



## Spatial distribution of some heavy metals in the sediments of Tangerang coastal waters, Banten Province, Indonesia

Sigid Hariyadi, Akrom Muflih, Ali Mashar, Bambang Widigdo, Yusli Wardiatno

Department of Aquatic Resources Management, Faculty of Fisheries and Marine Sciences, Bogor Agricultural University, Jalan Raya Darmaga, Kampus IPB, Bogor 16680, West Java, Indonesia. Corresponding author: S. Hariyadi, sigidhariyadi@gmail.com

**Abstract.** The pollution of the Tangerang coastal waters, Province of Banten is caused by industrial, household, and agricultural activities. The heavy metals such as cadmium (Cd), copper (Cu), mercury (Hg), and lead (Pb) were detected in the sediment and their spatial distribution was mapped. This study was conducted in Tangerang coastal waters, a part of Banten Province, Indonesia. The sediment samples were collected in April and August 2013. Some water parameters were also observed. The Spearman rank correlation test was taken to determine the correlations between heavy metal concentration and particle size, and between heavy metal concentration and total organic carbon. The distribution of heavy metals in the sediments was spatially different in each area. The concentration of Hg in general exceeded the threshold. The Spearman rank correlation showed no relationship between the concentration of heavy metals and the size of the particles or the total organic carbon ( $p > 0.05$ ).

**Key Words:** coastal degradation, heavy metal pollution, industrial waste, particle size, western Java.

**Introduction.** Tangerang District is one of the areas in Indonesia, which is still being developed in an industrial point of view. The development involves many sectors, including industry, housing, and agriculture. According to the Central Bureau of Statistics (2013), there were 119 large and medium-sized industries in Tangerang District in 2012. These developments are consequently followed by the increased amount of waste products, both harmful and harmless in nature. In addition, household and agricultural activities contribute waste as well to the coastal waters. The waste materials produced are released to the environment through the air, soil, and water, but most of it is dumped in the water. This is because water is considered to be able to restore its condition fairly quickly via flushing, mixing, and absorption by aquatic biota. One of the hazardous wastes in Tangerang coastal waters are heavy metals.

Several heavy metals are used in industries as raw material, reagents, catalysts, and solvents. Industrial activities which use heavy metals are the textile, metal galvanizing, paint or color ink, printing, and agro-chemical industries (Rochyatun et al 2005). Oil drilling, gold and coal mining, electric plants, pesticide and ceramic industries, and metal smelting involve heavy metals in their production process (Suhendrayatna 2001). The waste materials from these activities usually enter the waters through rivers that ended into Tangerang District's coastal areas. Heavy metals are difficult to degrade and they are accumulated in organisms and sediments. Cd (cadmium), Pb (lead), and Hg (mercury) are non-essential metals (they are not needed by biota), whereas Cu (copper) is an essential metal (important to biota in trace amounts) (Sanusi 2006; Eisler 2007). These metals are poisonous to aquatic biota, especially if the concentration in the water exceeds the threshold (Connell & Miller 1995; Arifin & Fadhlina 2009; Pinet 2009; Olubunmi & Olorunsola 2010). Heavy metals' poisonous and hazardous nature is demonstrated by its physical and chemical characteristics. Its physical character which is difficult to degrade makes this kind of pollutant accumulate in aquatic environment, while



**Study steps.** This study was divided into three steps: (1) The determination of the study locations: the sampling locations were determined using GPS (Global Positioning System). The sampling locations consist of 18 composite stations (sampling lines) and 52 sub-stations or sampling points (Figure 1). The determination of the stations was based on the representation of the areas in the study of the distribution of heavy metal in sediments; (2) Sediment sample-taking procedure: the sediment samples were taken using the Van Veen grab. The samples taken from each location were put in a plastic flip bag and labeled; and (3) Sediment sample handling and analysis: the samples collected from the 52 sub-stations were made into proportionate composites based on the areas (sampling line) become 18 station samples. Then the sediment samples were taken to the laboratory to be analyzed for heavy metal concentration, organic materials, and textures.

**Heavy metal concentration analysis.** Heavy metals in the sediment were analyzed using inductively coupled plasma-optical emission spectroscopy (ICP-OES), method which is used to simultaneously analyze several chemical elements. The metal analysis was also done using an atomic absorption spectrophotometer (AAS) following American standard analysis procedures (APHA 2012).

**Sediment grain size and organic material analyses.** The particle size analysis was done through fractioning using 10 fractions. The sediment samples were dried (Sulaeman et al 2005), then each filter size sample was weighed and the percentage of weight was calculated. After that, the median was calculated using the interpolation technique and classified based on Wentworth's scale. The organic materials were analyzed for the total organic carbon (TOC) based on the Walkley & Black value (C) in percentage (Sulaeman et al 2005).

### **Data analysis**

**Comparative analysis.** The heavy metal concentration data was compared with the thresholds from various countries and international institutions, that is, the Australian and New Zealand Environment and Conservation Council-Interim Sediment Quality Guidelines, Hong Kong-Interim Sediment Quality Value-*low*, National Oceanic and Atmospheric Administration (Burton 2002), United States Environmental Protection Agency-effects range low (EPA 2007), and Canadian Council of Ministers of the Environment-threshold effect level (ECMDEPO 2007). The thresholds are defined as values which would have a negative effect on biota if the concentration exceeds them.

**Spatial distribution of heavy metal analysis.** The heavy metal data and the coordinates were processed spatially based on the Geographic Information System approach using the relevant software. The spatial analysis of the distribution of heavy metal used the interpolation method. Interpolation is a value estimation process in unmeasured areas to form a value distribution for the whole area. The technique employed in the interpolation method was Inverse Distance Weight (IDW). IDW is a technique which shows interpolation results that are similar to the sample data found closer than the ones later found. The weight would change linearly according to its distance to the sample data (Childs 2004).

