Spatial Distribution of Heavy Metals in the Surface Sediments of the Southern Coast of Pacitan, Indonesia

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Received February 24, 2017; Accepted September 20, 2017

ABSTRACT

This study was conducted to analyze the spatial distribution of heavy metals from four different coastal areas in southern Pacitan, Indonesia namely Pantai Watu Karung (WK), Pantai Teleng Ria (TL), Pantai Pancer (TP) and Pantai Soge (SG). Data collected in this study included: temperature, salinity, DO, pH, sediment, organic matter and heavy metals in the sediments (Pb, Hg and Cd). The results showed different distribution patterns of heavy metals. Heavy metal concentrations, especially Pb and Hg, were found to be higher in Pantai Soge, while the concentration of Cd was higher in Pantai Pancer. An ANOVA test showed the distributions of Pb and Cd were significantly different ($p < 0.01$) between sampling sites. Variability of the physicochemical parameters influenced the variabilities of heavy metal concentrations among sampling sites. Overall, heavy metal concentrations in the study areas were rather low; however, attention was needed due to heavy activities in the coastal areas of southern Pacitan that can contribute to heavy metal pollution.

Keywords: *heavy metals; environmental factors; coastal areas; Pacitan; Indonesia*

ABSTRAK

Penelitian ini bertujuan untuk menganalisa distribusi spasial logam berat pada empat wilayah pantai yang berbeda di perairan selatan Pacitan, Indonesia, yaitu: Pantai Watu Karung (WK), Pantai Teleng Ria (TL), Pantai Pancer (TP) dan Pantai Soge (SG). Data yang diukur meliputi suhu, salinitas, DO, pH, ukuran sedimen, bahan organik dan juga logam berat di sedimen (Pb, Hg dan Cd). Hasil penelitian menunjukkan pola distribusi logam berat yang berbeda antar pantai di lokasi penelitian. Konsentrasi logam berat Pb dan Hg ditemukan lebih tinggi di Pantai Soge, sedangkan konsentrasi Cd ditemukan lebih tinggi di Pantai Pancer. Analisa statistik ANOVA menunjukkan distribusi Pb dan Cd berbeda signifikan di antara stasiun penelitian ($p < 0.01$). Variabilitas faktor fisika-kimia lingkungan mempengaruhi variabilitas logam berat yang ditemukan. Secara keseluruhan, konsentrasi logam berat di daerah penelitian masih cukup rendah, namun perhatian tetap diperlukan karena tingginya aktifitas manusia di perairan selatan Pacitan yang dapat mengakibatkan polusi logam berat.

Kata Kunci: *logam berat; faktor lingkungan; pantai; Pacitan; Indonesia*

INTRODUCTION

The accumulation of heavy metals in sediments is one indicator of environmental pollution. Heavy metals enter aquatic systems from natural sources (i.e. weathering of rock and soil), as well as from anthropogenic activities [1-3]. Aquatic sediments accumulate heavy metals by adsorption or precipitation processes. This accumulation depends on the size characteristics of the sediments [1,4]. There is a linear correlation between heavy metal concentration and the fraction of particles, i.e., metals increased with decreasing grain size of sediments [1]. A study conducted in coastal waters of Marseille found higher concentrations of heavy metals in the finest fraction of sediments [5]. The accumulation of heavy metals in sediments also depends on hydrographic conditions such as temperature, salinity, pH and DO. Heavy metal concentrations were greatly affected by changes in pH and sediment disturbance (by physical mixing) rather than by changes in dissolved oxygen concentration or salinity [6]. Moreover, pH controls the adsorption and precipitation process of heavy metals in the sediment [7]. A low pH value or acidic environment increases heavy metal concentrations because heavy metals are generally more mobile when pH is less than 7 [8]. In addition, heavy metal concentrations have a significant correlation with organic matter content as dissolved and particulate organic matter can act as a scavenger for metals that accumulate in bottom sediment [1,9].

