ASSESSMENT OF Fe, Cu, Zn, Pb, Cd & Hg IN AMBON BAY SURFACE SEDIMENTS

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Received : January 2017 Accepted : June 2017

ABSTRACT

In this study, the concentration of Fe, Cu, Zn, Pb, Cd and Hg were determined in marine surface sediment to assess the impact of pollution sources around the Ambon Bay by application of Atomic Absorption Spectrometry (AAS). The metals concentrations found were in the following ranges (in mg kg^{-1} DW): 27,598 to 51,716 for Fe, 13.7 to 44.8 for Cu, 51.3 to 163 for Zn, 0.1 to 0.66 for Cd, 0.04 to 0.44 for Hg and 14.4 to 24.5 for Pb. The results indicated the strong contribution of heavy metals pollution from the urban waste, residential sources, farming industry, shipping activities and ship repair activity from dockyard around the Ambon Bay. This study provides the most updated information regarding heavy metals (Fe, Cu, Zn, Pb, Cd and Hg) concentrations in Ambon Bay marine sediment.

Keywords: *marine pollution, sediment, heavy metals, Ambon Bay*

INTRODUCTION

Ambon Bay is an important area for fishery and aquaculture that also grows rapidly in terms of population number and industrial activity. The increasing industrialization and human population concentrated in coastal regions have generally led to an increase discharges of pollutants which affect marine environment quality. The kind of waste that needs to be concerned is heavy metal. Heavy metals are causing concern in particular due to their bioaccumulation and toxic effects in most living beings. Heavy metal contamination due to anthropogenic activity has been reported in Ambon Bay waters in the past 20-30 years (Sellano *et al*., 2008; LIPI, 2008). The anthropogenic activities that could potentially contribute to heavy metal pollutants into Ambon Bay are the urban waste, farming, shipping activities and ship repair activity from dockyard around the Ambon Bay (LIPI, 2008).

Iron (Fe), copper (Cu), zinc (Zn), lead (Pb), cadmium (Cd) and mercury (Hg) are some of heavy metals that we should be aware of because their toxic effects to humans. At low levels, Cu, Fe and Zn are important for enzymatic activity, but toxic at higher concentrations in living organisms (Prashanth *et al*., 2015). Cd, Pb and Hg are known as the three most toxic metals in the environment without a known essential role in living organisms (Baker *et al*., 2003; Diaz *et al*., 2006, Ebrahimi and Taherianfard, 2010). In aquatic organisms, the toxic effects of heavy metals on aquatic invertebrates cause reduction of the developmental growth, increase of developmental anomalies, reduction of fishes survival even cause extinction of entire fishes population in very polluted waters (Khayatzadeh & Abbasi (2010).

Sediment is a good indicator for long-term studies related to monitoring of coastal waters (Zachariadis, 2001). Measurement of heavy metals in the sediment is widely used as indicator of pollution because the concentration of heavy metals in sediments is relatively stable and its role as a deposition media for diverse contaminants (Arifin *et al*., 2012, Tupan *et al*., 2014). Study of heavy metal in sediment have been conducted since early 1980s in Indonesia. The review of the study of heavy metals pollution in sediment that have been carried out in several coastal waters of Indonesia in 1983 to 2008 reported that population growth and industrial development along the coast have a significant role to pollution loads into coastal marine environment (Arifin, 2001, Arifin *et al*., 2012). However, most of study was conducted in western part of Indonesia and only a few in eastern part of Indonesia.

To our knowledge, there were only three sets of data available about heavy metal concentrations in Ambon Bay marine sediment. The first one was reported by Edward and Tarigan (1987), they found low concentrations (in mg kg^{-1} dry weight) of Cd (0.625), Cu (0.119), Pb (0.429), Zn (3.032) and Hg (0.0697) in sediment of Ambon Bay. The second one was published by Marasabessy *et al*. (2009) indicated an increase of Pb and Zn concentrations about 16 times higher than those observed in 1987. The latest study was conducted by Tupan (2014) in two sites of Ambon Bay, namely Poka and Lateri for Pb contamination in sediment. Sediment of the bay were sampled and measured for Pb in 2011 and found the value in Poka and Lateri ranged in: 5 to 15 mg kg^{-1} DW. The significant increase of pollutants input into the study locations give cause for concern. Whereas routine monitoring would be highly beneficial to ensure the appropriate environmental quality for human, as well as marine biota living in this water.

