni, Pb (Figure 4b), however the correlation was not strong enough (represented by short line in the Figure 4b) since they are from different origin. In addition, in June Zn had different origin Enrichment Factor can support the interpretation of PCA whether metals abundance is altered by natural or anthropogenic source.

Enrichment Factor (EF) varied as the season changed. EF in March could be arranged in order of Zn>Cd>Pb>Cu>Ni; whereas EF scores in June were Zn>Cd>Pb>Cu>Ni. Ni in both months was interpreted as normal concentration (no enriched) with EF score <1. In contrast, all metals were classified in moderate to severe enrichment. Zn in June exhibited severe enrichment ranging from 1.5 to 19.2 (Figure 5a and Figure 5b).

### The Status of Metals Pollution

Index of geoaccumulation ( $I_{geo}$ ) unveiled the general state of sediments in Jakarta Bay in which most of the observation station was in unpolluted to lightly polluted state. March and June indicated the different of  $I_{geo}$  in which extreme condition was exhibited.  $I_{geo}$  in March were arranged in order of Zn>Cu>Cd>Pb>Ni and  $I_{geo}$  in June were Cd>Pb>Ni>Cu>Zn. High  $I_{geo}$

were showed by Cu, Cd and Zn in March. 1 station suffered very high pollution by Cu and Cd, whereas 2 stations indicated very high pollution by Zn. However, In June only Pb and Cd caused very high pollution which two stations experienced very high pollution of Pb and three stations suffered very high pollution of Cd (Figure 6a and Figure 6b).

Several studies of heavy metals concentration, as listed in Table 4, had been conducted, yet there were no continuous data to get a better understanding of metal concentration in the bay. Cu, Pb, and Zn showed fluctuated concentration over 3 different years of observation. Decreasing concentration these 3 metals occurred between 2003 and 2006, however in 2010 the concentration jumping to an extreme value. The heavy metals concentration in this study showed no significant differences with metals concentration in another coastal area (Table 4).

### DISCUSSIONS

The temporal distribution of the metals showed insignificant different whereas the spatial distribution varied. This could be related to three possibilities; i.e. the difference of anthropogenic discharge, river flows and rainfall occurring those two months of sampling. Each river has different flow that could affect variability of the transported metal, for example, the debit of Angke river varied 7.4-27.5 m<sup>3</sup>/s and that of Ciliwung river ranged 28.3-61.8 m<sup>3</sup>/s (BPLHD Jakarta, 2013). Between those two months, these varieties of those river flows were also influenced by rainfall. The rainfall of these two months were different; i.e. 101-300 mm March 2013(BMKG, 2013a) and 0-150 mm June (BMKG, 2013b). Moreover, the strong current and high