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DOI: 10.22146/ijc.22400

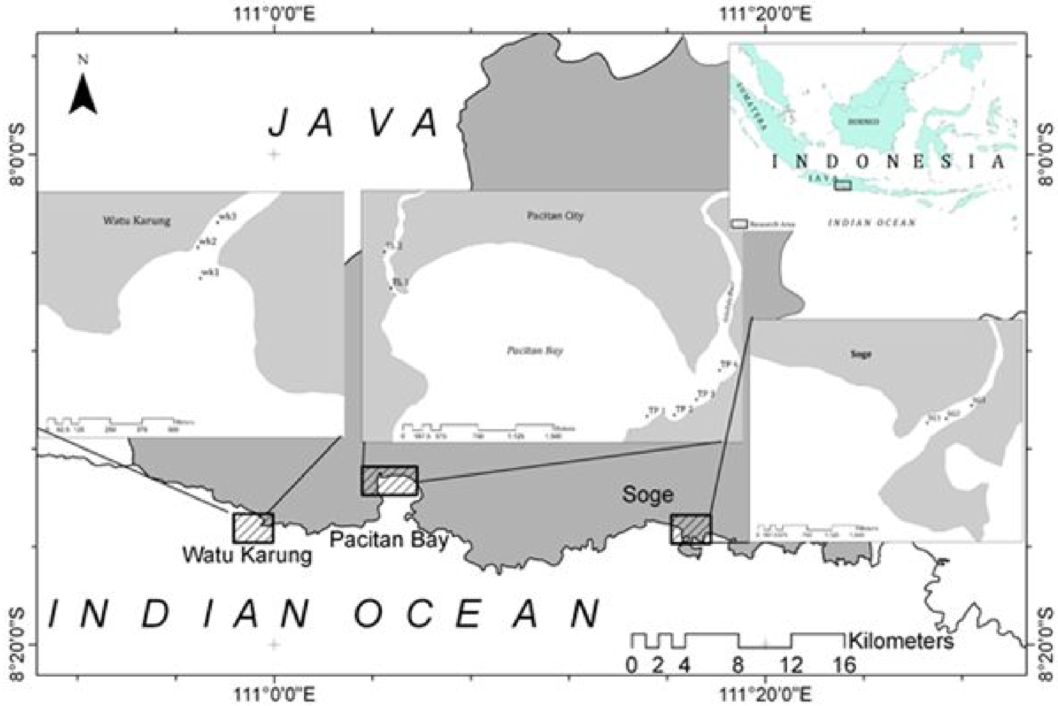

Fig 1. Study areas are four different coastal areas in the southern region of Pacitan: Pantai Watu Karung (WK); Pantai Teleng Ria (TL) and Pantai Pancer (TP) at Pacitan Bay; and Pantai Soge (SG)

Long-term accumulation of heavy metals in sediments can have significant effects on organisms and cause toxicity. Toxicity levels of heavy metals have been studied extensively [3,10]. Pb, Hg and Cd are known to be toxic elements and they cannot be decomposed. Therefore, these heavy metals can cause harm to the environment and humans.

The distributions of heavy metals have been extensively studied and reported elsewhere [1,11-13]. However, studies on the distribution of heavy metals in Indonesia, especially in the southern ocean, are scarce, with limited publication. The objectives of this study were to understand the distribution of heavy metals (Pb, Hg and Cd) in the surface sediments of the coastal waters of Pacitan and to describe what factors caused the distribution. The study areas were located in the southern part of the Indian Ocean lying between 111º0'00"E-111º05'00"E and 8º12'00"S-8º15'00"S in the region of Pacitan, East Java, Indonesia (Fig. 1). There were four different coastal areas: Pantai Watu Karung (WK) which is small fishing port close to a mangrove habitat; Pantai Teleng Ria (TL) which is an enclosed and recreational beach at Pacitan Bay; Pantai Pancer (TP) which is on the other side of Pantai Teleng Ria at Pacitan Bay; and Pantai Soge (SG) which is close to aquaculture (shrimp ponds). These areas have significant input from human activities and city development such as the construction of industrial and residential estates, recreational facilities and the fishing industry. They are subjected to negative impacts of pollutant discharge such as elevated heavy metals in the water column and sediments.