**Correlation analysis.** The Spearman Rank Correlation analysis (Fowler & Cohen 1990) was used to reveal the relationship between heavy metal concentration and the size of particles and also with organic materials in the sediments. The correlation analysis measured the strength of the relationship between two variables through a number called a coefficient.

**Results and Discussion.** The results of the heavy metal analysis showed that some have exceeded the threshold, while others have both partially exceeded and not exceeded the threshold (Table 1). The concentration of Hg exceeded the thresholds of all

the threshold references used in all the stations in both observations (April and August). The concentration of Cu exceeded the thresholds (NOAA, USEPA-ERL, and CCME-TEL) in Dadap and Kronjo areas in August.

The concentrations of Pb and Cd have not exceeded the thresholds. Thresholds are the upper limit of the concentration of heavy metals that would have negative effects at low levels (slight biological effects) on biota. The condition of the concentration of the four heavy metals indicated that there was Hg pollution from various industries. In addition, accumulation of heavy metals can be caused by three factors, the high concentration in industrial and city waste, the effect of sea currents in the waste conduits, and flux from soil (Kalloul et al 2012). The precipitation of metals in coastal areas is controlled by natural processes such as diffusion by the currents on the coast, geochemical, biogenic, and inorganic processes (Balachandran et al 2006). Most of the metals formed naturally in sea water are supplied by the rivers, rock disintegration, and sediments from the land. Heavy metals are important in controlling various biological processes (photosynthesis and cellular metabolism). When the concentration of metals in a certain area exceeds the threshold, the area is at risk of endangering its ecology (the environment of benthic and trophic-level organisms through transfer in the food chain), requiring special attention. High heavy metal concentration is very poisonous (lethal) to aquatic biota. Metals from anthropogenic sources are usually waste materials from industrial processes.

Table 1  
Metal concentrations in sediments of Tangerang coastal waters, Banten Province – Indonesia and the quality standards

No.	Comparison	Code	Type of heavy metal			
			Cu	Pb	Cd	Hg
1	The quality standards (ppm)	1 <sup>a</sup>	65	50	1.5	0.15
		2 <sup>b</sup>	65	75	1.5	0.28
		3 <sup>c</sup>	34	46.7	1.2	0.15
		4 <sup>d</sup>	36	35	0.6	0.17
<i>Observations in April</i>						
2	Average concentration ± SD (ppm)	K	4.73±0.89	< 0.20±0.00	< 0.10±0.00	14.10±2.60
		M	6.30±1.41	11.50±0.00	< 0.10±0.00	8.55±1.20
		R	4.94±2.01	1.95±2.47	< 0.10±0.00	6.12±0.54
		T	9.44±1.29	< 0.20±0.00	< 0.10±0.00	4.72±0.83
		D	16.97±10.15	< 0.20±0.00	< 0.10±0.00	19.57±0.85
<i>Observations in August</i>						
3	Average concentration ± SD (ppm)	K	31.00±18.29	11.23±5.25	< 0.10±0.00	0.94±0.32
		M	26.95±5.93	14.32±0.53	< 0.10±0.00	1.57±0.54
		R	20.13±8.60	11.88±6.80	< 0.10±0.00	3.35±3.25
		T	23.67±2.39	12.81±2.98	< 0.10±0.00	4.46±0.99
		D	45.73±16.42	13.93±1.32	< 0.10±0.00	1.02±0.40

K: Kronjo, M: Mauk, R: Rawakidang, T: Tanjung pasir, D: Dadap; <sup>a</sup>ANZECC ISQG-low, Australian and New Zealand Environment and Conservation Council, Interim Sediment Quality Guidelines (Burton 2002); <sup>b</sup>Hong Kong ISQV-low, Interim Sediment Quality Value (Burton 2002); <sup>c</sup>NOAA, National Oceanic and Atmospheric Administration (Burton 2002) and USEPA-ERL, United State Environmental Protection Agency- Effects Range Low (EPA 2007); <sup>d</sup>CCME-TEL, Canadian Council of Ministers of the Environment-Threshold Effect Level (ECMDEPQ 2007).

**Water quality.** Water quality reflects the condition of the waters and determines the processes that will be undergone by the metals suspended in the sediments. The water quality parameters which affect the form of metals in waters are salinity, pH and dissolved metals. The observation results showed a pH of approximately 8, a salinity of 20-30 ppt, dissolved Cu < 0.002-0.011 ppm, dissolved Pb < 0.002-0.014 ppm, and Cd < 0.001-0.014 ppm. Olubunmi & Olorunsola (2010) stated that heavy metal in the form of crystal structures in sediments is not affected by the dilution effect (the lowering of concentration) caused by rain. The observation results showed that the pH of the waters was approximately 8; therefore, biochemical processes tend to be stable. Balintova et al

(2012) stated that the variance of pH affects the sediment partition of copper in polluted waters. The salinity of 20-30 ppt portrays a vast observation area from estuary to the outer coastal area. The decline of pH through decreased salinity could change the kinetics of elements and support desorption of metals from the sediment to a dissolved phase (Sylaios et al 2012).

***Spatial distribution of the heavy metals.*** In general, the distribution of heavy metals is affected by the position of the land, the amount of input, the waters' dynamics and bio-geochemistry. According to Laing et al (2007), the availability of metals is affected by the influence of the dynamic system (the tide flow), the hydraulic system, organic materials and salinity to metal mobility and bioavailability. Geographically, Tangerang District has a number of rivers. There are two in Kronjo, that is, Sipanjang and Cipasilian, and in Mauk there are three, that is, Cimandiri, Cileuleus, and Cimaug. In Rawakidang there are two rivers, Cirarab and Anak Cisadane, in Tanjung Pasir there is only one main river, Cisadane, which is divided into two as it flows to the estuary. In Dadap there are two rivers, Dadap River and Kamal River (Jakarta) and they are located facing the Muara Angke Port. These rivers are the source of input from land. The river which is potentially the largest contributor of pollutants is Cisadane. This river passes through Bogor and Tangerang City. The status of the river is highly polluted (Saputra 2009). Accumulations of heavy metal such as Pb, Cd, and Cu were found in the sediments of the coast before the rainy season, because of the activities on land, the huge input from untreated industrial waste (Ravichandran & Manickam 2012).