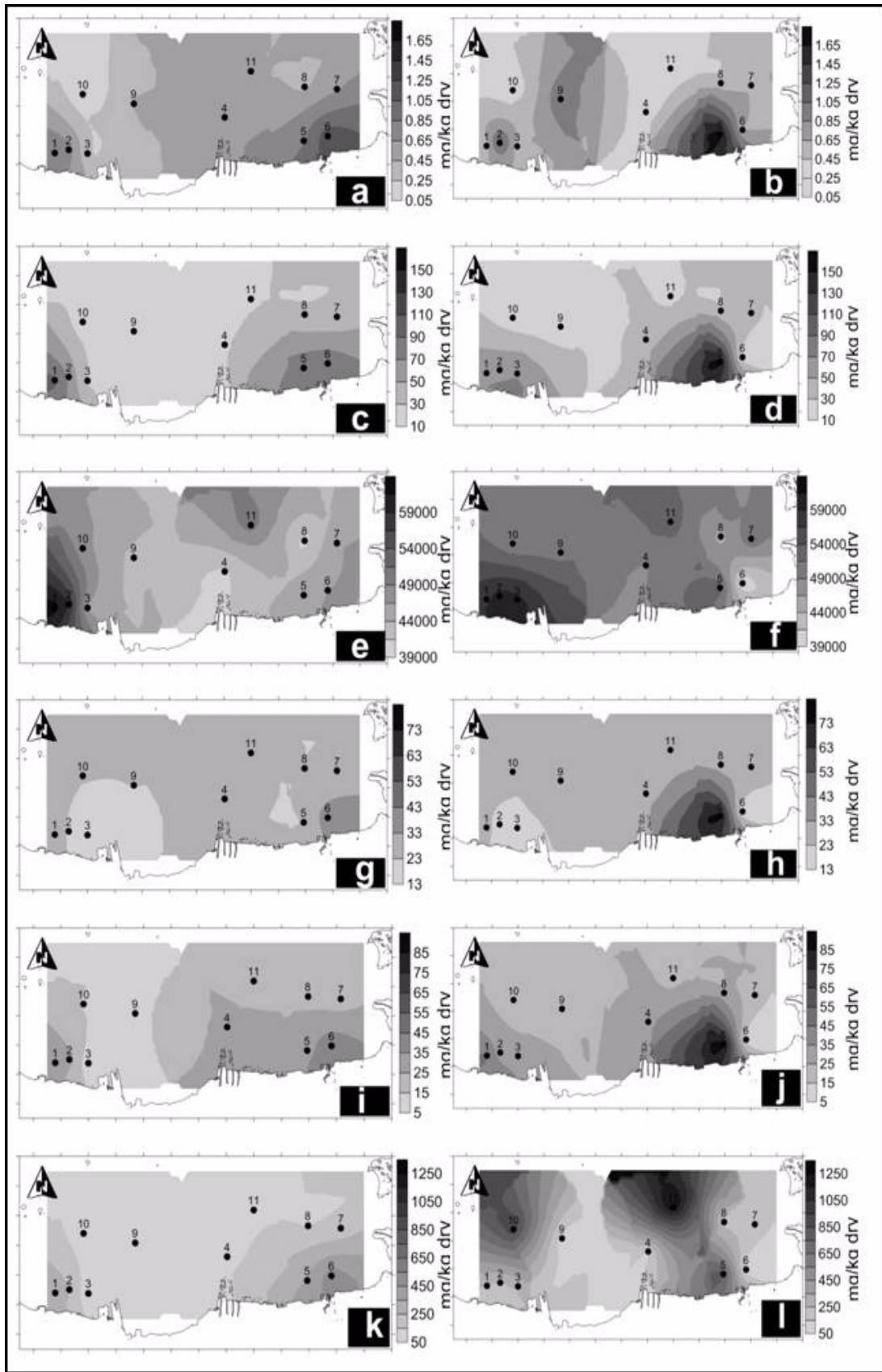


Figure 3. Spatial distribution of metals: Cd in March (a) and in June (b); Cu in March (c) in June (d); Fe in March (e) and in June (f); Ni in March (g) and in June (h); Pb in March (i) and Pb in June (j); Zn in March (k) and in June (l)

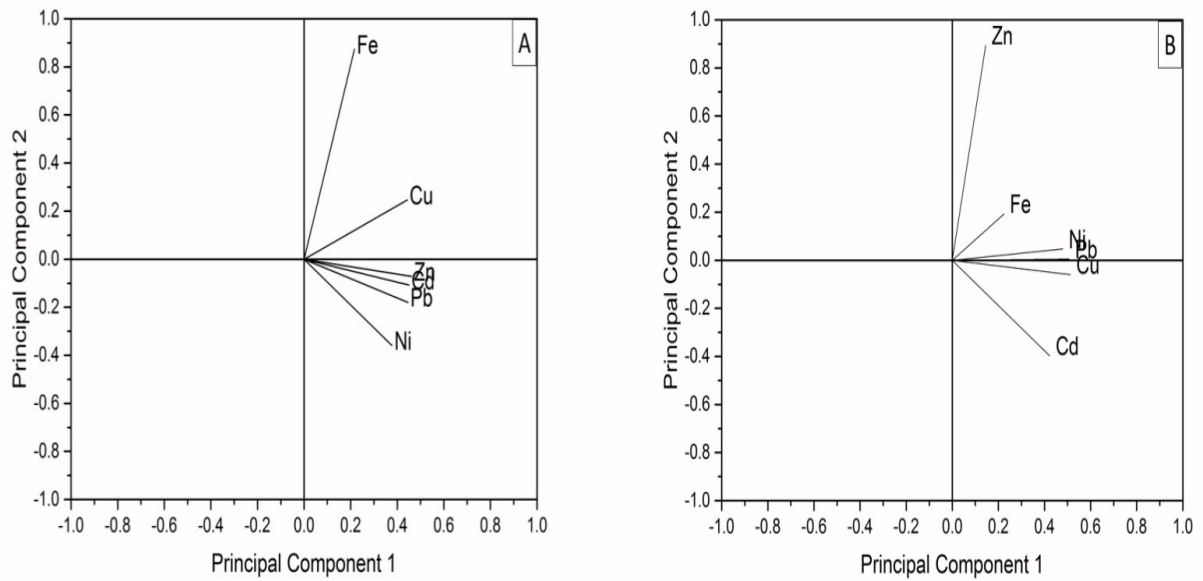


Figure 4. Loading plot of Principal Component Analysis for metals in sediment: A. March; B. June