EXPERIMENTAL SECTION

Sampling Collection

Samples were collected from four different coasts with 2-4 stations on each coast along the southern areas of Pacitan, Indonesia (Fig. 1). Environmental data collected were temperature, salinity, dissolved oxygen and pH, and these were measured using a thermometer, salinometer, DO meter and pH meter, respectively. Surface sediment samples were collected using an Ekman grab and in order to avoid any leakage of the fine material, the grab was firmly closed and only opened upon arrival on the boat. Sediment samples were taken from the inner part of the grab to minimize metal concentration from the grab`s wall using polyethylene (PE) spoon. The samples were placed in tightly sealed plastic containers. The Ekman grab and PE spoon was cleaned using distilled water between sampling sites to avoid contamination from the previous sample. Measurements of the environmental data and sediment samples were conducted in the surface layer at 5–10 cm depth. About 1 kg of sediment of samples were collected for analysis of grain size, heavy metal content and organic matter. The collected

Table 1. Mean (± SD, n = 3) of the water quality parameters

Sampling station	Temperature (°C)	Salinity (psu)	Dissolved Oxygen (mg/L)	pH
WK	31.12 ± 1.44	22.67 ± 6.60	15.38 ± 0.92	7.92 ± 0.03
TL	27.65 ± 1.20	33.83 ± 4.01	13.15 ± 2.62	8.21 ± 0.09
TP	29.39 ± 2.67	29.67 ± 1.15	10.90 ± 1.15	8.36 ± 0.26
SG	31.68 ± 3.60	na	11.23 ± 5.39	8.12 ± 1.10

Note: na (not available)

Table 2. Granulometric composition of sediment and organic matter content (%)

Sampling station	Gravel (%)	Medium sand (%)	Fine sand (%)	Silt+Clay (%)	Organic matter (%)
WK	0.53	44.57	14.89	40.40	0.90
TL	0.20	3.80	79.81	16.20	0.54
TP	1.94	45.81	44.62	7.64	0.63
SG	1.18	58.88	38.49	1.45	0.52

samples were preserved in a cool box with dry ice until further analysis in the laboratory.

Sediment Analysis

Grain size analysis was performed by drying out the wet sediment samples in an oven at 105 °C for 4 h. About 100 g of the sediment samples were homogenized and passed through a range of sieves with different mesh sizes for grain size analysis (sand, silt and clay). The starting weight of the sample and the separated weight of each fraction were used to calculate the percentage size of each fraction. The organic matter content was determined by drying out sediment samples at 70 °C for 24 h. Dried sediment samples were then combusted in an oven at 550 °C for 4 h. Grain size and organic matter analysis were conducted according to [1,14].

For metals analysis, dried sediment samples were crushed to the finest possible fraction using an acid-washed pestle and mortar. About 2 g of the sample was digested at room temperature in a beaker with 10 mL of 70% reagent grade nitric acid overnight. The digested samples were then analyzed using a Shimadzu Flame Atomic Absorption Spectrophotometer Model AA-6800 using standards to allow determination of metal concentrations within each sample. To monitor the performance of the instrument, blank and standard solutions were used to obtain data quality by developing calibration curves.

Statistical Analysis

A one-way ANOVA was used to compare the metal concentrations of the sampling sites (significant values, $p \le 0.01$). All statistical calculations were carried out with SPSS 16.0 for Windows.

RESULT AND DISCUSSION

Hydrographic Conditions

Environmental data describing the hydrographic conditions of the sampling stations are shown in Table 1. The temperature of the study sites was in the range of 27.65–31.68 °C and the salinity was about 22.67–33.83 psu. The temperature range was the result of the different time of the measurement. However, since sampling was conducted on the same day, the difference of the temperature among sampling sites was not significant. The high range of salinity among the sampling stations was highly related to the input of freshwater from the river (WK and TP). Due to the error of the salinometer, salinity at Pantai Soge could not be measured. Dissolved oxygen was found in the range of 10.90–15.38 mg/L and the pH value was in the range of 7.92–8.36. Overall, the data showed quite similar values for hydrographic conditions among the sampling sites.