Considering the harmful consequence of heavy metal pollution and the rapid increase of population and industrial growth in Ambon, hence in the present study we reported the present status of Fe, Cu, Zn, Pb, Cd and Hg concentrations in surface sediments in Ambon Bay that suspected to be contaminated by anthropogenic activities.

MATERIALS AND METHODS

Description of the study area

Ambon Bay consists of an inner and outer area that is connected by a narrow and shallow threshold with an average depth of about 12 m (Fig. 1). The inner bay has an average depth of 30m with about 6 km^2 of area. The outer bay opens into Banda Sea, and has an area of about 100km^2 with an average depth over 100m. Ambon Bay is the commercial and industrial hub to Ambon city. As the main administrial and commercial city of Maluku province, Ambon has developped to become a small area with rapid growth in terms of population number and industrial activities. Ambon city has 359km^2, 0.57 percent of the total area of Maluku province but it has 24 percent of the total populations in Maluku Province. Population growth rate of Ambon City in 2000 to 2009 is 3.65, increased to 5.63 in 2009 to 2010, then in 3.75 in 2010 to 2015 (BPS, 2015). Metekohy *et al*. (2016) reported a rapid increase in demand for residential land in Ambon City in the period 2009 to 2015. In 2003, the amount of building land area in Ambon is 675 hectares, grew to 921 hectares in 2009 and 1322 hectares in 2015.

Seven coastal sampling sites were selected in the Ambon Bay by purposive sampling. They were located in the area with anthropogenic pressure and water circulation. Three sampling sites (Passo, Waiheru, and Poka) located in Ambon Inner Bay (AIB), one site (Ambang Galala) in threshold and the other three (Batu Merah, Waihaong, Tawiri) in Ambon Outer Bay (AOB), as illustrated in Fig. 1. Compared to AOB that opens to Banda Sea, water mass circulation in the AIB is relatively lower because of the tight and shallow threshold between the two sites of bay. Passo is a dense population area in the AIB, higher than Waiheru and Poka. Waiheru has a local vegetable farming that is suspected to contribute pollutant into the bay through process of fertilization. Batu Merah and Waihaong are an urban area with a high population. Around Waihaong there is a shipyard and the biggest port in Ambon. Tawiri village is far from Ambon city.