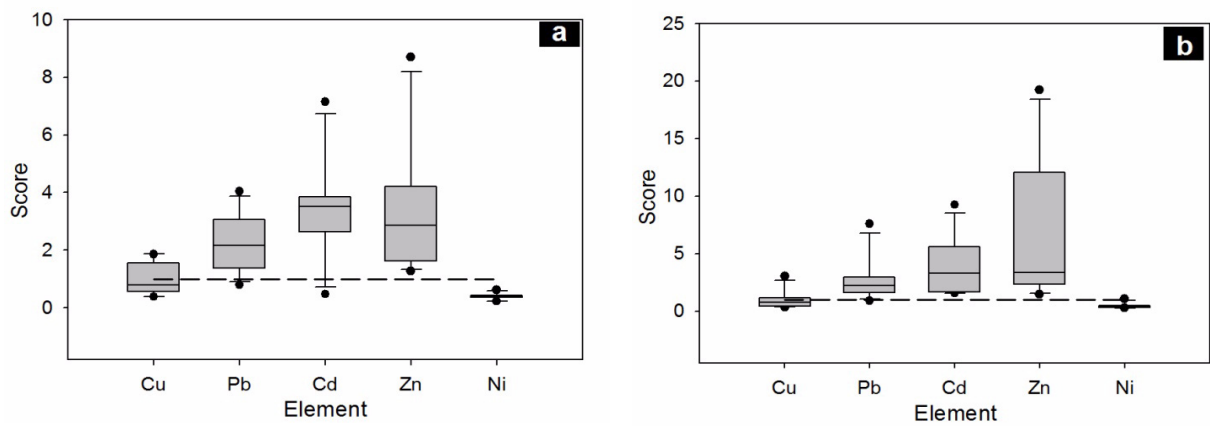


Figure 5. The box plot for EF score: a. EF in March; b. EF in June

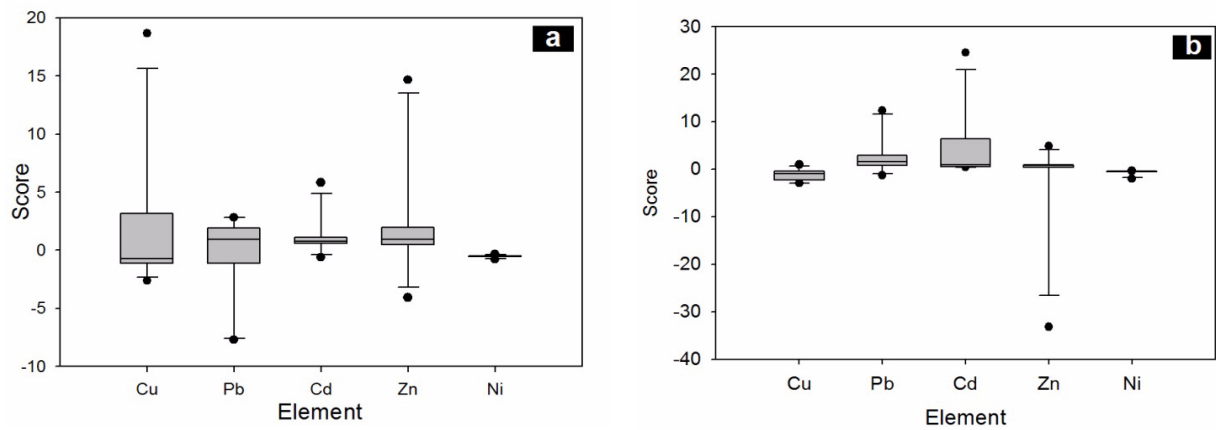


Figure 6. The box plot for Igeo score: a. March; b. June

Table 4. The previous studies of heavy metals concentration in Jakarta Bay and the comparison with other locations