Grain Size and Organic Matter

The granulometric composition in the sediment samples from all stations showed a relatively high content of medium to fine sand fractions with low fractions of gravel (Table 2). Pantai Pancer and Pantai Soge were composed of similar fractions with a high percentage of medium to fine sand. On the other hand, the sediments of Pantai Watu Karung were mainly medium sand and silt+clay. Pantai Teleng Ria showed a very high content of fine sand (79.81%) compared to the other fractions that were present in a low percentage.

The highest distribution of organic matter was at Pantai Watu Karung with an average of 0.90% followed by Pantai Pancer (0.63%), Pantai Teleng Ria (0.54%) and Pantai Soge (0.52%). The mangrove habitat at the

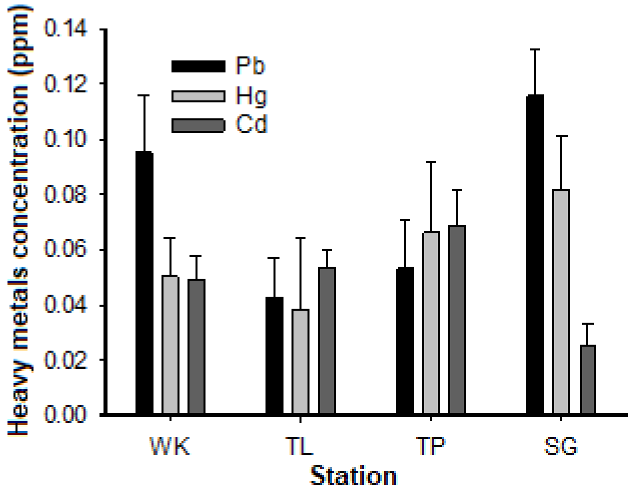

Fig 2. Surface distribution of Pb, Hg and Cd concentrations in the study areas

Table 3. One-way ANOVA showing variation in heavy metals and organic matter between study sites

Source	Df	F	p
Site	3	15.07	< 0.01
Pb	8		
Site	3	2.36	0.15
Hg	8		
Site	3	13.34	< 0.01
Cd	8		
Site	3	5.66	0.02
Organic matter	8		

Pantai Watu Karung sampling site might be the reason for the high percentage of organic matter in this area. In addition, the high percentage of silt+clay in this sampling site was also the result of a high percentage of organic matter. For the other three sampling sites, the percentage of organic matter did not differ very much. This was supported by the ANOVA results (Table 3) in which the distribution of organic matter was not different between sampling sites ($p < 001$). However, between each sampling site, the highest percentage of organic matter was always found in the station closest to the river input or mangrove habitat. Mangroves are well known to be the source of organic matter deposition to the mangrove bottom habitat and the habitat surrounding [1,4,15-16].

Heavy Metal Distributions

The distribution of Pb, Hg and Cd concentrations are shown in Fig. 2. The spatial distribution of these heavy metals in the study areas showed a higher concentration of Pb compared to Hg and Cd. Concentrations of Pb, Hg and Cd were in the range of 0.033–0.134 ppm, 0.02–0.083 ppm, and 0.017–0.083 ppm, respectively.

Among the sampling sites, the average concentration of Pb was found to be the highest at Pantai Soge (0.116 ppm), followed by Pantai Watu Karung (0.095 ppm), Pantai Pancer (0.053 ppm), and Pantai Teleng Ria (0.043 ppm). The ANOVA results (Table 3) showed the distribution of Pb among sampling sites was significantly different ($p < 0.01$). The average concentration of Hg was also found to be the highest at Pantai Soge (0.082 ppm), followed by Pantai Pancer, Pantai Watu Karung, and Pantai Teleng Ria with values of 0.071 ppm, 0.050 ppm, and 0.039 ppm, respectively. However, a statistical test did not show a different distribution of Hg between sampling sites (Table 3). Unlike the distribution of Pb and Hg, the average concentration of Cd was found to be the lowest at Pantai Soge (0.025 ppm) and the highest at Pantai Pancer (0.068 ppm), followed by Pantai Watu Karung (0.049 ppm) and Pantai Teleng Ria (0.054 ppm). The distribution was significantly different between sampling sites ($p < 0.01$).