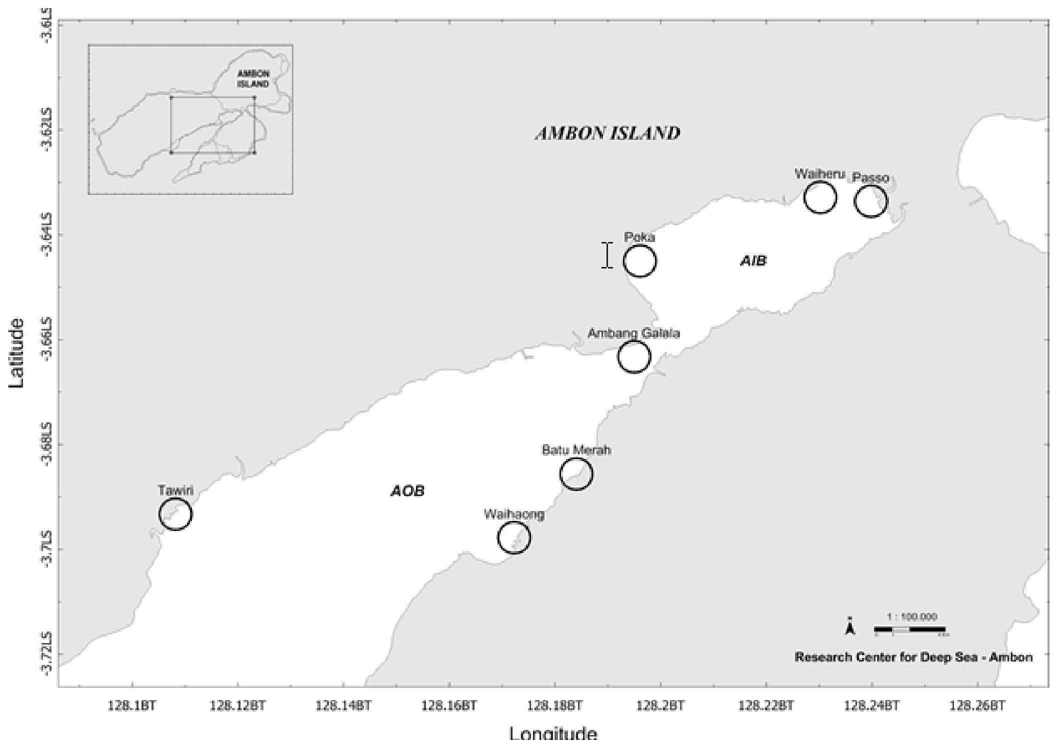

Figure 1. Map of the seven stations around Ambon Bay (Indonesia).

It has a small population, but it has a docking ship area.

Sampling and Storage Procedure

Marine surface sediments were collected at 7 stations from 25 to 28 August 2015 using a stainless steel grab sampler (0 to 10cm from the bottom surface) at 10 to 12m depth. About 1 kg sediments collected from each site were labeled and stored in the cool box before transfer into the laboratory. In each sites measurements of pH, salinity and temperature in water column were done by using pH meters and Conductivity Temperature Depth (CTD) Model ASTD-687. The Sampling location position was tracked by Global Positioning System (GPS)-Garmin, Model 76CSx.

Digestion Procedures

Samples were first dried to a constant weight at 60˚C and homogenized by using a mortar. Total concentrations of Fe, Cu, Zn, Pb and Cd were determined by Flame AAS SpectrAA-20 using procedures based on the standard methods described by USEPA 3050B (1996) at 95±5˚C, as follows: one gram subsample is digested with repeated additions of 15 ml nitric acid (HNO_3) and 4 ml hydrogen peroxide (H_2O_2). This was followed by an addition of 10 ml hydrochloric acid (HCl) then filtrated with Whatman no. 41 filter paper. The resulting solution was diluted in 100 ml with pure water and analyzed directly by FLAA.

Total concentration of Hg was determined by a cold-vapor atomic absorption method using procedures based on the standard methods described by USEPA 7471B (1998) at 95±3˚C. Briefly, a well homogenized sediment sample (0.5-0.6g) was digested by reagent water, aqua regia and potassium permanganate. This was followed by an addition of sodium chloride-hydroxylamine sulfate to reduce the excess permanganate. The resulting solution was subsequently diluted using reagent water and stannous sulfate prior to analyze them using FAAS with vapor generated accessories. All analysis were carried out with blanks. All glassware were cleaned by refluxing with nitric acid and thoroughly rinsed with H_2O before used. The reported results are expressed as $mg.kg^{-1}$ dry weight.

Quality Assurance

The validity of the analytical methods for all metals was monitored by the analysis of Certified Reference Material from National Research

Table 1. Measured and certified values of Fe, Cu, Zn, Pb, Cd and Hg concentrations ($mg.kg^{-1}$) using standard sediment reference Certified Reference Material from National Research Council of Canada (CRM NRCC) PACS-2.

PACS 2 (n = 3)	Certificate Value	Measured Value
Fe	40,900 ± 600	42,368 ±256
Cu	310 ± 12	306 ± 8.6
Zn	364 ± 23	381± 310
Pb	183 ± 8	190 ± 9.8
Cd	2.11 ± 0.15	2.23 ± 0.25
Hg	3.04 ± 0.2	3.08± 0.41

Council of Canada (CRM NRCC) PACS-2. Values were consistently within the certified range (Table 1).

Analysis Method

Due to the lack of specific regulation about heavy metal concentrations in coastal sediments in Indonesia, all of the results were compared to the existing standards as determined by Environment Canada Threshold Effect Limits (ECTEL), NOAA (National Oceanic and Atmospheric Administration), and Australian and New Zealand Environment and Conservation Council (ANZECC) guidelines (Burton, 2002). We also compared the results with prior existing reports from Ambon Bay and other bays. No guidelines given for Fe concentration, and for that reason, we compared only with the existing reports.