***Cadmium (Cd).*** Cd is found in very small amounts (trace amounts) and is not water soluble. The cadmium content level in the Earth's crust is approximately  $0.2 \text{ mg kg}^{-1}$ . Natural sources of Cd are greenockite (CdS), hawleyite, sphalerite, and otavite (Moore 1991). Cd is commonly used in metallurgy, metal galvanizing, pigment, battery, electronics, lubricant, glass, ceramic, textile, and plastic industries. The observation results for April and August in a whole showed that the concentration of Cd is below the equipment's sensitivity level or undetected. The concentration of Cd was less than 0.10 ppm in the five observed locations.

***Copper (Cu).*** Cu is a heavy metal which is naturally found in waters and is an essential element for animals and plants. In plants and algae, Cu is a component of plastocyanin which plays a role in the transportation of electrons in photosynthesis (Boney 1989). Cu compounds which are not easily dissolved in water, such as copper carbonate, copper hydroxide, and copper sulphate will undergo precipitation in waters with high alkalinity. The observation results in April as a whole is 1.80-28.47 ppm (Figure 2). The highest concentration of Cu was found in Dadap at a value of 25.52-28.47 ppm. The distribution of Cu is affected by currents (tides), the amount of input from land especially through river, and the utilization by aquatic biota. The observation results in August as a whole was 7.43-62.37 ppm (Figure 3). The concentration of Cu in Dadap and Kronjo was very high, 56.27-62.37 ppm. The metal came from mining activities, industrial zones, and precipitation from the atmosphere. The high concentration of Cu in Dadap waters that is also part of the Jakarta Bay, suggested that there were high input of Cu from activities in the area and on the upper land.

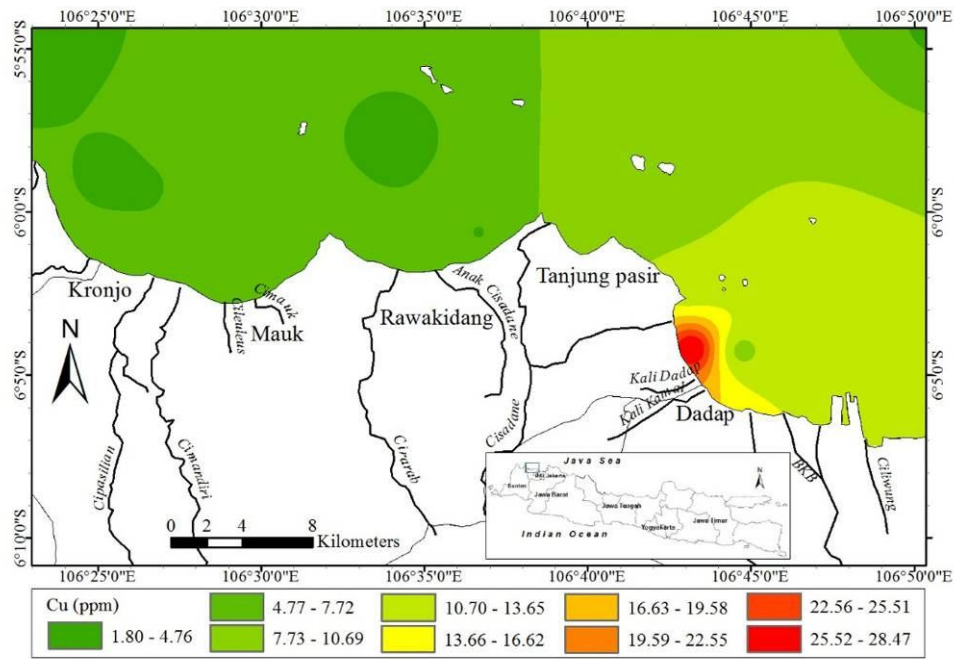


Figure 2. Distribution of copper (Cu) in the sediments of Tangerang coastal waters, Province Banten - Indonesia in April 2013.

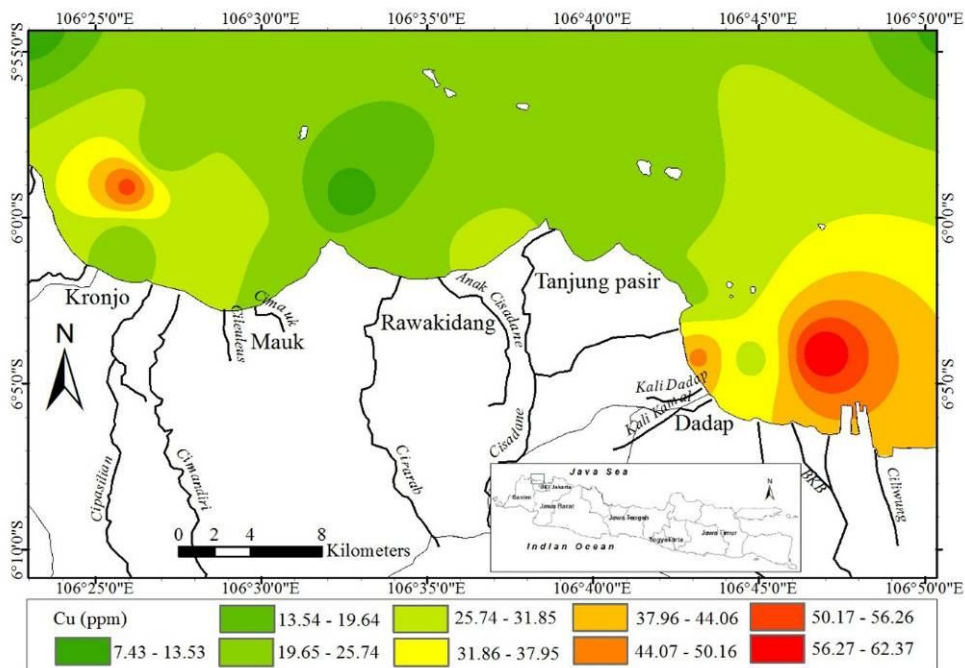


Figure 3. Distribution of copper (Cu) in the sediments of Tangerang coastal waters, Province Banten - Indonesia in August 2013.