The distribution pattern of organic matter was rather similar to that for Pb and Hg but different from Cd (Fig. 3). While Pb, Hg and organic matter were higher in Pantai Watu Karung and decreasing toward Pantai Teleng Ria and started to increase again at Pantai Pancer, Cd concentration was the highest at Pantai Pancer.

The coastal areas, where the study was conducted, are highly influenced by human activities. Not only as recreational beaches with many visitors like Pantai Teleng Ria, Pantai Pancer and Pantai Soge, but also as the places for fishing activities like Pantai Watu Karung. These activities indicated that they are vulnerable to pollution. All of the sampling sites are close to a river that will bring pollutants from the urban areas.

There have been many studies on heavy metal content in sediment all around the world with diverse results depending on the heavy metal input, sediment characteristics and hydrographic conditions [6,12,17-19]. Compared to other studies, the heavy metal content found in this study was rather low [1,14,20-21]. Based on the study of the Yellow River [22], it was found that heavy metal contents of Pb and Cd were in the range of 21–34 ppm and 0.15–0.36 ppm, respectively. Another study conducted in Bangkalan, Madura found Cd content in the range of 6.7–9.6 ppm [23] and this is very high compared to this study which only found Cd in the range of 0.017–0.083 ppm. The results of this study were similar to the study in Pasuruan Port where the Cd concentration in sediments was in the range of 0.034–0.1 ppm [24].

Hydrographic data illustrated the actual data range for marine environment with high temperature, seawater range of salinity and slightly alkaline condition

