Assessing Heavy Metals Contamination in Suspended Particulate Matter in Jakarta Bay, Indonesia

Penilaian Pencemaran Logam Berat dalam Materi Partikulat Tersuspensi di Teluk Jakarta, Indonesia

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ABSTRACT: Suspended Particulate Matter (SPM) is an important compartment within water column due to its capability in adsorbing pollutant like heavy metals. However, there was limited information regarding SPM and its heavy metals content in Jakarta Bay. Therefore, this study was proposed to understand the spatial distribution, source and to assess metal content in SPM in Jakarta Bay. The samples were collected from 24 stations in April 2011 and were then analyzed using acid digestion processes adopted from USEPA 3050b. The generated data were then modeled to determine the spatial distribution of metals in SPM. The result revealed that the concentration of Cd, Cu, Ni, Pb and Zn in SPM were as follow: 10-110; 21-472; 14-356; 7-107; 87-4255 mg.kg⁻¹, respectively. Since Cd, Pb and Zn in SPM were majorly concentrated in the area closed to harboring activities, the activities was suspected in contributing of high input of those metals, meanwhile, Cu and Ni was mostly came from riverine runoff. The computation of Index of Geoaccumulation (Igeo) revealed that the major area in Jakarta Bay was unpolluted by Cu, Pb, Zn, and Ni in SPM however 96% of the selected area were strongly contaminated by Cd in SPM. Thus, this study emphasized that harbor area as a dominant source of metals in SPM in Jakarta Bay and required to be well managed.

Keywords: Jakarta Bay, Suspended Particulate Matter, heavy metals, contamination.

INTRODUCTION

Intensive anthropogenic activities in recent decades accelerate the elevation of pollutants (El-Sorogy and Youssef, 2015; Rahman and Singh, 2016). Metals become one type of the hazardous pollutants in an environment due to their persistence, recalcitrance, toxicity and bioaccumulation properties (Kang et al., 2015; Li et al., 2013). In a marine environment, the coastal area is a primary receiver of metals through riverine runoff (Kontas, 2007; Mohammed et al., 2012). Originally, a river is transporting metals in many forms i.e. metal soluble phase, colloid phase and adsorbed phase like on particulate matter and sedimentary form (Huang et al., 2012). However, in an estuary where freshwater and saline water meet each other, those metals change its behavior and many complex processes such as adsorption-desorption, photocatalytic reaction, absorption, chemical reaction, and deposition-
Redeposition are involving in the alteration of metals form (el-Nemr et al., 2006; El-Said et al., 2014; Sun et al., 2015).

Many rivers exhibit a high content of suspended particulate matter (SPM) consisting of either oxide or organic particle (Dali-youcef et al., 2006; Wang et al., 2016). One of the most substantial attention comes to the capability of the SPM in adsorbing metals. The SPM experience an equilibrium state between donating and receiving metals from/to dissolved phase. The fluctuation of metals content on the SPM occurs in the response to that process (Buggy and Tobin, 2008). Moreover, finer SPM can interact and bind together making a bigger aggregate as the physicochemical character of the waters changed. As the SPM is getting bigger, the gravity is attracting that SPM downward into bottom sediment. In a coastal region with low disturbance, that deposited SPM is highly likely to be buried deep in the bed sediment and act as metals archive (Liu et al., 2016). However, when that waters receive any disturbances like tidal movement, hurricane incident, benthic burrowing organism activities, the changes of physicochemical properties of sediment, those deposited SPM may be re-suspended into above water column acting as a source of metals pollution (Han et al., 2017). Indeed, the continuity of these processes will eventually affect the net concentration of the metal content in SPM in the coastal ecosystem like in Jakarta Bay.

Jakarta Bay is objected to a heavy anthropogenic stressor and experienced a degraded condition due to high load of chemical and biological pollutant (Retno et al., 2016). The Bay is located close to the capital city, the megacity of Jakarta, home to over 30 million inhabitants (Ladwig et al., 2016). The city is reflected by the massive number of motorcycle and private car utilization, poor waste disposal management in the residential and industrial area, and an active harbor activity driving an intensive discharge of anthropogenic efflux (Rinawati et al., 2012). In addition, the Jakarta Bay is receiving the freshwater input from 13 rivers carrying anthropogenic waste and pollutant like heavy metals from the land (Hartati et al., 2016; Rinawati et al., 2012). As regards to those many heavy metals sources in the bay and massive riverine input, metal content in SPM may become serious pollution in the Jakarta Bay especially metal associated with anthropogenic activities like Cd, Cu, Ni, Pb, and Zn. Therefore this study was aimed to understand the spatial distribution of Cd, Cu, Ni, Pb and Zn in SPM, and to assess the condition of metal content in SPM.