| Location                   | Element, mg/kg dry |             |             |             |             |              | References                 |
|----------------------------|--------------------|-------------|-------------|-------------|-------------|--------------|----------------------------|
|                            | Cd                 | Cu          | Fe          | Ni          | Pb          | Zn           |                            |
| Jakarta Bay (2003)         | <0.001-0.44        | 0.79-193.75 | -           | 0.42-128.47 | 0.25-77.42  | 71.13-128.47 | (Rochyatun and Rozak 2007) |
| Jakarta Bay (2006)         | -                  | 9.70-11.8   | -           | -           | 18.60-21.20 | 44.90-61.90  | (Hosono et al. 2011)       |
| Jakarta Bay (2010)         | -                  | 10.80-107   | -           | -           | 13-106      | 85-845       | (Riyadi et al. 2012)       |
| Shantou Bay, China         | 0.67               | 48.52       | -           | 22.95       | 51.63       | 153.28       | (Qiao et al. 2013)         |
| Pearl river estuary, China | -                  | 6.2-100     | -           | 10.60-54.10 | 16.0-96.30  | 55.10-268    | (Ip et al. 2007)           |
| South yellow sea, China    | 0.06-1.54          | 6.0-32.90   | -           | -           | 6.2-39.30   | 24.60-244    | (Yuan et al. 2012)         |
| Beibu Bay, China           | 0.16               | 58.26       | -           | -           | 27.99       | 67.28        | (Dou et al. 2013)          |
| Nakaumi, Japan             | -                  | 8-51        | -           | 7-50        | 12-35       | 42-208       | (Ahmed et al. 2010)        |
| Honjo, Japan               | -                  | 9-39        | -           | 6-31        | 13-34       | 41-167       | (Ahmed et al. 2010)        |
| Hungli estuary, Bengal     | -                  | 4.30-45.29  | -           | 6.86-52.45  | Nd-44.47    | 22.96-204.99 | atterjee et al. 2007)      |
| Jakarta Bay (March 2013)   | 0.08-1.18          | 16.40-94.80 | 40678-62313 | 13.70-38.4  | 8.10-41.70  | 72.10-503    | This study                 |
| Jakarta Bay (June 2013)    | 0.29-1.74          | 16.60-157   | 39573-60300 | 15.90-75.8  | 7.800-89.4  | 85.30-1270   | This study                 |

wave marking the rainy season played secondary effect on the metal distribution. Despite of higher concentration close to the river mouth, the metal was distributed uniformly in the bay.

The anthropogenic input played a significant impact on metal concentration in the bay, indicated by the higher concentration of metals in the location near the shoreline and the value of enrichment factor. The Enrichment factor indicated the state of normal to severe enrichment. The massive anthropogenic input of metals often occurred as it is also reported in various studies such as the Shantou Harbor and Shantou City discharge in China became the main source of metals in sediment (Qiao et al., 2013). The variety of river flow influenced the fluctuation of its contained organic matter. Metals tend to be adsorbed into organic matter which introduced into the estuary and coastal area by river and sewage (Alagarsamy, 2006).

The difference of anthropogenic activities between western and eastern part of the bay could also cause this distribution. The power plant, harbor, and industrial activities are observed in the eastern part of the bay, whereas harbor is not found in the western part. Generally, these activities appeared as major metal sources (Horta-Puga et al., 2013). Several anthropogenic activities lead to the escalation of metals abundance. The main activities that release heavy metals into the Jakarta Bay are probably contributed by the consumption of oil and its derivatives. The harbor and power plant were supported in the massive utilization of fuel. The rise of lead (Pb) concentration in Deception Bay, Australia was promoted by fuel combustion and enhancement of Zn concentration mainly caused by boat building yards and anchorage (Brady et al., 2014). Lead (Pb) is an extremely stable element and very toxic to human and animals (Nobi et al., 2010). In addition, fuel combustion may become a

source of cadmium. Agricultural activities also bring potency in releasing Cadmium into the environment. Xu et al. (2014) reported the application of intensive phosphate fertilizer releasing a significant amount of Cd into the aquatic environment.

## CONCLUSION

The temporal distribution of metals indicated insignificant difference between March and June since the short time lag and the both months were in transitional season. In addition, the metals were evenly distributed in March than June. Moreover, the distance of the location to the source of metal negatively influenced the concentration. Hence, metals concentration in Jakarta Bay indicated enrichment status possibly due to anthropogenic activities, and it is classified as unpolluted to moderate polluted.

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