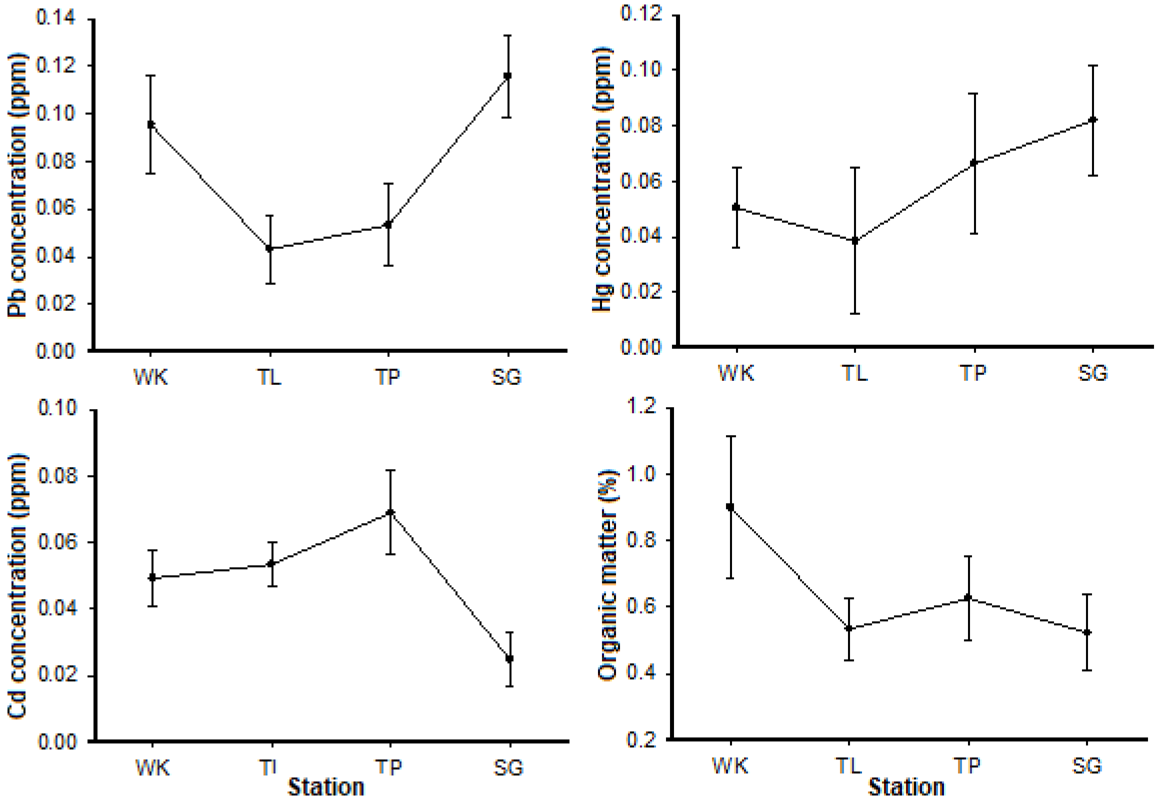

Fig 3. Heavy metal distribution and organic content in the sediment

of pH. The results of this study were in agreement with the study of [2] and [25] for temperature and pH. However, DO in this study was found to be the highest compared to both studies. Water quality parameters could change the bioavailability of metals in the pore waters and the release rate of metals from sediments [7], thus can modify heavy metals chemistry and toxicity on aquatic life [2]. For example, the study [25] found that metals especially Pb experienced flocculation better at lower salinity level and thus reduced its concentration in the environment due to changing of the metal to the particulate phase. This finding supported the result of this study that found higher Pb concentration at Pantai Watu Karung with the lowest salinity (Fig. 2 and Table 1). On the other hand, Pantai Teleng Ria with the highest salinity among study sites showed the lowest concentration of Pb in the sediment as the heavy metal concentration will tend to be higher in the water [26]. Dissolved oxygen can influence the release of heavy metals from sediment by affecting the rate of oxidation of organic compound [7]. It has been understood that organic matter plays an important role in metal binding [8] and the oxidation rate of organic matter is higher in the aerobic condition [7]. Low concentration of heavy metals in the surface sediments in this study might be related to high concentration of DO that easily released metals from the sediment to the overlying waters due to the binding of heavy metals with organic matter.

The study areas had a similar sediment composition with a high percentage of fine sand rather than gravel. All of the sampling sites were connected to rivers which gave high input of fine sediment. It has been known that fine sediment accumulated heavy metals more than bigger size sediment particles [1]. Pantai Watu Karung displayed the highest percentage of silt+clay due to the influence of the mangrove habitat and it can be one of the reasons why higher heavy metal concentrations was found in this beach compared to the other beaches. Furthermore, mangrove habitat contributed higher content of organic matter from its detritus to the sediment surface [16]. Finer size of sediment together with organic matter may accumulate higher concentrations of heavy metals [22].

Among the heavy metals, Pb was found to be the highest in most of the sampling sites except at Pantai Pancer. Pb is a type of heavy metal that is naturally present in the environment. It is the most concerning heavy metal pollutant in the environment. The input of Pb to the environment comes from many different sources such as from anthropogenic activities, soil and water-borne Pb, and also atmospheric inputs from industrial processes and combustion of fuels and waste materials [2] that make it present in a higher concentration than the other types of heavy metals. Higher concentration of Pb at Pantai Watu Karung and Pantai Soge compared to the concentration at Pantai Teleng Ria and Pantai Pancer clearly showed the effect of lower pH value especially at Pantai Watu Karung (pH = 7.92). Based on the experimental study on the effect of pH on metal release and sequestration [8], it was found that found a high concentration of Pb in the low-pH experiments. This metal showed little or no release from the sediment for the high- and mid-pH experiments. In addition, Pb concentration decreased in the environment with high disturbance of the sediment [6]. Pantai Teleng Ria and Pantai Pancer that experienced high disturbance of the sediments from the waves and currents were found to have a lower concentration of Pb compared to the other study sites.

The lower concentration of Cd compared to the other two types of heavy metals may be due to of its low mobility, as stated by [20] in their study on heavy metal distribution in marine sediments from the southwest coast of Spain. The input of cadmium to the environment comes from either land or air (atmosphere). Although there might be no exact point source of cadmium in the study areas (as shown by the lower concentration of cadmium compared to the other heavy metal concentrations), cadmium input might come from the pollution of cadmium from the air. Cadmium can be released into the atmosphere during heating in metal refining processes in association with copper [8]. Unlike the distribution of Pb that is related to lower pH, higher Cd concentrations might be related to the increased value of pH. Cd concentration was found to be the highest at Pantai Pancer with a slightly higher value of pH among the study sites. Cd concentration in the sediment increases with the increasing of pH due to precipitation of dissolved Cd [2]. A study [7] found similar results which showed lower release rate of metals from the sediment to the overlying water at higher pH than lower pH causing a higher concentration of Cd in the sediment than in the water.