Material and Methods

Sampling Site

The collection of SPM was carried out in the Jakarta Bay at the geographical position of 106°45'E-106°58'E and 6°7'S-5°55'S. The samples were collected from 24 stations spreading from river mouth to the center of the bay in April 2011. In this study, nine (9) major rivers ended in the Jakarta Bay were selected as the representative of the suspected source of SPM. As regards to their position, from western part to eastern part, those rivers can be put in such an order: Dadap, Kamal, Cengkareng, Angke, Ciliwung, Sunter, Cakung, Cikeas/CBL, Citarum. Figure 1 represented the illustration of sampling points/stations and the location of each river.

Sample and Data Collection

The physicochemical properties of seawater were directly measured in the field. pH value was recorded...
using cybersan pH 300 series. On the other hand, salinity was measured using CTD SBE 19 and the data acquisition was confirmed using SEASAVE from Sea-Bird Electronic Inc. and converted using DATCNV and BINAVG program.

The term of suspended particulate matter (SPM) in this study represents all solid material in water column larger than 0.45 \( \mu m \) in size following the study of Nguyen et al., (2005). Surface water was collected from the observation points using pre-cleaned Van dorn water sampler at 1 m in depth. Then, 1 L of that water was filtered using pre-cleaned whatmann® filter paper, cellulose-Nitric grade, and 0.45 \( \mu m \) of pore size. The separated SPM was sealed in a petri dish and preserved at -20\( ^\circ \)C.

A method based on acid digestion procedure was executed to analyze the metal content in SPM following the protocol of Loring and Rantala (1992). This method mainly employed acids i.e. HNO\(_3\), H\(_2\)O\(_2\), and HCl to dissolve metals in SPM. All acids were purchased from Merck, Darmstadt, Germany. The whole processes were carried out under reflux system operated at 95 \( ^\circ \)C for at least 8 hours. In this procedure, the addition of acid came in sequence. The first was the addition of 10 mL of HNO\(_3\), followed by the addition of 20 mL of H\(_2\)O\(_2\) and the last was the addition of 10 mL of HCl. After those acid digestion steps completed, the filtrate was injected to FAAS (Flame Atomic Absorption Spectrophotometer) Varian SpectrAA 20-plus® for the measurement of Cd, Cu, Ni, Pb, and Zn. All glassware was immersed in HNO\(_3\) (1+1) prior. The quality control data were presented in Table 1. The accuracy test was performed by analyzing certified material PACS-2® manufactured by National Research Council Canada and the precision test was carried out using replication of sample.

### Data Analysis

The pH, salinity, SPM concentration, metal concentration in SPM was presented in MAP generating using GIS 10.1 software (product of ESRI, Redlands, California, USA). The relation of those aforementioned variables was presented using Pearson’s correlation. The assessment of metal concentration in SPM was computed using Index of Geoaccumulation (\( I_{geo} \)) (Caeiro et al., 2005) formulated as follow:

\[
I_{geo} = \log_2 \frac{C_{sample}}{1.5 \times C_{background}}
\]

The \( I_{geo} \) is widely used to assess metal in sediment, however, from another study, the index was also powerful to assess metal concentration in SPM (Suja et al., 2017). The background concentration was selected from average metal concentration in riverine SPM as listed by Viers et al., (2008). The score of \( I_{geo} \) was interpreted as follow: <0-1 is uncontaminated, 1-3 signify moderate pollution, 3-5 represent strong pollution and >5 denote extreme pollution (Suja et al., 2017).

### RESULTS

#### The Characteristics of Sea Water

The physicochemical properties like salinity and pH were directly recorded during the field sampling, parallel with the collection of seawater sample. The set of data revealed that the salinity was varied from 2 to 32 psu, and pH ranged from 6.7 to 8.2, as presented in Figure 2. The low salinity was detected at the river mouth, i.e. Angke River (2 psu), Dadap River (9 psu), Sunter River (6 psu) and Ciliwung River

![Figure 2](image-url)

**Figure 2.** The distribution of (a) sea surface salinity and (b) pH during the observation time.
(6 psu). Opposite, the middle of Jakarta Bay reflected seawater salinity (28-31 psu). Meanwhile, pH of the surface water was distributed uniquely in which pH < 7 was detected only in 3 stations. Interestingly, those stations were located in the western part of Jakarta Bay i.e. in the river mouth of Angke River and Dadap River, however, all river mouth in the eastern part of the bay exhibited pH > 7. The neutral pH values (7.2-7.5) were observed in the river mouth of Cakung to Citarum in the eastern part of the bay.