RESULTS

Temperature range in the AIB was 26.05 to 26.12°C, while in the AOB was 25.85 to 25.91°C (Table 2). Salinity range in the AIB was 33.89 – 33.91 psu and 33.76 – 34.13 psu in the AOB. pH range in the AIB was 7.78 – 8.05 and 7.46 -7.89 in the AOB. The range of temperature, salinity and pH of Ambon Bay are in accordance with the national standard of marine water quality (Decree of the Indonesian Minister of environment no. 51, 2004).

Fe, Cu, Zn, Pb, Cd and Hg were found in all sites (Figure 2). The concentrations of heavy metals in all sites ranged from (DW, in mg.kg-1): 0.1 to 0.66 for Cd, 13.7 to 44.8mg.kg^{-1} (dry weight) for Cu, 27,598 to 51,716mg.kg^{-1} (dry weight) for Fe, 14.4 to 24.5mg.kg^{-1} (dry weight) for Pb, 51.3 to 163mg.kg^{-1} (dry weight) for Zn and 0.04 to 0.44mg.kg^{-1} (dry weight) for Hg. The highest concentration of Cd, Cu, Fe, Pb were found in Tawiri. Meanwhile, the highest concentration of Zn and Hg were detected in Batu Merah.

Based on the quality standards for marine sediment under Environment Canada Threshold Effect Limits (EC TEL), NOAA and Australian and New Zealand Environment and Conservation Council (ANZECC) level of some metals found below the guidelines. The level of metals has exceed the rules were Cu (Passo, Galala, Batu Merah, Waihaong and Tawiri), Zn (Batu Merah) and Hg (Batu Merah). There was no guidelines found for Fe (Burton, 2002).

DICUSSION

Temperature, Salinity and pH of Ambon Bay

Temperature in the AIB is higher than in the AOB. The higher temperatures can increase particulates solubility in the water, otherwise at low temperature the heavy metals have a tendency to settle in the bottom (Maramis and Hastuti, 2009). The pH found in all sites were in range of 7.46 to 8.05 (slightly alkaline) and this may hinder metals dissolution in water and tend to settle at the bottom. Heavy metals free ions are released into the water column at low pH and tend to decrease when the pH increases (Hill, 2010; Maramis and Hastuti, 2009). The salinity in the AOB is lower than in the AIB. The dissolved of some metals like Cd and Zn are influence by salinity. The dissolved Cd and Zn increases at

Table 2. Results of temperature, salinity and pH of Ambon Bay

No.	Longitude	Latitude	Location	Temperature (°C)	Salinity (psu)	pH
1	128.2384	-3.67110	Passo	26.12	33.89	7.78
2	128.2317	-3.63487	Waiheru	26.05	33.89	7.99
3	128.1971	-3.64527	Poka	26.10	33.91	8.05
4	128.1950	-3.66328	Ambang Galala	25.91	34.09	7.46
5	128.1823	-3.68473	Batu Merah	25.87	34.13	7.86
6	128.1692	-3.69776	Waihaong	25.91	34.02	7.85
7	128.1111	-3.69483	Tawiri	25.85	33.76	7.89