Compared to the other sampling sites, Pantai Soge displayed a very high concentration of Pb and Hg. This may be due to the fact that this study site is close to fish and shrimp cultures. Stable and insoluble heavy metals are easy to dissolve in aerobic conditions such as in the culture systems [27] and this type of water is one of the sources of heavy metal deposition to the surrounding waters. On the other hand, the lower concentration of heavy metals at Pantai Teleng Ria and Pantai Pancer might be due to the dynamics of the coast (strong waves and currents) which result in mobilization and deposition of heavy metals to the deeper sediments.

In general, organic matter content in the study areas showed values less than 5% and according to the ecological quality status proposed by [28], it is classified as high-good quality. Moderate quality of organic matter in the environment is shown with percentage content between 5 to 10% and organic matter content more than 10% shows the poor-bad quality of the environment. The results of this study are almost similar to the study of [2,29] but much lower compared to the study of [14]. As stated by [2,14], the variability of organic matter in the environment is influenced by the river input, organic productivity due to the discharge of domestic waste and the rate of sedimentation.

Organic matter plays an important role in the accumulation process of metals in the sediment. Organic matter has the ability to absorb heavy metals and deposit it to the sediment [1]. Low concentration of heavy metals in this study might due to low percentage of organic matter (high-good quality environment).

The percentage of organic matter showed a decreasing value from Pantai Watu Karung to Pantai Soge. A higher percentage of organic matter at Pantai Watu Karung was also followed by a higher concentration of heavy metals compared to the results of the other study sites. This richness may result from the influence of the mangrove habitat at Pantai Watu Karung. The mangrove habitat contributed to the distribution of heavy metals due to its physicochemical properties, such as the ability to trap material from the water column and its high content of organic matter [16]. Therefore, the distribution of heavy metals in this study was related to the distribution of organic matter. The higher the organic content, the more metals accumulate in the sediments. Based on [12], it was stated that the distribution of heavy metals in mangrove sediment may be influenced by the quality and quantity of sedimentary organic matter and the cycling of Fe and S in the redox condition. Moreover, the sediment in Pantai Watu Karung that was mainly composed of silt+clay (Table 2) may be the reason for the higher heavy metal content, especially Pb and Cd in this area. Sediment grain size is very important in the accumulation process of heavy metals as decreasing grain size results in the increase of heavy metal content [30-31].

In general, there is inconsistency in the relationship between the distribution of the heavy metal and organic matter in this study (Fig. 3). However, the concentration of Hg can be seen to slightly follow the distribution of organic matter. In most of the stations, a higher concentration of Hg was found when the percentage organic matter was also high, especially at Pantai Teleng Ria and Pantai Pancer. Pantai Watu Karung and Pantai Soge showed the opposite pattern; a high percentage of organic matter but a low concentration of Hg.

CONCLUSION

This study showed the variability of the heavy metal distribution pattern on the southern coast of Pacitan. Pb was found to be higher than Hg and Cd. Pantai Soge exhibited higher concentrations of Pb and Hg, compared to the other stations, while Cd was found to be the highest at Pantai Pancer. These variabilities were observed due to the variability of the physicochemical properties of the sampling sites. Pb was found higher at higher salinity and pH. On the other hand, higher pH resulted in a higher concentration of Cd especially at Pantai Pancer. Compare to other studies, heavy metal concentrations in this study was rather low and it might be related to the low content of organic matter that can be considered as the good quality environment. However, attention to the heavy metal pollution in the study areas is needed because of its characteristics that are persistent and toxic.

ACKNOWLEDGEMENT

This research was funded by the Fisheries and Marine Science of Brawijaya University Research Grant (BOPTN 2015 No. SP DIPA-042.04.2.400072/2015). We also would like to thank Maria Deswita who helped us in the field sampling and data analysis.

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