*Mercury (Hg)*. Hg is a trace element in the Earth's crust, the availability is 0.08 ppm. Hg is very toxic because of its high volatility, which enables it to quickly spread from natural and anthropogenic sources (Abi-Ghanem et al 2011). Hg is a liquid metal at normal temperature. The observation results in April showed that the concentration of Hg as a whole was 2.35-20.19 ppm (Figure 4). The concentration of Hg in Kronjo and Dadap was very high, 18.22-20.19 ppm. The observations in August as a whole showed a concentration of 0.34-9.08 ppm (Figure 5). The highest concentration of Hg was found in Rawakidang area at 8.12-9.08 ppm.

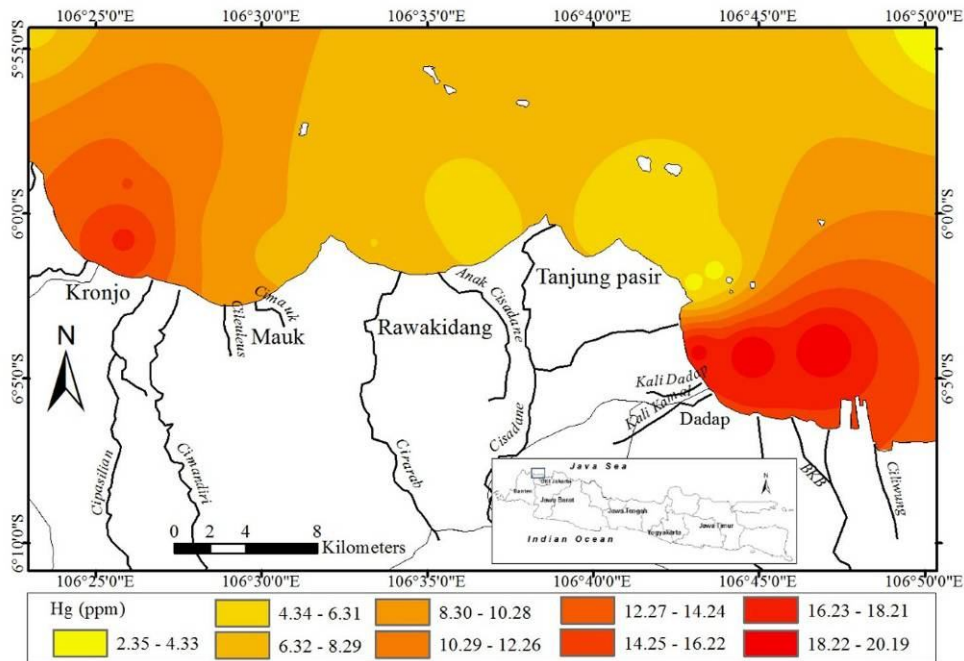


Figure 4. Distribution of mercury (Hg) in the sediments of Tangerang coastal waters, Province Banten - Indonesia in April 2013.

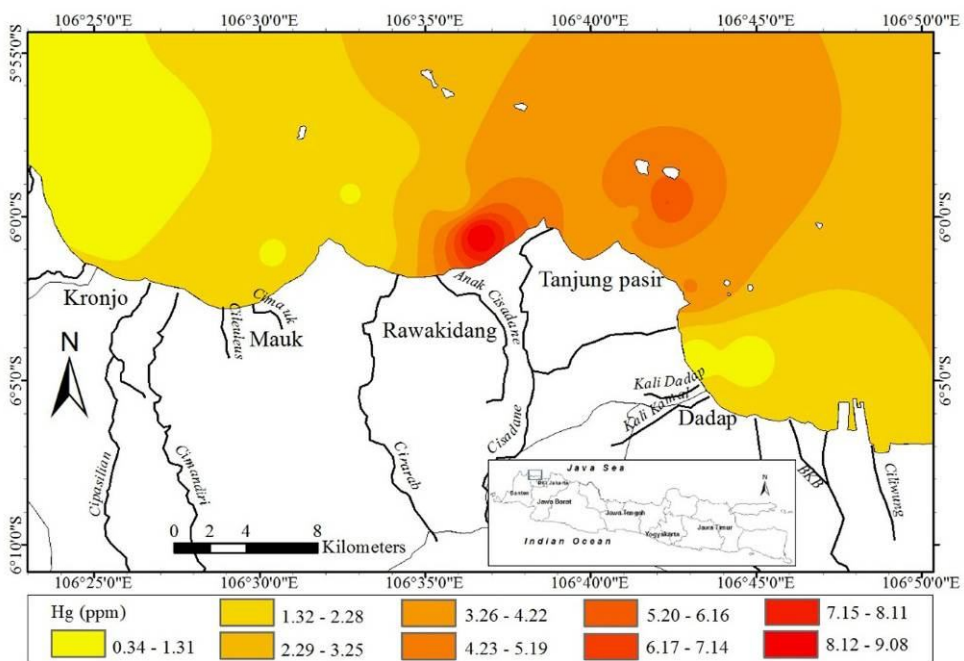


Figure 5. Distribution of mercury (Hg) in the sediments of Tangerang coastal waters, Province Banten - Indonesia in August 2013.