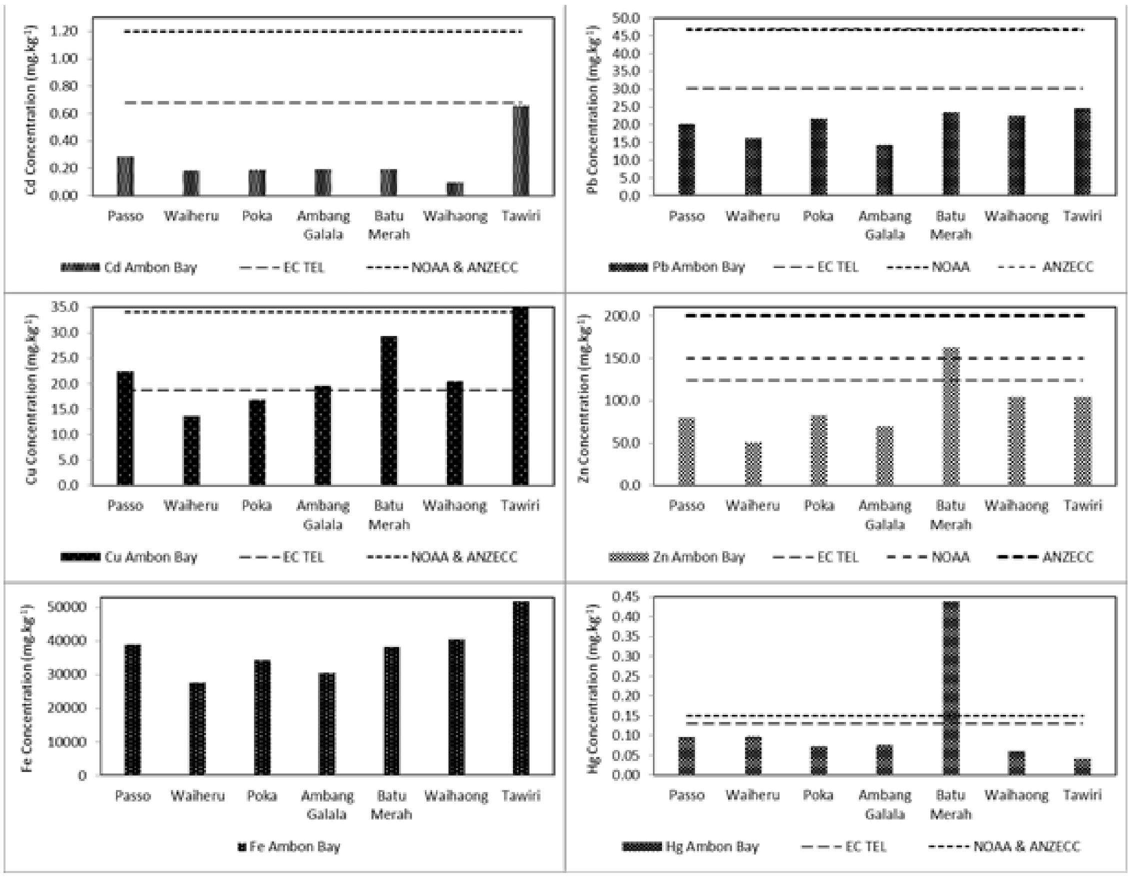

Figure 2. Fe, Cu, Zn, Pb, Cd and Hg concentration of Ambon Bay (mg.kg^{-1} DW).

low salinity (Gerringa *et al*., 2001). However, the variation of temperature, pH and salinity in this study did not have a significant role to the level of concentration in all sites generally. This was an indication that concentration metals were more influence by antropoghenic factor. Moreover the higher temperature in the AIB might be influenced by the limited mass circulation water in the AIB caused by the narrow and shallow threshold between AIB and AOB.

Fe, Cu, Zn, Pb, Cd and Hg Concentrations

The metal concentrations found in marine sediment of Ambon Bay can be related to discharge from urban waste and industrial activities around the bay. The highest level of Cd was detected in Tawiri site. This can be explained by docking activities at this site such as stripping and painting of ships. Cd is widely used in the binding of pigment in paint manufacturing (Hutagalung, 1984 and Yu, 2005). On the other hand, the high concentration of Cd in Tawiri may be affected by submarine volcano in the Banda Sea that dragged to the coastal area through the upwelling process. The influence of submarine volcano on high heavy metals input in to the marine environment was reported by Mrajitha (2010), following observation carried out on Adonara Island (East Florest, Indonesia). However, this hypothesis should be confirmed by further study.

Sediment of Passo site contained 0.29mg.kg^{-1} of Cd. Marasabessy *et al*. (2009) reported that the Cd concentration in Passo was undetectable or below the detection limit of AAS (0.002mg.kg^{-1}). This rapidly increasing might be happening due to the growth of residential waste and runoff from commercial land areas such as shopping centers at this site (Hill, 2010). In Waiheru, the concentration of Cd has increased from 0.09 to 0.19mg.kg^{-1} in seven years. This may have derived from agricultural activities due to the usage of Cd containing fertilizers and fungicides (Yu, 2005).