One of the important variables in this study was the quantification of SPM from each station and Figure 3 was a model for the spatial distribution of the SPM in the Jakarta Bay. In that model, the SPM was concentrated close to the coastal area. The highest SPM concentration (106 mg/L) was observed right in the River Mouth of Angke. This value was extraordinary high compared to other river mouths such as the river mouth of Dadap River (the SPM concentration was 66 mg/L) and River Mouth of Ciliwung (SPM concentration=33 mg/L). In general, the SPM concentration in the middle of the bay was ranged from 9.8-12 mg/L.

Metals Concentration

The trace metals consisting of Cd, Cu, Ni, Pb and Zn in SPM were analyzed based on acid digestion procedure. Figure 4 was box-whisker plot reflected the range of the data set of metal in SPM and from the data set, those metals concentration in SPM can be arranged in such order: Zn>Ni>Pb Cu>Cd.

The data set of metal in SPM was also modeled using ArcGIS 10.1® to get a better illustration of their spatial distribution and the result was presented in Figure 5. Those two figures revealed that each metal in SPM was showing different behavior. Cd in SPM was ranged from 10-110 mg.kg\(^{-1}\) and extreme concentration was observed only in one station closed to Priok Harbor. However, other stations exhibited relatively lower Cd concentration (< 50 mg.kg\(^{-1}\)). Cu in SPM was ranged from 21-472 mg.kg\(^{-1}\) and was concentrated in front of Angke River. The area next to Priok Harbor also showed relatively higher Cu concentration. Comparing with previous metals, Ni in SPM displayed different distribution pattern. Rather than concentrated in Angke river mouth or Priok Harbor, Ni was more discharged from the eastern part of the bay right in the river mouth of Citarum (concentration=356 mg.kg\(^{-1}\)) and gradually decreased toward the sea. The lowest concentration of Ni in SPM (14 mg.kg\(^{-1}\)) was recorded in the western part of the bay at station 9.

The distribution of Pb in SPM was similar to the distribution of Cd in SPM, although the main source was different. Pb in SPM was concentrated in the front of river mouth of Ciliwung (302 mg.kg\(^{-1}\)) and Sunter (296 mg.kg\(^{-1}\)). The two river mouths were located next to Priok Harbor. Surprisingly, the lowest Pb in SPM value (7.6 mg.kg\(^{-1}\)) was measured in front of Citarum River. Comparing with other metals, the distribution of Zn was similar with Cd and Pb in which Zn in SPM was concentrated in the area of Priok Harbor (concentration=4255 mg.kg\(^{-1}\)).

The relation of metals and physicochemical parameter was presented in Pearson’s correlation. The analysis of Pearson’s correlation was carried out based on the confidence level at 95% and the result was presented in Table 2. From that correlation, this study...
Figure 5. The distribution of metals: (a) Cd, (b) Cu, (c) Ni, (d) Pb, (d) Zn in Suspended Particulate Matter (SPM)
found that salinity was positively correlated with pH (significant, p<0.05), however, these two parameters were negatively correlated with SPM (significant, p<0.05). Among those metals, Pb was positively and significantly correlated with Cd and Zn. Surprisingly, all metals in SPM were insignificantly correlated with SPM. Comparing with other locations, metals in SPM in Jakarta Bay was as high as metals in SPM from other estuaries excluding Cd. Cd in SPM in Jakarta Bay was relatively higher, especially in Priok Harbor. The list of metals in SPM in other locations was presented in Table 3.

Table 2. The Pearson's correlation describing the correlation of salinity, pH, the concentration of SPM and concentration of metals in SPM.

|        | salinity | pH      | SPM     | Pb     | Cd     | Cu     | Ni     |
|--------|----------|---------|---------|--------|--------|--------|--------|
| pH     | 0.64*    |         |         |        |        |        |        |
| SPM    | -0.73*   | -0.56*  |         |        |        |        |        |
| Pb     | -0.29    | 0.10    | -0.08   |        |        |        |        |
| Cd     | -0.17    | 0.06    | -0.31   | 0.44*  |        |        |        |
| Cu     | -0.24    | -0.33   | 0.17    | -0.21  | -0.14  |        |        |
| Ni     | -0.04    | 0.13    | 0.006   | -0.03  | 0.21   | 0.12   |        |
| Zn     | -0.34    | -0.07   | 0.06    | 0.51*  | 0.24   | 0.03   | -0.13  |