High concentrations of Hg can be directly or indirectly affected by land. On the Tangerang coast, around Kronjo and Mauk, there is a Steam-Generated Power Plant which uses coal as fuel and there is a large number of coal-trucks scattered around the area. The burning of coal releases carbon dioxide, mercury, smog, sulfurous compounds, and hydro carbon (Pirrone & Mason 2009). This indicates that the main source of Hg in the waters of Tangerang is coal burning. Hg is released to the atmosphere from a number of sources, such as, fossil fuel, power plants that use coal, smelting plants (metal and non-metal), chemical production, ore processing, waste-processing plants, and cement plants (Pirrone & Mason 2009).

*Lead (Pb)*. The distribution of Pb concentration precipitated in the sediments of Tangerang District's coast showed some differences. The observations in April showed a concentration of 0.20-11.49 ppm (Figure 6). The concentration of Pb was measured in the Rawakidang area at 0.20-3.70 ppm and in Mauk at 10.22-11.49 ppm, while the other stations showed concentrations below the sensitivity level of the equipment used (< 0.20 ppm). However, the concentration of Pb in August was 0.21-17.20 ppm (Figure 7). The highest concentration of Pb in August was found in all areas observed at 15.33-17.21 ppm except in parts of Kronjo and Rawakidang. Pb is the most soluble metal with high bioavailability in low pH, low organic material, and low concentration in the sediment. The Pb mobility in sediments has a positive correlation with the increase in temperature, the decrease in pH, and microbial activity (Eisler 2007).

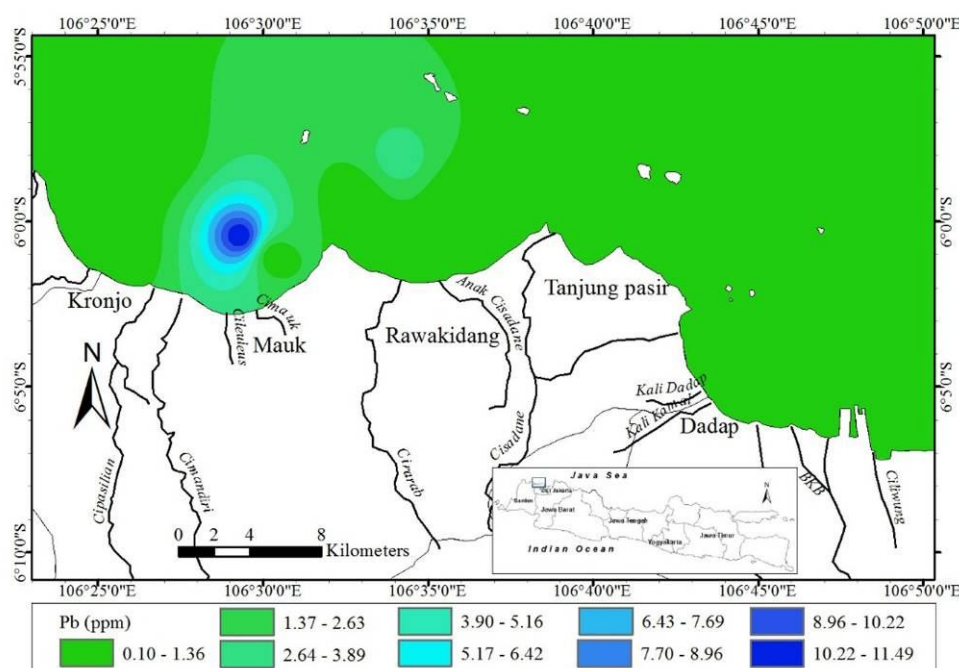


Figure 6. Distribution of lead (Pb) in the sediments of Tangerang coastal waters, Province Banten - Indonesia in April 2013.



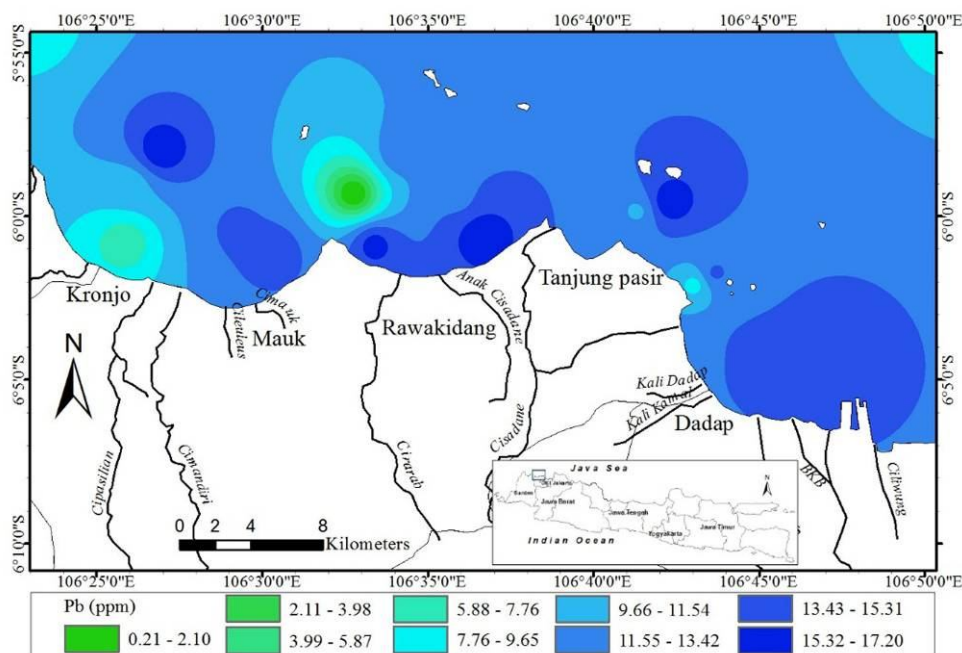


Figure 7. Distribution of lead (Pb) in the sediments of Tangerang coastal waters, Province Banten - Indonesia in August 2013.