Pb concentrations found in the Bay of Ambon ranged from 14.4 to 24.5 (DW, in mg.kg^{-1}). High concentration of Pb was detected in Batu Merah, Waihaong and Tawiri sites. In the 20th century, lead was used extensively as an additive in gasoline (WHO, 1977) and as a compound in red and yellow in paints manufacturing as well (Sudarmaji *et al*., 2006). Fuel oil has additives such as tetra ethyl lead and tetra methyl lead to improve the fuel quality (Rochyatun *et al*., 2006; Sudarmaji *et al*., 2006). Nowdays, the using of lead has been eliminated from the petrol supplies and paint manufactures of the majority of countries. However, in this study we found the higher Pb were found in the harbor activities and docking ship area. Tawiri site is a docking area. Batu Merah and Waihaong are close to Yos Sudarso Harbor and Wayame docking ships area. In comparison to the previous study conducted by Marasabessy *et al*. (2009) in Ambon Bay, even Pb concentration in this present study was higher in average. The Pb measured in Tawiri was 12 times higher than this previous data. This was an indication that lead may still be released into the environment by peeling the paint of old ships in docking area and contamination from the urban sewage run-off. Batu Merah and Waihaong are urban areas with a high population in Ambon.

The marine surface sediment of Ambon Bay contained 0.09 to 0.44 (DW, in mg.kg^{-1}) of Hg. The work of Marasabessy *et al* (2008), reported that no Hg was detected (the value was lower than the detection limit of AAS, 0.002mg.kg^{-1}) in all sites of Ambon Bay. The presence of Hg contamination possibly caused by natural process such as volcanic activity, however normally Hg contamination in the environment majority comes from the anthropogenic sources. The studies reported that Hg might generally exist in the urban sewage such as gasoline and oil combustion (Hg content of coal is about 1 ppm), disposal electrical equipment (batteries, lamps) and compounds as a seed dressing in agriculture (Yu, 2005).

In this study we found Cu, Fe and Zn (DW, mg.kg^{-1}) varied between 13.7-44.8 for Cu, 27,598 to 51,716 for Fe and 51.3-163 for Zn. At low concentration, Cu, Fe and Zn are essential metals for normal growth, disease resistance, production and reproduction (Prashanth *et al*., 2015). According to the quality standards for marine sediment established by EC TEL standards, the Cu and Zn levels in some sites have exceeded the limits. Cu has exceeded the limit in Passo, Ambang Galala, Batu Merah, Waihaong and Tawiri. Zn in Batu Merah has exceeded the limit. No guideline given for Fe concentration in

Table 3. Guidelines for metals in marine sediment

Metals	EC TEL[1]	NOAA[2]	ANZECC[3]	Ambon Bay[4]
Fe	-	-	-	27598-51716
Cu	18.7	34	34	13.7-44.8
Zn	124	150	200	51.3-163
Pb	30.2	46.7	47	14.4-24.5
Cd	0.68	1.2	1.2	0.1-0.66
Hg	0.13	0.15	0.15	0.04-0.44

[1]EC TEL: Environment Canada Threshold Effect Limits (Burton, 2002); [2]NOAA: National Oceanic and Atmospheric Administration (Burton, 2002); [3]ANZECC: Australian and New Zealand Environment and Conservation Council (Burton, 2002); [4] This study.

sediment. Docking activities, urban sewage and various industries in and around Ambon Bay that possibly the source of high Cu and Zn in Ambon Bay. As comparison, Yulianto *et al*. (2006) found Cu ranged 19.88-57.70mg.kg^{-1} in industrial areas of Central Java.

Comparison were made to estimate the environmental consequences of the studied metals obtained in the present study to the marine sediment guidelines and some previous studies in Indonesia and overseas. Results showed that all of Cd and Pb detected in Ambon Bay are below the EC TEL, NOAA and ANZECC values. The level of Cu in all sites, except Waiheru and Poka has exceeded the limits of EC TEL, even the level of Cu in Tawiri was above the NOAA and ANZECC values. The Hg and Zn measured in Ambon Bay were clearly lower than guidelines, except Hg in Batu Merah site. In this study, we found 27527 to 51716 mg.kg^{-1} (dry weight) of Fe. So far, there is no quality standard of Fe in marine sediment established by the Indonesian Ministry of Environment, neither the previous study in Ambon Bay. Compared to other studies, Fe found in marine sediment of Ambon Bay are far higher than Fe found in the harbors area in South Africa, 10095.0 to 14603.0 mg.kg-1 (Fatoki and Mathabatta, 2001) but less than Fe found in Jakarta Bay (Suyarso & Lestari,2012

Heavy metals level in sediment of other sites of Indonesia and other world areas is presented in Table 4. Fe level in marine sediment of Ambon

Table 4. Comparison of Ambon Bay heavy metals concentration with other marine sediments concentration from different locations (in mg.kg^{-1}, dry weight).