*p<0.05
Table 3. The concentration of metals in SPM in other estuaries. BDL was Below Detection Limit Value and WRSPM represented the average of world river concentration of metals on SPM.

| Location                        | Cd   | Cu   | Ni   | Pb   | Zn   | Ref                      |
|---------------------------------|------|------|------|------|------|--------------------------|
| Kongsfjorden, Svalbard, Norway   | 0.45-19 | 26.7-187 | 13.5-81.6 | 5.6-66 | 68-406 | (Bazzano et al., 2014) |
| Kongsfjorden, Norwegian Arctic  | 0.22-13.5 | 43-609 | 15-148 | 22.3-212 | 145-1216 | (Bazzano et al., 2017) |
| Changjiang Estuary, China       | 0.14-0.75 | 40-70  | -    | 25-38 | 144-238 | (Yao et al., 2016)      |
| Mejerda Delta, Tunisia          | 0.4-2.6 | BDL-39.1 | 65.6-92.0 | 38.1-193.8 | 66.6-142.5 | (Helali et al., 2016) |
| Kali Estuary, India (pre-       | -    | 55.7-181.7 | 95.1-439.7 | 27.5-112.3 | 133.4-730.7 | (Suja et al., 2017) |
| monsoon)                        | -    | 93.5-209.3 | 121.6-266.1 | 30.8-583 | 142.3-414.9 |
| Average World River SPM (WRSPM) | 1.55 | 75.9  | 130  | 21.1-203.4 | 177.6-1678 | (Viers et al., 2008) |
| Jakarta Bay (1995)              | -    | BDL-403 | BDL-105 | BDL-904 | BDL-788 | (Williams et al., 2000) |
| Jakarta Bay                     | 10-110 | 21-472 | 14-356 | 7-107 | 87-4255 | This study              |

The Igeo was computed for assessing metals level in SPM. The analysis was carried out by neutralizing the data set of metals observed in this study with the background concentration and the distribution of Igeo score was presented in Figure 6. The score value was spreading from <0 to 4 implied the condition of metal in SPM was between uncontaminated to strongly polluted. The worst condition was contributed by Cd in SPM in which the score was ranged from 2.12 to 4.5 (moderate to strong pollution). Meanwhile, other metals were leading to unpolluted to strong pollution. Ni and Pb in SPM unpolluted the bay in each station, however, Cu and Zn were causing up to moderate pollution. Cu in SPM was scored from -2.5 to 2.05, Ni score fell between -3 and 1.7, Pb was scored between -3.5 to 1.7 and Zn donated score of -1.9 to 3.8.

DISCUSSION

Jakarta Bay is a complex coastal system for it is a shallow estuarine with numerous rivers ends to the bay. Generally, the input of that massive freshwater contributes to the alteration of physicochemical properties of seawater featured by the exchange between freshwater and seawater (Oursel et al. 2013). In this study, SPM has a negative and significant correlation with pH and salinity. That was triggered by the difference of source. Since the source of SPM was majorly contributed by riverine runoff, the SPM was observed higher in the river mouth and gradually decreased toward the sea. Opposite, high value of pH and salinity were donated by sea water. Thus, as a result of water mixing, pH and salinity value was lower in the river mouth and gradually increased toward the sea. The
SPM concentration in river mouth and sea water of Jakarta Bay was comparable with other estuarine like in the Mediterranean Sea. In Huveaune waters, Mediterranean, SPM concentration was 9.8-83 mg/L in river mouth and 4.5 mg/L in sea water (Oursel et al., 2013). In Tunis, Tunisia, SPM concentration was observed at 90 mg/L during the rainy season and 40 mg/L during the dry season (Helali et al., 2016).

Surprisingly, Pb, Cd, and Zn in SPM were positively and significantly correlated one another. This study suspected that the source of high Pb, Cd and Zn content in SPM came from the same source. This assumption was constructed from the evidence of the distribution pattern. The spatial distribution revealed that those metals were concentrated in Priok Harbor, river mouth of Sunter and Ciliwung. Priok Harbor is considered as the biggest harbor in Indonesia. Hence, considering the location, Priok Harbor is stationed next to Priok Harbor that was also suspected to contribute in metals input to the bay. Those two anthropogenic activities were massively utilizing fossil fuel as the main core for their activities. Fossil fuel like ores or oil was majorly stimulated metals contamination in many oceans especially estuarine area (Chae et al., 2006). This study was in-line with the finding of Pekey (2006) confirming that Cd, Pb, and Zn were exhibiting the similar properties and can be used as a tracker of specific industries like paint industry.