**Current patterns and metal distribution.** The results of the spacial heavy metal distribution mapping in April and August showed significant differences in concentration. In general, the distribution of metal in locations observed was high in the eastern area. The concentration of heavy metals (Cu, Hg, and Pb) in the observation stations as a whole showed temporal differences. One case was Dadap; in this area, Cu concentration was 25.52-28.47 ppm in April and 44.67-62.37 ppm in August. The concentration of Hg was 18.22-20.19 ppm in April and 0.34-2.28 ppm in August. The concentration of Pb was 0.20-1.27 ppm in April and 11.55-17.20 ppm in August. The differences in the concentration of heavy metals (Cu, Hg, and Pb) in the two observations were probably because of the difference in the equipment used in the analysis. The AAS method has a 0.002 ppm detection level for Cu and a sensitivity of 0.025 ppm, a 0.01 detection level for Hg with a sensitivity of 0.02 ppm, and a 0.05 detection level for Pb with a sensitivity of 0.5 ppm. On the other hand, the ICP method has a Cu detection level of 0.01 ppm with 0.98 ppm sensitivity, a Hg detection level of 0.01 ppm with a sensitivity of 0.1 ppm, and a Pb detection level of 0.05 ppm with a sensitivity of 1.0 ppm (APHA 2012). This shows that the AAS is more accurate compared with ICP.

In addition, the differences might be caused by the coastal dynamics in the form of surface current strength variation which is different for each season. The ocean surface current data in April and August showed that the current in the observation area, moved towards the west and northwest at a speed of 0.005-0.185 m s<sup>-1</sup> (Figures 8 and 9). This current's direction could affect distribution, including heavy metals in sediments. The mass of seawater moving towards the west would carry in influx of chemical compounds from the rivers to the west, especially in front of the estuaries. The medium velocity currents carry sediment particles in the form of fine particles (mud) and some of the finest sand, whereas the high velocity currents carry larger amounts of sand (Barnard et al 2013). The currents which are formed from vertical concentration gradients affect bottom sediments through the processes of precipitation, vertical mixing (flotation), and currents from horizontal gradients which also affect the process of suspension (Winterwerp & van Kessel 2003).

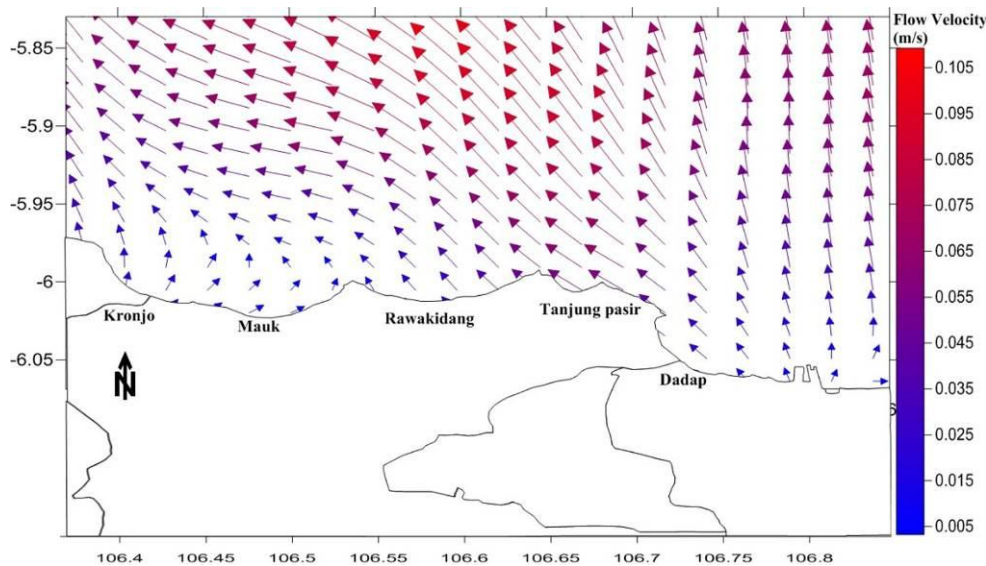


Figure 8. Currents pattern of surface in Tangerang coastal waters, Province Banten - Indonesia in April 2013 (Source: Marine and Fishery Research and Development Agency 2013).

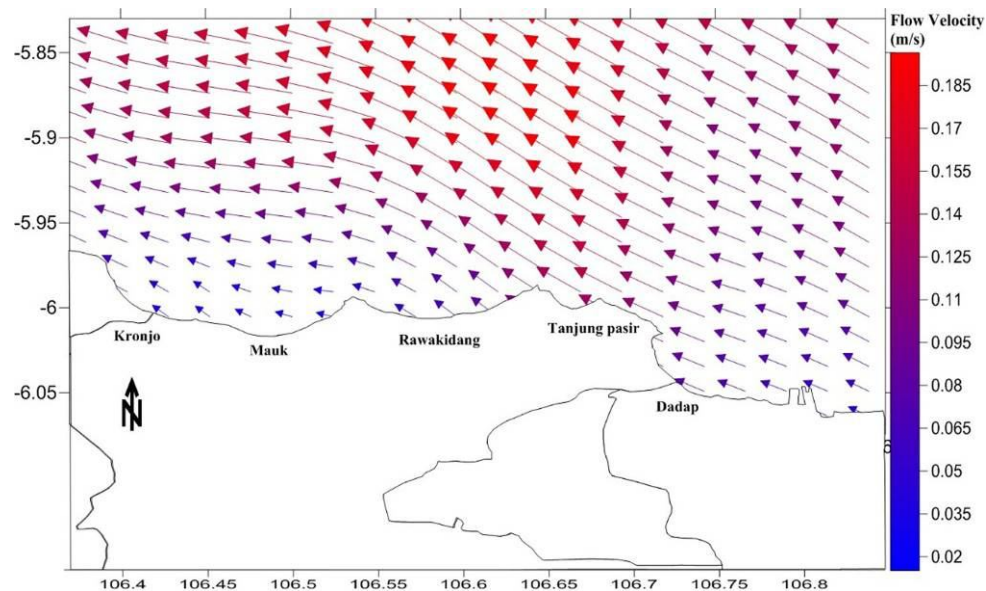


Figure 9. Currents pattern of surface in Tangerang coastal waters, Province Banten - Indonesia in August 2013 (Source: Marine and Fishery Research and Development Agency 2013).