Location	Fe	Cu	Zn	Pb	Cd	Hg
Ambon Bay[a]	*27,598–51,716*	*13.7-44.8*	*51.3-163*	*14.4-24.5*	*0.1 – 0.66*	*0.04–0 .44*
Jakarta Bay (Indonesia) [b]	50,282-65,093	*13.5-35.4*	69.7-244	10.1-27.7	0.069-0.639	0.04-0.154
Jakarta Bay (Indonesia) [c]	-	*15 - 169.5*	95.8–333.3	14- 58.1	0.012-0.75	-
Gresik Waters[d]	-	23.7 - 234	77.3 - 405	1.74 – 12.7	0.08 – 3.05	0.04 – 0.33
Mahakam Delta[e]	-	-	-	75.27 – 903.46	0.66 – 1.82	0.0005
Dumai Sea western waters[f]	-	-	-	5.16 – 12.69	0.02 – 0.1	-
Klabat Bay (Indonesia)[g]		0.067–6.35	0.74–38.83	1.07 – 32.54	0.007-0.08	-
Manila Bay (Philipines)[h]	-	-	-	7.3 - 19	-	-
Gulluk Bay (Turkey)[i]	-	25.2	80.8	20.0	0.56	-
Soline Bay (Croatia)[j]	-	21.1– 51.9	17.0–65.4	29.9 – 67.3	0.086–0.12	-

[a]This study; [b](Suyarso & Lestari, 2012); [c](Permanawati *et al.*, 2013); [d](Lestari & Budiyanto , 2013); [e](Salahuddinet al., 2012); [f](Syahminan *et al*., 2015); [g](Puspitasari and Hindarti, 2009); [h](Hosono *et al*., 2010); [i](Dalman *et al*., 2006); [j](Kljakovic-Gaspic *et al*., 2008).

Bay was lower than Jakarta Bay. The levels of Cu were lower than other bays except Klabat Bay and Gulluk Bay. Zn measured in Ambon Bay was lower than Jakarta Bay and Gresik Waters. Pb was detected above the Gresik Waters, Dumai Sea western waters, Manila Bay and Gulluk Bay. Cd level in Ambon Bay was higher than Klabat Bay, Dumai Sea western waters, Gulluk Bay and Soline Bay. Hg found in this study was higher than Hg in Jakarta Bay, Gresik Waters and Mahakam Delta (Suyarso & Lestari, 2012).

Generally, Zn and Cd in Ambon Bay are higher than others bay. The high Cd and Zn level in the sediment would pose hazards to human health. Contaminants in the sediment can be taken up by benthic organisms. When larger animals feed on these contaminated organisms, include human, the toxins are taken into their bodies, moving up the food chain with increasing concentrations (Khayatzadeh & Abbasi, 2010).

CONCLUSION

Marine surface sediment of Ambon Bay has been contaminated by heavy metals Fe, Cu, Zn, Pb, Cd and Hg. It indicates that growing population and industries in Ambon have a significant impact to the quality of the Ambon Bay sediment. There is urgently required for establishing a guideline for heavy metals pollutant in coastal sediments in Indonesia, considering that there might be a difference of heavy metal accumulation in Indonesian waters compared to the other regions in the world. This is due to geographical location and environmental parameter that could influence the input, presence and persistence of heavy metal in marine surface sediments in this region.

ACKNOWLEDGMENTS

The authors are thankful to Willem M. Tatipatta and Sudin A. Malik for their assistance in field sampling, and Mr. Rozak for his assistance in heavy metals analyses. This work was supported by research project Monitoring of Ambon Bay – Indonesian Institute of Sciences.

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