The investigation of this study revealed that the source of Pb, Cd, and Zn in SPM suspected mainly came from those harboring activities. Theoretically, in an estuary with a dominant riverine input, physicochemical variables affect the adsorption-desorption equilibrium of metals on SPM (Feng et al., 2017; Shigemitsu et al., 2013) and as the result, pH and salinity gradient toward the sea will eventually alter the metal content in SPM due to competitive binding between metals and free ion in seawater (Gao et al., 2015). Hatje et al., (2006) explained the effect of a strong electrolyte like NaCl in the binding capability of metal in SPM (Hatje et al., 2006). The presence of major cation like Na⁺, Ca²⁺, and Mg²⁺ also stimulates the competitive condition between metals and particulate matter. The unique distribution of Cd, Pb, and Zn was also discovered in Changjiang Estuary, Cina. In that estuary, Those 3 metals in SPM was reported low in the inner region and high in the outer region of the estuary (Yao et al., 2016). They explained that two factors influenced that kind of distribution: (1) metals released by ionic exchange or degradation of its organic matrix; and (2) the adsorption of metals onto either other existing particles or newly formed matrix.

Metals in SPM in Jakarta Bay were comparable with other metal contents in SPM from other estuaries. However, Cd in Jakarta Bay was significantly higher than other estuaries. In comparison, Cd in SPM in Kongfjorden, Norway was <1/20 lower (0.22-5.6 mg/kg) than that Cd observed in this study (Bazzano et al., 2017) and the average riverine metal concentration in SPM around the world was 1.5 mg/kg (Viers et al., 2008). Thus, Cd in SPM in Jakarta Bay can be acknowledged as one of the threatening pollutions in the bay and the computation of Igeo was applied to get detailed information about the pollution condition in the bay.

Originally, Igeo was used to assess the metal contamination in sediment from marine environment (Chatterjee et al., 2007; Li et al., 2013; Varol, 2011; Xu, et al., 2014). This index was computed by measuring the metals concentration with the concentration of metals from reference. The reference value was usually selected from a clean area, average metals concentration in the earth crust, and metal concentration during the pre-industrial era (Caeiro et al., 2005; Wang et al., 2013). However, some studies on metal content in SPM were utilizing Igeo to assess the contamination level (Suja et al., 2017). This calculation was computed based on the comparison of metal concentration with average world river of SPM (WRSPM) (Viers et al., 2008).

The computation of the Igeo revealed that Cd and Zn became a dominant pollutant in the bay especially in the area of Priok Harbor meanwhile Cu, Pb and Ni were insignificant pollutants in the bay. From the computation, this study found that 96% of observation point in the bay was strongly polluted by Cd in SPM. Commonly, Cd was the anthropogenic origin and the escalated level of Cd in the environment is mostly triggered by the increased volume of anthropogenic activity (Wang et al., 2014; Wang et al., 2013). Zn exhibited an over-layer concentration in the area closed to Priok Harbor leading to strong pollution of Zn in Priok Harbor. However, 16% of stations were at the moderate pollution and 80% of observation point was still uncontaminated by Zn. Moreover, Ni and Pb indicated an identical condition in which 16% of the stations was moderately contaminated, and 84% of stations were uncontaminated by these two metals. In addition, most of the stations were uncontaminated by Cu. The finding of this study emphasized that Priok Harbor was a noticeable source of contamination in Jakarta Bay especially in the term of metals pollution in SPM.
CONCLUSION

The physicochemical variable was affected by the presence of river resulting SPM was higher in inner region and lower in outer region, opposite with pH and salinity. Uniquely, those physicochemical variables were uncorrelated with metals concentration in SPM. The Priok Harbor was the main suspect of the pollution due to the concentrated of metals in SPM in the harbor area, especially for Cd, Pb and Zn. In order to get better picture on the contamination level in the bay, Index Geoaccumulation (Igeo) was computed. The Igeo revealed that most of the metals in Jakarta Bay were in uncontaminated condition. The index also described that 96% of stations in the bay was strongly contaminated by Cd in SPM. The harbor area was contributed in that contamination level especially the contamination of Cd in SPM. The profound studies on the source of metal in SPM, the effort on controlling the source and the remedial of the pollution are going to be the promising step in the near future on environmental management in Jakarta Bay.

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