**Sediment fraction size and organic materials.** The results of the sediment fractionizing in all observation stations showed that the fraction types are dominated by mud (Figure 10). The condition of the mud allows rapid decomposition of organic materials. The size of the sediment particles in the study locations is being strongly related to the condition of the adjacent environment during the sediment formation process. One of the sources of sediment input is abrasion or erosion products carried by the river to the coast. The factor which affects particle size is the sediment transport mechanism which in turn will determine the precipitation variations (Rahman 2006).

Coarse sediment particles easily precipitate, but it is more difficult for finer particles such as silt and clay which tend to get carried by the currents farther from the coast. The particle size classification results based on Wentworth's scale (Wentworth 1922) resulted in only two types, mud-sized particles ( $3.9 < M_d < 62.5 \mu\text{m}$ ) and very fine sand ( $62.5 < M_d < 125 \mu\text{m}$ ). Mud-sized particles dominated the sediment samples (Figure 10). The size of particles on the beach will shrink because of abrasion, resulting in finer particles farther from the beach. Mud sediment has a size close to that of clay; therefore it is postulated that the concentration of organic materials and metal sediments

would be higher. This is in line with the statement from Fortune (1993) that clay compounds have a higher proportion of organic materials and a higher affinity to metals.

Organic materials found in the sediments are spatially and temporally different. Observation results showed an average value of organic materials of 1.22 % in April and 1.68% in August (Figure 11). The concentration of organic materials on the coasts of Tangerang is high. Low concentration of organic materials found in the coastal sediments is a value of lower than 0.25% and high concentration is a value of more than 1% in the open sea sediments (Helali 2013). A concentration of organic materials which is higher than 4.6% reflects a precipitation of a combination between organic materials from dumping waste and debris from planktons (Sylaios et al 2012). Organic materials could absorb and release metals which were previously bound by a solid compartment of sediment. The mechanism found in the sediments promotes metal accumulation in the process of decomposition by decomposer microbes (Laing et al 2007).

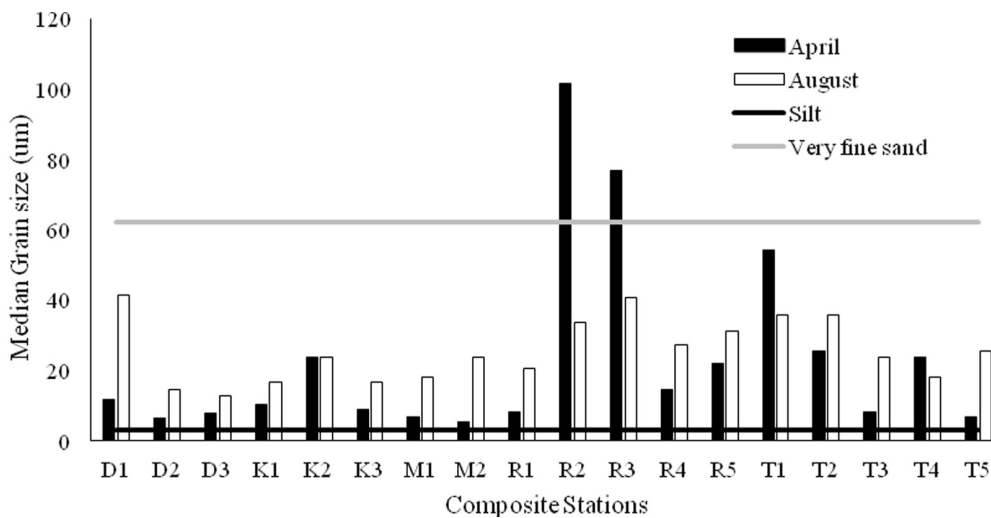


Figure 10. Classification of grain size particle of sediment in Tangerang coastal waters, Province Banten - Indonesia (median/Md) based on the Wentworth scale (D1, D2, D3, K1, .... and T5: composit stations as in Figure 1; D - Dadap, K - Kronjo, M - Mauk, R - Rawa-kidang, T - Tanjung Pasir).

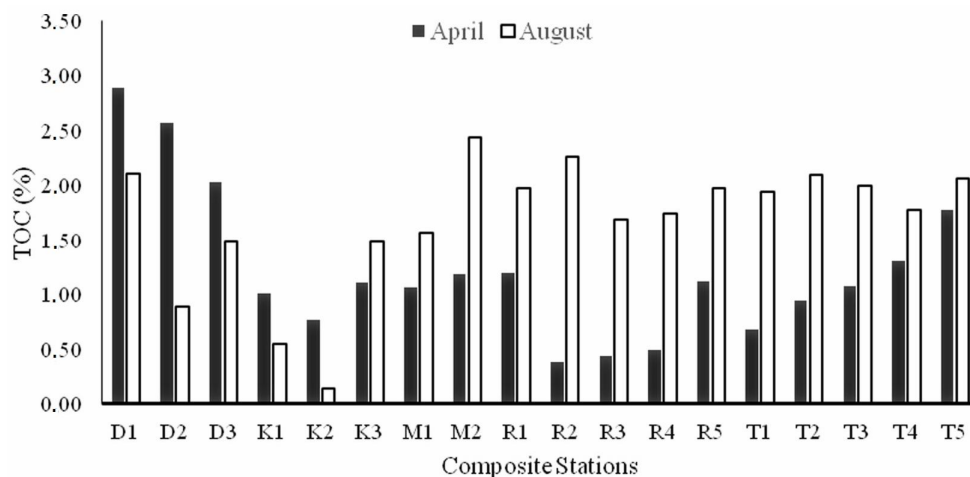


Figure 11. Total organic carbon (TOC) in sediment of Tangerang coastal waters, Province Banten – Indonesia (D1, D2, D3, K1, .... and T5: composit stations as in Figure 1; D - Dadap, K - Kronjo, M - Mauk, R - Rawa-kidang, T - Tanjung Pasir).

**Sediment components and concentration of heavy metals correlations.** Meador et al (1998) stated that the distribution and concentration of elements in sediments are influenced by several factors, such as, the texture of the sediment, the concentration of organic carbon in the sediment, the sediment's potential reduction and oxidation, and

bioturbation. The results of the Spearman Rank correlation show that there is no correlation between the size of the particles and the concentration of heavy metals; the correlation values are Cu ( $r = -0.05$ ;  $P=0.77$ ), Hg ( $r = -0.17$ ;  $P=0.33$ ), and Pb ( $r = 0.33$ ;  $p = 0.05$ ). Cd does not have a correlation with particles and organic materials because the values for Cd showed similar values for all stations. The results obtained in this study are different from previous studies. This is because the distribution of heavy metals in various particle sizes is affected by both natural and non-natural sediment formation. Togwell (1979) stated that the concentration of heavy metals in sediments is not only affected by the rock-weathering process, but also by the concentration of materials in the sediment, the mineral composition, and the (particle) size of the sediment itself. This is contradictory to Fortune's opinion (Fortune 1993) that there is no strong correlation between metals and particle size based on a study in the tropical area of Northern Australia.

The Spearman Rank correlation also showed that there was no correlation between TOC and heavy metal concentration. The value of the correlations are Hg ( $r = 0.02$ ;  $p = 0.91$ ), Pb ( $r = -0.21$ ;  $p = 0.23$ ), and Cu ( $r = -0.35$ ;  $p = 0.84$ ). The results were dissimilar to the opinions of Marcovecchio & Ferrer (2005) that Cu showed a strong correlation to organic materials in the sediment which could contribute to the ability to absorb metal into the system. Cd and Pb have no correlation to organic materials. This was similar to the finding of the studies by Marcovecchio & Ferrer (2005) and Helali et al (2013), they found that Cd and Pb show very limited correlation to organic materials in the sediments with an insignificant level of confidence. Organic materials in sediments, especially in the form of carbon, would enable the formation of bonds between organic carbon and metals. The results of the Spearman Rank correlation showed that there was no correlation between sediment texture and total organic carbon. The correlation was ( $r = (-0.19)-0.09$ ;  $p > 0.05$ ). The correlation tended to be negative, meaning that the finer the sediment texture, the more organic materials are bound in the sediment. Carbon is able to form bonds in sediment in the form of the complex bonds (complexation). The presence of metals in the sediment is affected more by concentration of TOC (Haerudin et al 2005). Therefore, the higher the concentration of organic carbon in sediments, the higher the concentration of pollutants in the sediments.

Observation results showed that there is heavy metal pollution by Hg; therefore, measures to control it need to be taken. The management strategy for heavy metal pollution in the coastal area of Tangerang is to formulate a guideline and a threshold for heavy metals in sediments, reinforce existing regional and national laws pertaining to the environment, and implement a holistic aquatic environment supervision program, especially in areas that are indicated to be polluted.

**Conclusions.** The distribution pattern of heavy metals in sediments shows that there is a varied distribution in the eastern season (April) and the transitional season (August). Several kinds of metal have high concentrations in the mid-waters. High concentrations of Cu were found in Dadap area in April, and they were high in Dadap and Kronjo in August. The concentration of Cd showed similar values ( $< 0.1$  ppm) in all areas. The concentration of Hg was higher in Kronjo and Dadap in April, but high in the Rawakidang area in August. The concentration of Pb was high in Mauk area in April, and was high in Dadap, Rawakidang, and Mauk areas in August. The concentration of Hg (0.34-20.19 ppm) as a whole has exceeded the threshold, meaning that there is Hg pollution in Tangerang's coastal area. The concentrations of Cu and Pb are still below the threshold. Spearman's correlation showed there was no correlation between heavy metals, the size of particles and total organic carbon ( $p > 0.05$ ).

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Authors:

Sigid Hariyadi, Department of Aquatic Resources Management, Faculty of Fisheries and Marine Sciences, Bogor Agricultural University, Jalan Raya Darmaga, Kampus IPB, 16680 Dramaga, West Java, Indonesia, e-mail: sigidhariyadi@gmail

Akrom Muflih, Department of Aquatic Resources Management, Faculty of Fisheries and Marine Sciences, Bogor Agricultural University, Jalan Raya Darmaga, Kampus IPB, 16680 Dramaga, West Java, Indonesia, e-mail: akrommuflih@gmail.com

Ali Mashar, Department of Aquatic Resources Management, Faculty of Fisheries and Marine Sciences, Bogor Agricultural University, Jalan Raya Darmaga, Kampus IPB, 16680 Dramaga, West Java, Indonesia, e-mail: alimashar75@gmail.com

Bambang Widigdo, Department of Aquatic Resources Management, Faculty of Fisheries and Marine Sciences, Bogor Agricultural University, Jalan Raya Darmaga, Kampus IPB, 16680 Dramaga, West Java, Indonesia, e-mail: bbg\_widigdo@yahoo.co.id

Yusli Wardiatno, Department of Aquatic Resources Management, Faculty of Fisheries and Marine Sciences, Bogor Agricultural University, Jalan Raya Darmaga, Kampus IPB, 16680 Dramaga, West Java, Indonesia, e-mail: yusli@ipb.ac.id

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